Stratospheric Controlled Perturbation Experiment (SCoPEx) Advisory Committee

Final Report

Final Report of the SCoPEx Advisory Committee

Prepared by:

Sikina Jinnah, Louise Bedsworth, Shuchi Talati, Michael B. Gerrard, Michael Kleeman, Robert Lempert, Katharine Mach, Leonard Nurse, Hosea Olayiwola Patrick, and Masahiro Sugiyama

March 2024

This report was equally prepared by the Co-Chairs of the SCoPEx Advisory Committee (Talati, Besdworth and Jinnah) with contributions and endorsement from the full Advisory Committee.

Suggested Citation:

Jinnah, Sikina, Louise Bedsworth, Shuchi Talati, Michael B. Gerrard, Michael Kleeman, Robert Lempert, Katharine Mach, Leonard Nurse, Hosea Olayiwola Patrick, and Masahiro Sugiyama. 2024. Final Report of the SCoPEx Advisory Committee. 18 March 2024. SCoPEX Advisory Committee. Available at: https://scopexac.com/finalreport

Table of Contents

Executive Summary **4**

Introduction **7**

Advisory Committee Membership 8

Mission, Values, and Operating Guidelines 8

Timeline of Committee Work 11

Research Governance Framework **13**

Financial Review **16**

Legal Review **17**

Engineering Safety Review **18**

Scientific Merit Review **20**

Societal Review **23**

Acknowledgements **25**

Appendices - Work Products

Appendix A - Financial 26

Appendix B - Legal 51

Appendix C - Engineering Safety 76

Appendix D - Scientific Merit 140

Appendix E - Societal 216

Executive Summary

Introduction

Harvard University established the Stratospheric Controlled Perturbation Experiment (SCoPEx) Advisory Committee (AC) in 2019 to provide advice to Harvard Vice Provost for Research and the SCoPEx research team on if and under what conditions SCoPEx could proceed. Harvard established the AC in anticipation that SCoPEx would be one of the first outdoor experiments of solar geoengineering or solar radiation modification (SRM).

Specifically, the AC was asked to provide recommendations on the: scientific quality and importance of the proposed experiment; risks associated with the proposed research; effectiveness of risk management; the need, objectives and possible formats for stakeholder engagement; and other issues as deemed necessary by the Advisory Committee.[1]

The AC met regularly between 2019 and 2023 as a full Committee as well as in several sub-committees focused on the various elements of its governance framework. The AC also met periodically with the SCoPEx research team to request information and obtain updates on the research team's plans, as well as with the Harvard Vice Provosts for Research and for Climate and Sustainability, to which the AC officially reported. The AC is now concluding its work with the release of this report documenting our activities over the 4 year period and presenting our final recommendations.

Governance Framework and AC Recommendations

Through a consensus-based process the Committee developed and implemented a research governance framework for SCoPEx, which the AC hopes will inform future governance of any outdoor solar geoengineering experiments. The Committee consulted with outside experts throughout its work, bringing in experts to share knowledge and experiences from similar processes.

Through this framework, the AC reviewed and provided recommendations on 5 elements of the SCoPEx proposal:

1. Engineering and Safety
2. Finances
3. Legal Issues
4. Scientific Merit
5. Societal Engagement

[1] Excerpt from SCoPEx Terms of Reference - See Appendix F

Table 1 below summarizes the four core elements of the framework and the key governance recommendations from the AC.

Table 1: SCoPEX Advisory Committee Governance Framework

Focus Area	Goal	AC Recommendations
Engineering Safety Review	Determine if the proposed experiment poses any significant or imminent safety concerns.	Current plan cleared. If SCoPEx resumes in the future, an Engineering Safety review should be conducted to account for any technology and other updates.
Scientific Merit Review	Assess the scientific merit of SCoPEx.	Sufficient to proceed. If SCoPEx resumes in the future, an updated and accessible Research Plan and an engagement plan developed in accordance with the AC's Guidelines will be needed. If changes are made to SCoPEx, the Scientific Merit Review should be conducted again.
Financial Review	Ensure all funding sources for the SCoPEx project are publicly disclosed and reviewed by the AC for possible conflicts of interest.	No conflicts of interest were identified. If SCoPEx resumes in the future, the financial review must be updated.
Legal Review	Ensure that the proposed experiment meets all regulatory requirements.	No legal obstacles or conflicts were identified for the proposed launch in Sweden, which was canceled in light of various factors. If SCoPEx resumes in future, a legal review should be conducted for any potential launch site location, since legal requirements vary from state to state.
Societal Review	Ensure that different sets of public and stakeholders have opportunities to meaningfully engage with the research team on the proposed experiment.	No engagement effort was conducted. If SCoPEx resumes in future, Harvard or the Research Team should produce and implement an engagement plan in accordance with the AC Guidelines.

Key Reflections on Societal Engagement from the Advisory Committee's Work

As one of the first bodies charged with designing and implementing a governance framework for a specific solar geoengineering experiment, the AC had the challenge and opportunity to build a model from the ground up. Our work was deeply informed by a rich body of academic literature on, and previous efforts of solar geoengineering governance, as well as various expert reports that recommend frameworks for governing solar geoengineering in general. Our work was the first to comprehensively and empirically apply these concepts and frameworks to a concrete empirical case. This process was particularly challenging in our work on developing guidelines for societal engagement. We reflect here on some elements of those discussions, which were thorny and are in need of further attention from policy makers and others who may wish to govern solar geoengineering research going forward.

Much of the AC's time was spent deliberating the specifics of how any societal engagement process for SCoPEx should work. Who should be engaged? In what format? Who should do it? Who should pay for it? Why should some groups be engaged but not others? How should researchers be expected to respond to data gathered through an engagement process? Was a global engagement necessary for a small-scale project with minimal safety risks? What is the threshold at which any experiment should include an engagement process? The AC unanimously agreed that engagement was a critical element of the review for SCoPEx. However, the AC faced the challenge of designing an appropriate engagement process around a small-scale experiment, with the knowledge that its execution had important implications at the societal level (i.e., moral hazard).

Although the Committee did not reach consensus on the conditions under which engagement is recommended for solar geoengineering research broadly speaking, the AC did agree that our governance framework should be a model for governance of future solar geoengineering experiments. All elements of the framework may not be appropriate for all experiments or programs, and implementation of specific elements will need to be determined on a case-by-case basis. Making this determination of propriety is a critical issue that should be addressed by policy makers as soon as possible to enable efficient, effective and transparent governance expectations for researchers seeking to engage in outdoor experimentation in future.

The AC also agreed on four core principles for societal engagement, when conducted, which reflect insights from the academic literature but we deem as sufficiently important to future solar geoengineering research efforts as to amplify them here:

1. Start engagement efforts as early as possible
2. Include social scientists with engagement expertise on research teams during the research design process
3. Don't presuppose what communities will be concerned about
4. Develop a plan to be responsive to community concern

Introduction

Harvard University established the Stratospheric Controlled Perturbation Experiment (SCoPEx) Advisory Committee in 2019 to provide advice to Harvard Vice Provost for Research and the SCoPEx research team on if and under what conditions SCoPEx could proceed.

A team of researchers at Harvard University proposed the SCoPEx project, which would be one of the first outdoor solar geoengineering experiments. The experiment was designed to advance understanding of stratospheric aerosols that could be relevant to solar geoengineering. It was designed to measure aspects of the aerosol microphysics and atmospheric chemistry that are currently highly uncertain in the simulations. It was not designed to be a test of solar geoengineering per se. Rather, it was designed to improve understanding of near-field plume evolution and with solar and infrared radiation (SCoPEx Research site). The project received internal University funding through Harvard's Solar Geoengineering Research Program (SGRP).

Because of the important implications associated with conducting an outdoor solar geoengineering research experiment, the research team and Harvard leadership committed to establishing a governance process to advise on the work. Harvard established a search committee in 2018 to establish the Terms of Reference for an Advisory Committee and identify candidates to serve as Advisory Committee Chair. The Search Committee determined that the Advisory Committee should provide recommendations on the:

- Scientific quality and importance of the proposed experiments, including scientific review and processes and standards for transparency;
- Risks associated with the proposed research program, including environmental, social and reputational risks;
- Effectiveness of risk management including regulatory compliance management of environmental health and safety;
- The need, objectives and possible formats for stakeholder engagement; and
- Other issues as deemed necessary by the Advisory Committee.[1]

When established, the Advisory Committee reported to Harvard's Vice Provost for Research. The Committee jointly reported to the Vice Provost for Research and the Vice Provost for Climate and Sustainability, once the latter position was established in 2021.

The Advisory Committee met regularly between 2019 and 2023. Through a consensus-based process the Committee developed and implemented a research governance framework for SCoPEx, which we hope will inform governance of any future outdoor solar geoengineering experiments. The Committee consulted with outside experts throughout its work, bringing in experts to share knowledge and experiences from similar processes. The Committee created and engaged an expert panel to guide the selection of peer reviewers to conduct the scientific merit review.

[1] Excerpt from SCoPEx Terms of Reference - See Appendix F

Advisory Committee Membership

Following the appointment of the Committee Chair, additional Advisory Committee members were appointed by Harvard with the guidance of the Committee Chair and other experts. Committee members were chosen to reflect a range of disciplines, geographies, gender, race, and age. In addition, Harvard prioritized selection of Committee members with no clear external bias regarding solar geoengineering. Over the course of the Committee's tenure, additional Committee members were appointed to bring additional topical expertise and geographic representation to the Committee.

Advisory Committee Members (Terms of Service)

Louise Bedsworth (April 2019 - March 2024)[2]

Michael B. Gerrard (July 2019 - March 2024)

Sikina Jinnah (September 2021 - March 2024)[3]

Michael Kleeman (July 2019 - March 2024)

Kevin Knobloch (July 2019 - April 2021)

Robert Lempert (July 2019 - March 2024)

Katharine Mach (July 2019 - March 2024)

Leonard Nurse (July 2019 - March 2024)

Hosea Olayiwola Patrick (November 2021 - March 2024)

Rajul (Raj) Pandya (July 2019 - October 2022)

Masahiro (Masa) Sugiyama (September 2021 - March 2024)

Shuchi Talati (July 2019 - Jan 2021, April 2022 - March 2024)[4]

Committee Staff

Sally Klimp (Nov 2019 - March 2024)

Mission, Values, and Operating Guidelines

The Advisory Committee developed and adopted the following mission and values to guide its work.

Mission Statement:

The purpose of the Advisory Committee is to provide advice on the research and governance of SCoPEx, operating independently from the Research Team. The Committee's goal is to ensure that the SCoPEx project is undertaken in a transparent, responsible, and legitimate manner by ensuring that it contributes to scientific understanding and establishes means for meaningful public engagement in the experiment.

[2] Committee Chair, April 2019- July 2020, Co-Chair Leadership Committee April 2022- January 2024

[3] Co-Chair Leadership Committee April 2022- January 2024

[4] Co-Chair Leadership Committee April 2022- January 2024

SCoPEx Advisory Committee Values

Integrity and Impartiality

In order to have impact, the work of the Committee must be respected and credible. Each member on the Committee was chosen for their experience as well as their reputation for integrity in international environmental research and governance. The Committee membership is intended to represent a wide range of perspectives, experiences, and expertise that are relevant to governing the experiment. Any circumstance that may present bias in the Committee process must be clearly identified and satisfactorily resolved to avoid inappropriate influence in the review process.

Expert and Evidence-Based Assessment

The Committee will invite and consider diverse scientific, cultural, philosophical, and ethical input while conducting their work in evaluating the governance and scientific review of SCoPEx. It will make decisions and recommendations based on this input, using its own expertise, while maintaining fidelity to the evidence and striving to be impartial.

Transparency

The Committee membership, terms of reference, operating guidelines, important updates, and relevant materials will be posted and shared on the Advisory Committee's website in a timely manner.

Advancement of Science

The Committee will consider and evaluate the potential efficacy of solar geoengineering research based on the current state of knowledge. The Committee is committed to advancing knowledge through its work and will assess and, to the extent feasible, identify strategies and options to mitigate any risks associated with SCoPEx.

Engagement, Collaboration, and Social Responsibility

Given the broad societal implications of solar geoengineering research and its potential contribution to the eventual deployment of solar geoengineering at scale, the public should be involved in decisions involving such research. Accordingly, the Committee is committed to embedding principles of engagement, collaboration, and social responsibility into its own work as well in our recommendations to the Research Team and Harvard University.

The Committee will seek engagement from a diverse range of stakeholders, inviting and welcoming diverse perspectives into the conversation. We will make concerted efforts to consult, especially with those who have experienced historical barriers to participation, including indigenous and local leaders, environmental justice communities, scientific experts, informal and formal community leaders, legal experts, moral and ethical teachers, and environmental leaders, prior to any release of materials in the atmosphere taking place. We recognize that some people or communities may have larger barriers to overcome in an engagement process and we are committed to finding ways around those barriers.

The Committee operates to increase all interested communities' understanding of solar geoengineering, to understand the perspectives of different communities and stakeholders, to gain and attend to input from all interested persons.

The Committee will also work with a view to ensuring that the Research Team establishes a similar goal and process for engagement and collaboration.

Purpose of this Report

This report documents the Advisory Committee's governance framework and the activities over the course of its lifetime. This report presents the Advisory Committee's framework and describes the activities undertaken to support implementation of the framework. Appendices contain supporting materials for each element of the framework. The following pages contain a timeline of Advisory Committee activities and milestones.

It is the hope of the Committee that in addition to sharing information on the Committee's work that this report can inform future research governance activities.

SCoPEx Advisory Committee - Timeline of Activities

- **May 2018 -** Harvard establishes search committee for Chair of SCoPEx Advisory Committee (Peter Frumhoff, Jane Long, Chris Field)
- **May-June 2018 -** Search Committee establishes Terms of Reference for the Advisory Committee
- **April 2019 -** Louise Bedsworth appointed as Chair by Search Committee and Harvard
- **July 2019 -** Harvard forms initial Advisory Committee based on Chair and Search Committee recommendations
- **October 2019 -** First Committee meeting
- **November 2019 -** Harvard hires Executive Coordinator (Sally Klimp) to support Advisory Committee
- **December 2019 -** AGU workshop
- **January 2020 -** AGU workshop
- **June 2020 -** Research Team Responds to Advisory Committee Financial Disclosure Questions
- **July 2020 -** Bedsworth steps down as Advisory Committee Chair
- **August 2020 -** Advisory Committee publishes Proposed Societal Engagement Process for SCoPEx for public comment
- **October 2020 -** Research Team responds to first draft of Societal Engagement Guidelines
- **November 2020 -** November 10: Research Team sends letter to Advisory Committee regarding potential platform test in Sweden, requesting decision by February 2021
- **November 2020 -** November 10: Research Team sends initial research plan to Advisory Committee
- **November 2020 -** November 30: Advisory Committee responds to Research Team requesting additional information regarding potential Sweden launch
- **November 2020 -** Advisory Committee publishes Financial Review
- **December 2020 -** Research Team responds to Advisory Committee request for information regarding platform test in Sweden
- **December 2020 -** Advisory Committee recruits reviewers for Engineering Safety Review
- **January 2021 -** Committee publishes final Proposed Societal Engagement Process
- **January 2021 -** Advisory Committee publishes Engineering Integrity and Safety Review
- **February 2021 -** Advisory Committee publishes Initial Legal Review
- **February 24, 2021 -** February 24: Advisory Committee receives letter from Saami Council stating opposition to SCoPEx platform test in Sweden

General Advisory Committee/SCoPEx Advisory Committee activities | Finance | Legal | Engagement/Societal | Engineering Safety Review | Scientific Merit Review

March 2, 2021 - March 2: Advisory Committee issues letter in response to letter from Saami Counci

March 31, 2021 - March 31: Advisory Committee releases statement calling for delay for Swedish flight pending engagement process

Swedish Space Corporation and Harvard announce decision not to conduct platform test in Sweden

March 2021 - March 25: Advisory Committee receives letter from Board of the Swedish Royal Advisory Academy of Sciences calling for the Advisory Committee to cancel SCoPEx

April 2021 - Committee issues solicitation for new Advisory Committee members

June 2021 - Indigenous Peoples Call on Harvard to shut down SCoPEx

October 2021 - CEC Workshop and Listening Session

December 2021 - AGU Town Hall

January 2022 - Advisory Committee establishes Panel of Experts to aid in Scientific Merit Review

March 2022 - Research Team releases updated Research Plan

April 2022 - Advisory Committee establishes Leadership Committee (Louise Bedsworth, Sikina Jinnah, Shuchi Talati)

April 2022 - Advisory Committee meets in-person

April 2022 - Advisory Committee creates sub-committee for scientific merit review (Masa Sugiyama (chair), Leonard Nurse, Sikina Jinnah) and secures five reviewers w/ advice of panel

May-July 2022 - Reviewers conduct and send first round reviewsReview

July 2022 - Panel submits summary report to Advisory Committee

August 2022 - Panel declines to advise on second round of review

October 2022 - RT responds to Peer Reviews and Panel Report

October 2022 - Advisory Committee publishes Local Engagement Guidelines

January 2023 - Advisory Committee receives second round of reviews

February 2023 - Advisory Committee creates summary report for second round of reviews

June 2023 - Research Team responds to Second Round review and Advisory Committee's Summary Report

August 2023 - Research team communicated planned suspension of SCoPEx with Advisory Committee

September 2023 - Advisory Committee shares dissolution plans with Harvard and RT

General Advisory Committee/SCoPEx Advisory Committee activities | Finance | Legal | Engagement/Societal | Engineering Safety Review | Scientific Merit Review

Research Governance Framework

The Advisory Committee followed a consensus-based process to develop a framework for making recommendation(s) for the SCoPEx project. The framework contains five elements that would serve as the primary inputs for the Committee to make recommendations to the Vice Provost for Research at Harvard University and the SCoPEx Research Team.

1. Engineering and Safety Review
2. Financial Review
3. Legal Review
4. Scientific Merit Review
5. Societal Review

Table 1 below summarizes each of the five elements, the process, and outcomes. This is followed by a summary and discussion of each element of the framework. The Appendices contained supporting documentation and work products.

Focus Area	Goal	Process	Deliverables and Outcome	Advisory Committee Recommen-dations
Engineering Safety Review	Determine if the proposed experiment poses any significant or imminent safety concerns.	The AC recruited three experts with expertise in flight dynamics to review the SCoPEx research plan (version 1.0).	The Engineering Safety Review was completed and published in January, 2021. Based on the external engineering and safety review, the AC determined that the proposed experiment poses no significant or imminent safety concerns.	If SCoPEx resumes in the future, an Engineering Safety review should be conducted to account for any technology updates.

Focus Area	Goal	Process	Deliverables and Outcome	Advisory Committee Recommen-dations
Scientific Merit Review	Assess the scientific merit of SCoPEx including the feasibility of the experimental approach and contribution to knowledge and under-standing of stratospheric particle dynamics.	The Scientific Merit Review included two rounds of review. In the first round, the AC recruited a Panel of 3 scientific experts who identified five scientists with relevant expertise to serve as Reviewers of the SCoPEx research plan (version 2.0). The Panel summarized the individual reviews and produced a Report for the AC. At the AC's request, the Research Team prepared responses to the Panel Report. The AC sent these responses to the same 5 scientists to complete a second round of review. Based on these reviews, the AC developed a set of recommendations for the Research Team.	Based on the second round of review, the AC recommended that the Research Team revise the scientific proposal in response to the reviews and make the revised document publicly available. With that recommen-dation, the AC concluded that the scientific merit of the proposed experiment is sufficient to proceed with planning a community engagement process.	The AC recommended that the Research Team produce an updated and accessible Research Plan and develop an engagement plan in accordance with the AC's Guidelines. If substantive changes are made to SCoPEx in any future iteration of the experiment the Scientific Merit Review should be conducted again.
Financial Review	Ensure all funding sources for the SCoPEx project are publicly disclosed and reviewed by the AC for possible conflicts of interest.	Reviewed project funding at regular intervals and had conversations with several funders to understand their relationship with the project. The financial review should be updated if SCoPEx resumes in future.	The Financial Review materials were published in November, 2020. The AC confirmed that all funding sources as of January 2021 are publicly disclosed and did not identify any COIs.	The AC recommended that the financial review be regularly updated throughout the course of the project. The Financial Review should be updated if SCoPEx resumes in the future.

Focus Area	Goal	Process	Deliverables and Outcome	Advisory Committee Recommen-dations
Legal Review	Ensure that the proposed experiment meets all regulatory requirements including permits and approvals in compliance with any potential location of a launch.	Conducted legal review for potential launch in Sweden (Setterwalls, 2021).	Review for Sweden completed in February, 2021. The AC determined that the proposed flight in Sweden was in compliance with all local and national laws.	Legal review should be conducted for any potential launch site location. Further review would be needed if SCoPEx resumes in future, and would be contingent on the location of any planned launch.
Societal Review	Ensure that different sets of public and stakeholders (e.g. potential local community where the experiment could take place as well as the larger global community) have opportunities to meaningfully engage with the research team on the proposed experiment.	AC developed preliminary recommendations for a global engagement process. The AC has also consulted the public directly through an online comment portal on our website and consulted with experts at various conferences and via Zoom. The AC developed guidelines for a local engagement process for the research team.	The proposed Societal Engagement Plan was completed in January, 2021. Local engagement guidelines were completed in October 2022.	If SCoPEx resumes in future, the Research Team should produce an engagement plan in accordance with the AC Guidelines and lead an engagement process upon its approval.

Financial Review

The Advisory Committee requested two documents from the Research Team: a financial statement that lists all monetary and in-kind contributions and a conflict of interest statement for major funding sources. The Advisory Committee then conducted a financial review of SCoPEx to ensure transparency and public disclosure of all funding information. The Advisory Committee identified monetary and in-kind contributions to support the research. The Committee asked the Research Team to answer the following questions regarding funding sources:

- Are all funding sources, including level of support, clearly identified and publicly listed?
- Does the project have any anonymous individual supporters? If so, why?
- Have institutional donors identified all sources of funds, including individual donors, corporations, etc.?
- Are any project funders associated with an institution that stands to benefit, financially or politically, from the results of this work? If yes, explain.
- Was any of the funding directed to specific activities?
- Does the project intend to create any new or novel technology and how will it be shared with the public? Is there an intention to create intellectual property?
- What are the relevant policies around funding?

The Advisory Committee issued its initial request for this information in January 2020. Responses from the Research Team are included in Appendix A. The Research Team requested that information on funding amounts from individual donors be kept private.

The Research Team provided updates on new donors over the course of the project. The Advisory Committee planned to regularly update and publicly post a financial review.

Financial Review Timeline and Milestones:

January 2020 - AGU workshop

June 2020 - Research Team Responds to Advisory Committee Financial Disclosure Questions

November 2020 - Advisory Committee publishes Financial Review

Appendix A Contents:

- Appendix A-1: Advisory Committee Request for Financial Review
- Appendix A-2: Research Team Response to Advisory Committee Request Appendix A-3: Advisory Committee Request for Additional Information
- Appendix A-4: Research Team Response to Request for Additional Information
- Appendix A-5: Additional Updates from the Research Team

Legal Review

The Advisory Committee legal review entailed assessing applicable local, state, federal and international regulatory requirements for the experiment and evaluating that the experiment is consistent with all applicable regulations and requirements. A location is necessary for such a review to be completed.

Harvard retained the Swedish Law Firm Setterwalls to conduct the independent legal review of the proposed equipment test flight in Sweden (available in Appendix B). Setterwalls concluded that the Swedish Space Act is not applicable on the First Phase of the Project and the Swedish Environmental Code does not apply to the First Phase. The review also concluded that no international law barred the proposed test flight, and that there is no requirement that Harvard obtains a license for the balloon and gondola according to the Swedish Civil Aviation Act and consequential legislation.

As the Research Team did not decide on a new location, a subsequent legal review was not conducted.

Financial Review Timeline and Milestones:

- **February 2021 -** Advisory Committee publishes Initial Legal Review for potential launch in Sweden

Appendix B Contents:

- Appendix B: Scope of Work and Legal Review Memo from Setterwells to Harvard University (February 18, 2021)

Engineering Safety Review

The Engineering and Safety Review assessed the engineering integrity and safety of the first proposed flight testing the equipment for SCoPEx. This review was conducted in the context of the potential platform testing in Kiruna, Sweden that would not release any aerosols. This platform test included testing the gondola's horizontal and vertical control using a winch system and propellers, as well as the power, data, navigation, and communication systems. They would not release any aerosols, nor fly an aerosol injection/release system.

The Advisory Committee recruited three scientists with expertise in balloon flight dynamics to review SCoPEx's experiment plan. Based on the feedback from reviewers and responses from the research team, the Committee found no significant or imminent safety concerns. The Committee agreed the research team has successfully met the requirements of this review.

Timeline for Engineering Safety Review Milestones:

- **November 2020 -** November 10: Research Team sends letter to Advisory Committee regarding potential platform test in Sweden, requesting decision by February 2021
- **November 2020 -** November 10: Research Team sends initial research plan to Advisory Committee
- **November 2020 -** November 30: Advisory Committee responds to Research Team requesting additional information regarding potential Sweden launch
- **December 2020 -** Research Team responds to Advisory Committee request for information regarding platform test in Sweden
- **December 2020 -** Advisory Committee recruits reviewers for Engineering Safety Review
- **January 2021 -** Advisory Committee publishes Engineering Integrity and Safety Review

Review Questions

We specifically asked reviewers to evaluate the platform test only, not the overall research plan and design. Documents shared with reviewers are available in Appendix C. Reviewer feedback and responses from the Research Team are available in Appendix C. Reviewers were asked to focus on the following questions:

1. Are there technical and/or other issues that have not been identified (or identified properly) that could compromise the platform test or cause it to not yield the desired data?

2. Are there unidentified risks or risks that have not been appropriately evaluated in the plan or correspondence from the research team?

3. Are there material improvements or prior research which have not, in your experience, been included in the plan for the test which might improve the project?

Reviewers of the SCoPEx Engineering Safety of SCoPEx

1. Henry Cathey, Aerospace Division Director and UAS Flight Test Site Director at the Physical Science Laboratory, New Mexico State University

2. Rodger Farley, Founder and CTO of Farley Flight Aerospace LLC

3. Christer Fuglesang, Professor Astronautics at KTH Royal Institute of Technology, Stockholm, Sweden Director KTH Space Center

Appendix C Contents:

- Appendix C-1: Letter from Research Team Requesting Authorization for Platform Test
- Appendix C-2: Research Plan Provided by Research Team
- Appendix C-3: Advisory Committee Response to Research Team Request
- Appendix C-4: Research Team Response to Questions from Advisory Committee
- Appendix C-5: Reviewer Feedback and Responses

Scientific Merit Review

The SCoPEx Advisory Committee developed a rigorous multi-step process to review the scientific merit of SCoPEx, and its potential contributions to knowledge and enhancing understanding of stratospheric particle dynamics. The Advisory Committee appointed a 3-member Panel of Experts with specialized expertise in stratospheric science and climate modeling to assist with the peer review process by helping to select reviewers and synthesizing and summarizing the reviews for the Advisory Committee.

With support from the Panel of Experts, the Advisory Committee selected 5 peer reviewers to evaluate the SCoPEx Research Plan and provide feedback on the scientific merit of the proposed experiment. Reviewers were selected for their knowledge of at least one of the following topics: global modeling, instrumental design, in situ campaigns, cloud/aerosol microphysics, and/or atmospheric chemistry. The Advisory Committee screened all potential reviewers for conflicts of interest (COI), using the US National Science Foundation guidelines for COI to eliminate reviewers.[5]

The SCoPEx scientific merit review process proceeded as follows:

Stage 1 (April 2022): The Advisory Committee established a Panel of three independent scholars with expertise in stratospheric science to help select reviewers, and to evaluate and summarize the reviews in a brief report for the Committee. The Panel assisted the Committee in identifying five independent experts to conduct a single blind review of the scientific proposal (dated 29 April 2022) The reviewers were selected on the basis of their expertise in the areas of climate modeling, instrument design, cloud and aerosol microphysics and atmospheric chemistry, among others.The Committee invited the Research Team to respond to the Panel's summary report of the reviews as well as the individual reviews themselves, and to revise the scientific proposal.

Stage 2 (October 2022): The Research Team prepared a detailed response to the Panel Summary Report and the first round of reviews. All five of the original reviewers evaluated the Research Team's response and provided additional comments. The Committee invited the Research Team to respond to the second round of comments from the reviewers.

Stage 3 (February 2023): The Committee created a Summary Report of the second round of reviews (in place of a panel report) for publication including a background of the Committee's work to date and recommendations on next steps

Stage 4 (Spring-Summer 2023): The Research Team submitted a detailed response to the second round of reviewer comments. The Committee reviewed the Research Team's response and recommended further revisions to the research proposal.

[5] See https://new.nsf.gov/policies/pappg/23-1/ch-2-exhibit-2#:~:text=Unless%20a%20waiver%20has%20been%20granted%20by%20NSF%2C%20a%20potential,student%2Fadvisor.

Specifically, the Advisory Committee invited the Research Team to respond, following the outstanding items raised in the reviews:

1. Rationale needed for using calcite, as opposed to sulfate aerosols, arguably a leading material for solar geoengineering, in the experiment;
2. Clarification needed on methodology proposed for evaluating the influence of turbulence on particle coagulation;
3. The need to demonstrate that the Research Team could maneuver the balloon (gondola) as detailed in the scientific plan;
4. A description of funding and resources required for accomplishing the experiment's scientific goals;
5. An explanation and justification for the specifications of the injector as the initial condition for aerosol evolution;
6. A detailed possible timeline for the proposed SCoPEx test flights and decision points.

Stage 5 (Spring-Summer 2023): The Committee reviewed all materials, including all 5 reviews from both the first and second rounds, the Panel's summary report, and the Research Team's responses to the reviewers. On the basis of these data, the Advisory Committee concluded that the scientific merit of the proposed experiment is sufficient to proceed with planning a community engagement process. The Committee further recommended that the Research Team revise the scientific proposal in response to the reviews and make that document publicly available.

Prior to beginning any societal engagement, the Advisory Committee requested the following from the Research Team:

1. Respond to any outstanding specific questions raised by the reviewers;
2. Revise a publicly available experiment plan to reflect all changes made through the review process;
3. Ensure the revised experiment plan clearly identifies milestones, decision points, and potential off ramps if equipment does not perform as expected or other experiment performance issues arise.

The Advisory Committee did not receive this information from the Research Team prior to ending its work in January 2024.

Scientific Merit Review Timeline and Milestones:

- **January 2022 -** Advisory Committee establishes Panel of Experts to aid in Scientific Merit Review
- **March 2022 -** Research Team releases updated Research Plan
- **April 2022 -** Advisory Committee creates sub-committee for scientific merit review (Masa Sugiyama (chair), Leonard Nurse, Sikina Jinnah) and secures five reviewers w/ advice of panel
- **May-July 2022 -** Reviewers conduct and send first round reviewsReview
- **July 2022 -** Panel submits summary report to Advisory Committee
- **August 2022 -** Panel declines to advise on second round of review
- **October 2022 -** RT responds to Peer Reviews and Panel Report
- **January 2023 -** Advisory Committee receives second round of reviews
- **February 2023 -** Advisory Committee creates summary report for second round of reviews
- **June 2023 -** Research Team responds to Second Round review and Advisory Committee's Summary Report

Appendix D Contents:

- Appendix D-1: Revised Research Plan
- Appendix D-2: Terms of Reference for Panel of Experts
- Appendix D-3: Round 1 Panel Summary Report Appendix D-4: Research Team Response to Round 1 Panel Report
- Appendix D-5: Advisory Committee Summary of Round 2 Reviews
- Appendix D-6: Research Team Response to Second Round Reviews

Societal Review

The Advisory Committee developed a process to identify and review societal concerns and interests surrounding SCoPEx. The Societal Review process targeted both local and global publics and stakeholders to build an inclusive and collaborative engagement process. The societal review was a novel and essential piece of our work and one that we hope will serve as a model to guide societal engagements to help inform future governance (see the Note on Engagement section below). The Committee produced recommendations for a robust engagement plan, which would provide the opportunity for mutual dialogue and create a pathway to connect the feedback from stakeholders to Harvard University and the Research Team. Our recommendations were designed to be responsive to different perspectives, social values, and needs. The documents were made publicly available for stakeholders and other interested parties.

The local and global tracks for engagement resulted in guidance for consultation with residents and stakeholders in the immediate vicinity of the planned balloon launch as well as a broader set of global engagement activities throughout the committee process.

The Committee first developed an outline for the Societal Engagement Plan for SCoPEx specific to the particle release portion of the experiment based on the work of a subcommittee which focused on engagement which researched literature in the field and interviewed academics who had done comparable engagements. We then updated the Plan based on feedback from the public, relevant scholarly experts, and the Research Team. The final Societal Engagement Plan was made available on our website in Jan 2021 and can be found in Appendix E.

Building on the Societal Engagement Plan, the Committee subsequently developed more fine grained Local Engagement Guidelines. These guidelines emphasized the need for an independent engagement facilitator and local partner(s), an understanding of local knowledge, conditions and concerns, and the use of deliberative methods (a structured, two-way process, where participants consider evidence and diverse perspectives, 'deliberate' options, ask questions, and provide feedback on the proposed experiment and associated activities that can inform next steps and future work). The Guidelines have been available on our website since October 2022 and are included in Appendix E.

The Committee also established a subcommittee for global engagement in April 2022, which produced a series of recommendations, including an literature-based analysis of existing research on global public attitudes towards solar geoengineering research. The subcommittee further recommended that this review should focus on publics in the Global South, as well as other publics who might be underrepresented in the literature (e.g. indigenous populations in the Global North). The subcommittee recommended that this review should identify publics whose perceptions aren't well known in making any further recommendations on a global engagement process.

Due to the suspension of SCoPEx in Fall 2023, the research team did not conduct a local engagement process, nor did they review the existing research on global perceptions. However, at time of writing, this review is being conducted by scholars outside the SCoPEx process.

In parallel to developing guidelines for the Research Team on societal engagement and review, the Committee also performed some of its own engagement activities (listed below) to inform its work. These included town halls and workshops, engaging publicly with external stakeholders, and soliciting public comment on documents.

The AC worked closely with the research team to share guidelines for engagement. The research team and Harvard leadership both supported this work as a way to develop a model for future governance of such experiments.

Societal Review Timeline and Milestones:

- **August 2020 -** Advisory Committee publishes Proposed Societal Engagement Process for SCoPEx for public comment
- **October 2020 -** Research Team responds to first draft of Societal Engagement Guidelines
- **January 2021 -** Committee publishes final Proposed Societal Engagement Process
- **February 24, 2021 -** February 24: Advisory Committee receives letter from Saami Council stating opposition to SCoPEx platform test in Sweden
- **March 2, 2021 -** March 2: Advisory Committee issues letter in response to letter from Saami Counci
- **March 2021 -** March 25: Advisory Committee receives letter from Board of the Swedish Royal Advisory Academy of Sciences calling for the Advisory Committee to cancel SCoPEx
- **October 2021 -** CEC Workshop and Listening Session
- **December 2021 -** AGU Town Hall
- **October 2022 -** Advisory Committee publishes Local Engagement Guidelines

Appendix E Contents:

- Appendix E-1: Proposed Societal Engagement Plan from Advisory Committee
- Appendix E-2: Local Engagement Guidelines Appendix
- E-3: Letter to SCoPEx Advisory Committee from Saami Council
- Appendix E-4: Letter from SCoPEx Advisory Committee to Saami Council

Acknowledgements

The SCoPEx Advisory Committee is deeply grateful for the time and counsel of various experts who supported our work over the years. Centrally, we want to recognize the contributions of our Executive Coordinator, Sally Klimp. Her skill, expertise and professionalism were essential in managing this complex project with diverse interests, disciplines, and geographies. We are also grateful to the search committee Jane Long, Peter Frumoff and Chris Field for their support in establishing the committee, to the Harvard research team, Frank Keutch and David Keith, for briefing us on their work, and Harvard leadership, John Shaw, James Stock, and Rich McCullough, for providing us the leeway to operate independently to produce a document that we hope will guide solar geoengineering research governance for years to come. Finally, we appreciate the many experts who guided our thinking through engaging presentations and discussion, including: Chad Baum, Lisa Dilling, Marion Hourdequin,Katharine Mansell, Naoyuki Mikami, Kevin Noone,Tim Nuthall, Karen Parkhill, Edward Parson, and Alice Siu.

Appendix A

Financial Review Documents

Appendix A-1

Advisory Committee Request for Financial Review

Financial Disclosure Review Process

Products Submitted For Review

The Research Team will submit two documents to the Advisory Committee.

Financial Statement

The Research Team/Solar Geoengineering Research Program will provide the Advisory Council with a Financial Statement that will include all sources of funding, both monetary and in-kind, for the proposed project. The Financial Statement must include the following elements:

- **Funding Sources**: Identify all sources of monetary and in-kind support for the proposed experiment.

- **Disclosures**: Provide answers to the following questions:
    - Are all funding sources, including level of support, clearly identified and publicly listed?
    - Does the project have any anonymous individual supporters? If so, why?
    - Have institutional donors identified all sources of funds, including individual donors, corporations, etc.?
    - Are any project funders associated with an institution that stands to benefit, financially or politically, from the results of this work? If yes, explain.
    - Was any of the funding directed to specific activities?
    - Does the project intend to create any new or novel technology and how will it be shared with the public? Is there an intention to create intellectual property?
    - What are the relevant policies around funding?

The Financial Statement will be publicly available through the Advisory Committee's website.

Conflict of Interest Statement

The Research Team will provide a conflict of interest statement for experiment and major funding sources.

Appendix A-2

Research Team Response to Advisory Committee Request

June 10, 2020

Dr. Louise Bedsworth
Chair of the SCoPEx Advisory Committee &
Executive Director of California Strategic Growth Council
1400 Tenth Street
Sacramento, CA 95814

Dear Louise,

In response to the SCoPEx Advisory Committee's request regarding the Research Team's and Harvard's Solar Geoengineering Research Program's (SGRP) financials and conflict of interest principles, I have enclosed three documents:

- The Statement
- Appendix A
- Appendix B

The Statement and Appendix A can be made public at your discretion. Appendix B can be shared privately with the SCoPEx Advisory Committee with the agreement that the information not be released publicly.

Please don't hesitate to reach out with questions. I am happy to setup a call to discuss.

Yours,

David Keith

David Keith
Gordon McKay Professor of Applied Physics, School of Engineering and Applied Sciences (SEAS); and,
Professor of Public Policy, Kennedy School of Government,
Harvard University

To: SCoPEx Advisory Committee
From: David Keith
Date: June 10, 2020
Subject: Statement Regarding Financials and Conflict of Interest

---

In response to your request regarding the Research Team's and Harvard's Solar Geoengineering Research Program's (SGRP) financials and conflict of interest principles, I have enclosed a statement detailing the information you requested. This statement can be made public at your discretion.

Separately, I have shared Appendix A and Appendix B. You are welcome to make Appendix A public. You can review Appendix B privately with the agreement that this information not be released publicly. Such a structure is meant to ensure that the SCoPEx Advisory Committee has the ability to review certain details while at the same time protecting donor privacy. We hope this serves as a useful template for other institutions if they carry out small scale outdoor solar geoengineering research.

Financial Disclosure

Question: Identify all sources of monetary and in-kind support for the proposed experiment.

Response: Experimental hardware and operations are funded from internal Harvard research funds provided to Professors David Keith and Frank Keutsch. Additional research funding is provided by Harvard's Solar Geoengineering Research Program (SGRP).

SGRP is funded by the following foundations and individuals. All donations are philanthropic gifts.

J. Baker Foundation
The Blue Marble Fund
OW Caspersen Foundation
The Crows Nest Foundation
The William and Flora Hewlett Foundation
Constance C. and Linwood A. Lacy Jr. Foundation
The Open Philanthropy Project
Pritzker Innovation Fund
Ronin Private Investments LLC
The Alfred P. Sloan Foundation
The Tansy Foundation
Teza Technologies LLC
VoLo Foundation
The Weatherhead Center for International Affairs

Laura and John Arnold
G. Leonard Baker, Jr.
Alan Eustace
Howard Fischer
Ross Garon
Bill Gates
Jonathan Goldberg

Drew Myers
John Rapaport
Chris and Crystal Sacca
Michael Smith
Andrew Stark
Bill Trenchard

SCoPEx also received in-kind support from NOAA, which provided the POPS instrument that will provide size-resolved measurements of particle concentration. These measurements are important for understanding the physical and chemical interactions of stratospheric aerosols under ambient and perturbed conditions.

Question: Are all funding sources, including level of support, clearly identified and publicly listed?

Response: Yes. All funding sources are publicly listed online.

The SCoPEx FAQ states the following: "Who is providing the funding? Experimental hardware and operations are funded from internal Harvard research funds provided to Professors David Keith and Frank Keutsch. Additional research funding is provided by Harvard's Solar Geoengineering Research Program (SGRP). All donations to SGRP are philanthropic."

The SGRP website publicly lists all of the foundations and individual donors who have supported the program. It does not publicly list levels of support to protect our donor's privacy, which is common practice amongst NGOs that accept philanthropic gifts, including those that currently support solar geoengineering research, such as the Environmental Defense Fund, Natural Resources Defense Council, and Union of Concerned Scientists. However, because it is important for the SCoPEx Advisory Committee to be able to assess the proportional make up of donations, we have provided the amounts of each donation to the committee in Appendix B. Note that we provide that information solely for the use of the committee to assess conflicts of interest and other financial implications of the donations, but that we do so under the agreement that the committee not release this information publicly. We have also included the SGRP gift letter in Appendix A.

Question: Does the project have any anonymous individual supporters? If so, why?

Response: No, we do not accept anonymous donations.

Question: Have institutional donors identified all sources of funds, including individual donors, corporations, etc.?

Response: No, while we do ask a range of questions, we have not asked foundations or individual donors to provide information as to where *all* of their funds (in the case of foundations) or wealth (in the case of individuals) was generated from. This is in part because it would not be possible as a matter of privacy to ask individual donors to share their *entire* financial portfolio. That said, we do research to identify all publicly available sources of funding, and we ask a range of questions to determine whether the potential foundation or donor has a conflict of interest and could benefit significantly from slowing down the rate of greenhouse gas reductions. In such a case, we would not accept the donation. We provide more information about this latter piece below.

Question: Are any project funders associated with an institution that stands to benefit, financially or politically, from the results of this work? If yes, explain.

Response: Not to our knowledge.

Question: Was any of the funding directed to specific activities?

Response: There was one case in which a donation was directed to a specific activity. Before SGRP formally launched, The Alfred P. Sloan Foundation provided funding to SGRP and the Emmett Center on Climate Change and the Environment at the University of California, Los Angeles to host the Forum on U.S. Solar Geoengineering Research. This Forum was held at the Conference Center of the Carnegie Endowment for International Peace in Washington, DC on March 24, 2017. All information about the Forum, including the funding source, was and is listed publicly online. The event was also livestreamed at the time to ensure the conversations were transparent.

Otherwise, to date, all funds donated to SGRP have supported the program broadly.

Question: Does the project intend to create any new or novel technology and how will it be shared with the public? Is there an intention to create intellectual property?

Response: No, we do not intend to create intellectual property.

One of SGRP's core principles is to operate in a way that is open access across all activities. As we list publicly on our website, we aim to provide "full transparency with open-access publications and liberal data sharing," and we "discourage patents and any form of IP protection."

Because of this, key SCoPEx personnel have personally committed to not file for patents associated with SCoPEx, including Frank Keutsch, David Keith, Norton Allen, Martin Breitenlechner, John Dykema, Mike Greenberg, Michael Litchfield, Terry Martin, Marco Rivero, and Yomay Shyur. In fact, David Keith and John Dykema authored a blog post on this topic, explaining why they oppose commercial work on solar geoengineering and will not file solar geoengineering patents.

To be clear, SGRP actually would have liked to forbid patenting for any solar geoengineering related technologies it supported, but there is not a legal way to do so. Still, while it technically may be true that Harvard owns intellectual property arising from research conducted using university resources, based on Harvard's IP Policy and the individual Participation Agreements faculty and researchers sign, as a practical matter the university will not file to protect or enforce intellectual property against the wishes of the contributing faculty member. Moreover, neither SGRP nor its donors can make any claim on the intellectual property related to the experiment or other research endeavors.

As it relates to activities outside of Harvard, we cannot prevent third-party contractors from filing for patents. That said, the work currently being carried out by the third-party contractors is generally not directly related to the science or hardware that would be useful in *actual* solar geoengineering deployment; their work is more focused on balloon designs and other hardware that is highly unlikely to be used if solar geoengineering were implemented on a large scale (since aircraft are more likely to be used for deployment compared to balloons). For example, our balloon flight provider could file for a patent for a new technology they create related to their balloon design, but that would not be relevant to actual solar geoengineering deployment.

On this point, we would like to make clear that we are not conducting SCoPEx to develop hardware that can be used for deployment. In fact, this is one of the reasons why we chose to loft the particles using a balloon rather than an aircraft. Overall, the purpose of SCoPEx is NOT to advance our understanding of the aircraft or other platforms for deployment of solar geoengineering. It aims to reduce the uncertainty around specific science questions by making quantitative measurements of some of the aerosol microphysics and atmospheric chemistry required for estimating the risks and benefits of solar geoengineering in large atmospheric models.

Question: What are the relevant policies around funding?

Response: In addition to Harvard's standard funding policies, SGRP follows two further policies:

1) We do not accept anonymous donations.
2) We do not accept donations from corporations, foundations, or individuals if the majority of their current profits or wealth come from the fossil fuel industry unless they can clearly demonstrate that they do not have a conflict of interest and present a strong track record of supporting efforts to address climate change.

We are concerned that fossil fuel companies or other interests will seek to exploit solar geoengineering as a pretext for delaying reductions in greenhouse gas emissions. We do not want donors who are (or could reasonably be construed as being) motivated to support solar geoengineering research to protect fossil fuel industries. For purposes of excluding such donors, we consider a rough weighting system as a guide. We rate the donor's ties to fossil fuels on a 1 to 5 scale, where 1 has no connection with fossil fuels and 5 has nearly all of their current wealth and social connections tied to coal. Then, we rate the donor's commitment to climate from 1 for a donor who has long devoted a majority of their time and resources to climate action to 5 for a donor who has no visible interest in climate. We then take the product of the two ratings, rejecting donors with a multiplicative combined rating that is larger than 10.

We would like to elaborate on this last point. We take issues of conflict of interest very seriously. And we take the "moral hazard" concern very seriously—the idea that research or even discussion on solar geoengineering could reduce incentives to mitigate. The world must reduce greenhouse emissions to zero, and remove carbon dioxide from the atmosphere, to address the root cause of climate change. Solar geoengineering does and will not change this fact.

We offer a few examples of our funding decisions:

- We would not accept funding from Exxon both because the company would benefit from prolonging the use of fossil fuels and because it has clearly undermined efforts to meaningfully address climate change. In other words, we would rate Exxon with a 5 x 5 = 25.
- We would accept funding from Tom Steyer or The Rockefeller foundation because they no longer would benefit from a delay in fossil fuel use even though their wealth was generated from investments in the fossil fuel industry (N.B. neither have donated to SGRP, this is illustrative.) Here, we would rate Rockefeller as 3 x 2 = 6.

Question: The Research Team will provide a conflict of interest statement for experiment and major funding sources.

Response:

Conflict of Interest – "Moral Hazard"
As we noted above, we take issues of conflict of interest very seriously. And we take the "moral hazard" concern very seriously—the idea that research or even discussion on solar geoengineering could reduce incentives to mitigate.

The world must reduce greenhouse emissions to zero, and remove carbon dioxide from the atmosphere, to address the root cause of climate change. Nothing about solar geoengineering changes this fact. But we, like others, are concerned that fossil fuel companies or other interests will seek to exploit solar geoengineering to slow down or block mitigation.

To address this concern in our own work, SGRP does not accept donations from corporations, foundations, or individuals if the majority of their profits or wealth come from the fossil fuel industry unless they can clearly demonstrate that they do not have a conflict of interest and present a strong track record of supporting efforts that address climate change.

Conflict of Interest – Harvard University
Harvard University also has a strict set of policies regarding institutional conflict of interest. SGRP embraces these policies.

Below, we offer an example when we initially questioned whether there was a conflict of interest. We immediately and proactively reached out to university officials, who then independently followed established policies and principles and ultimately determined there was not a conflict.

Background
Last year David Keith had the opportunity to serve on Harvard's university-wide committee to develop guidelines for Institutional Conflict of Interest. This process evaluated conflicts of interest that reflect on the institution as a whole and created a policy for dealing with them. One of the topics discussed at length was dealing with fellowships and similar affiliations from people with ties to donors. That experience alerted David to the importance of this issue and to the various ways it is handled at Harvard and other institutions. His view is that the way the Mossavar-Rahmani Center for Business and Government and SGRP handled Wake Smith's fellowship application was consistent with the way Harvard's conflict of interest policy will eventually emerge and with the way these matters are treated at peer institutions.

Context
Wake Smith is a retired aerospace executive who has become interested in advancing solar geoengineering research. In 2018, Wake collaborated with Gernot Wagner to write an article that drew upon his experience in the aerospace industry. After Wake independently began his research, he and his family wished to support solar geoengineering research and donated funds to SGRP through their personal foundation "The Crows Nest Foundation" (though which they make nearly all of their philanthropic donations). Last fall, Wake became a M-RCBG Senior Fellow at the Harvard Kennedy School's Mossavar-Rahmani Center for Business and Government (M-RCBG).

This circumstance presents the appearance of a conflict of interest in that people might conclude that the decision to grant the M-RCBG Senior Fellowship was influenced by Wake's donation. But this was not the case, and internal documentation demonstrates this.

First, Wake's donation was explicitly acknowledged in the Wagner and Smith paper. It stated: "WS began work on this analysis independently. He subsequently became a donor of Harvard's Solar Geoengineering Research Project, co-directed by GW."

Second, David Keith raised the issue of potential conflict of interest in writing as soon as he heard about Wake's formal application from colleagues at M-RCBG. They discussed it and the Harvard Kennedy School independently carried out a review process and concluded that there was not substantive conflict of interest for the following reasons. (a) Wake's donation was to SGRP and no SGRP funds have flowed to M-RCBG. (In other words, Wake's contribution did not go to or benefit the program that he applied to.) (b) Wake's donation to SGRP was small, less than 0.5% of total funds raised by that date. (c) Wake's application was exceptionally strong and would have been awarded based on merit (had a donation never been made). Nevertheless, because of reasonable concerns about the appearance of conflict of interest M-RCBG added a note disclosing the donation on the webpage that announced Wake's fellowship.

Separately, after Wake was accepted, we took action related to a different matter. After Wake's acceptance, M-RCBG listed David as Wake's faculty mentor because of his subject-matter knowledge of solar geoengineering. David, however, did not realize this was the case, so once he was alerted to this fact, he corresponded with M-RCBG and they immediately replaced him with Joe Aldy as Wake's faculty mentor. In hindsight, David should have understood that he was to be listed as Wake's faculty mentor and declined that duty.

We hope this example demonstrates the often complex questions that can be raised around potential conflict of interest, and how SGRP and Harvard University handles these circumstances, following established policies and principles.

Appendix A

Appendix A may be made public.

Solar Geoengineering Research Program Fund

The gifts of alumni and friends of Harvard University establish the *Solar Geoengineering Research Program Fund*. Housed in the Harvard University Center for the Environment (HUCE), this current-use fund shall support the activities of Harvard's Solar Geoengineering Research Program (SGRP), a research endeavor currently under the leadership of David Keith, Gordon McKay Professor of Applied Physics and Professor of Public Policy, to bring together an interdisciplinary group of faculty from schools such as the Faculty of Arts and Sciences, the Harvard Paulson School of Engineering and Applied Sciences, and the Harvard Kennedy School of Government, to accelerate the understanding of the effectiveness and risks of solar geoengineering.

Toward this end, this fund may support any costs of the project including, but not limited to:

- student research and support;
- exploratory seed grants for faculty;
- post-doctoral fellows, visiting scholars and researchers;
- experimental research and associated equipment;
- workshops, symposia, and conferences;
- administration and staff.

The fund will be managed by Harvard in accordance with its policies, including gift policies. The investment, administration, and distribution of the fund shall be accomplished in accordance with University policies, including gift policies, governing endowment and certain other institutional funds, which may be amended from time to time. Under current policies, a portion of the amount made available for annual spending may be applied to defray direct and indirect facilities and administrative costs.

If, at some time in the future, the designation of these funds is no longer appropriate, the Edgerley Family Dean of the Faculty of Arts and Sciences, in consultation with the Faculty director(s) of HUCE, may direct the fund to another purpose deemed to be best in keeping with the original purpose of this fund.

The following page offers information on how to make a gift to this fund. If you have questions about how to make a gift, please visit the Harvard website or contact Jane Van Velden at jane_vanvelden@harvard.edu.

Appendix A-3

Advisory Committee Request for Additional Information

Professor David W. Keith
Gordon McKay Professor of Applied Physics
John A. Paulson School of Engineering and Applied Sciences
Professor of Public Policy, Harvard Kennedy School of Government
Harvard University, Pierce Hall, 29 Oxford Street
Cambridge, MA 02138

Dear Professor Keith,

As you are aware, the overarching mission of the SCoPEx Advisory Committee is to ensure that the SCoPEx project is undertaken in a transparent, responsible, and legitimate manner and that it meaningfully contributes both to science and to building appropriate public engagement and trust. Operating independently from the research team we consider our mandate is to provide advice and guidance on the research and governance of the project.

Our work comprises four complementary elements:

- A technical review
- A legal and regulatory review
- A societal review, and
- A financial review

The results of these reviews will inform the committee's recommendations on whether and how SCoPEx should move forward, and we are grateful for your assurance that our input will be taken seriously and fully considered as part of the research process.

It is against this background that we have undertaken the financial review. This review is based on the information you have kindly shared with the Committee, our informal discussions with you (on 08/20 and 10/2/22) and Harvard's public disclosure (https://geoengineering.environment.harvard.edu/funding). This review has identified the following concerns/issues related to the financial disclosures. We request clarification from you, on the following, to assist in finalizing our review.

1. In your memorandum to the Committee dated June 10, 2020 it was stated *inter alia* that "We do not accept donations from corporations, foundations or individuals if the majority of their current profits or wealth come from the fossil fuel industry, unless they can clearly demonstrate that they do not have a conflict of interest and present a strong track record of supporting efforts to address climate change." In addition, you provided two examples as to why funding from fossil fuel derived wealth would be unacceptable (in the case of Exxon) but acceptable (in the case of Tom Steyer or Rockefeller Foundation), based on what the Committee considers to be a relatively subjective rating scale. In light of these criteria, can you clarify (a) how the contribution from Laura and John Arnold is consistent with your policy?

2. The Committee notes that at least one donor (Bill Gates[1]) has funded other experiments and research on large scale SRM. Do any project funders, whether

[1] The Committee is advised that this donation is not from the BMGF, but from Gates' personal funds.

individuals, foundations, institutions, corporations, affiliates or collaborators stand to benefit financially or politically from the results of this work? The Committee would also be grateful to be informed of the actions that you have taken (or intend to take) to mitigate such potential conflicts of interest.

3. The Committee is advised that there are different kinds of intellectual property that could be generated from SCoPEx, including copyright, patent, trademark, industrial design and geographical indicators. We would welcome a clear statement of the specific principles governing intellectual property generated by the project, and how these principles will apply to members of the SCoPEx project team, its contractors, collaborators and other third parties. As you have stated that all intellectual property from the project will be in the public domain would you consider publishing it under the relevant Creative Commons license structure?

In addition to the specific issues raised above, we seek your assurance that:

1. The Advisory committee will be kept updated on any changes to current Harvard and SCoPEx policy governing the acceptance of philanthropic donations.

2. The Advisory Committee will be informed of the identity of all additional sources and amount of monetary and in-kind support accepted for the SCoPEx experiment.

3. All funding sources will be clearly identified and publicly listed.

4. SCoPEx does not accept resources from anonymous donors.

5. Donations are accepted to provide broad project support, and are without conditionalities that direct the use of such funding to specific project activities.

We look forward to receiving a response to the matters raised, and welcome any further information that would assist the Committee in completing the financial review.

Appendix A-4

Research Team Response to Request for Additional Information

HARVARD
John A. Paulson
School of Engineering
and Applied Sciences

HARVARD Kennedy School
JOHN F. KENNEDY SCHOOL OF GOVERNMENT

November 20, 2020

Dear SCoPEx Advisory Committee,

Thank you for undertaking a financial review process for SCoPEx. We are happy to answer the follow up questions you have proposed. See enclosed document.

Please don't hesitate to reach out with questions. We can always setup a call to discuss further.

Yours,

David Keith
Gordon McKay Professor of Applied Physics, School of Engineering and Applied Sciences (SEAS); and,
Professor of Public Policy, Kennedy School of Government,
Harvard University

Question 1: In your memorandum to the Committee dated June 10, 2020 it was stated inter alia that "We do not accept donations from corporations, foundations or individuals if the majority of their current profits or wealth come from the fossil fuel industry, unless they can clearly demonstrate that they do not have a conflict of interest and present a strong track record of supporting efforts to address climate change." In addition, you provided two examples as to why funding from fossil fuel derived wealth would be unacceptable (in the case of Exxon) but acceptable (in the case of Tom Steyer or Rockefeller Foundation), based on what the Committee considers to be a relatively subjective rating scale. In light of these criteria, can you clarify (a) how the contribution from Laura and John Arnold is consistent with your policy?

Response: John and Laura Arnold's donation is consistent with our policy because we believe their contribution does not present a conflict of interest. Between their strong record of supporting efforts to address climate change and their investment in renewables (amongst oil and gas investments), we feel the Arnolds are <u>not</u> seeking to support and exploit solar geoengineering as a pretext for delaying reductions in greenhouse gas emissions—our (and others') main concern.

Yet, their contribution does provide a useful, concrete example of how complicated it is to determine a potential conflict of interest in the real world, since it is not uncommon for individuals, foundations, or corporations to have some fraction of wealth or profits connected to the fossil fuel industry. Indeed, in a world that depends on fossil fuels for energy, arguably any entity is so tied (including Harvard). We therefore provide more information below.

John and Laura Arnold are contributing to the climate effort in many important ways. John is the lead director at Breakthrough Energy and the Environmental Defense Fund's methane satellite detection project, and he and Laura have made significant philanthropic gifts to several climate organizations and initiatives, including Citizens Climate Education Corp, Clean Air Task Force Inc., Climate Leadership Council Inc., Energy Innovation Reform Project, and the Environmental Defense Action Fund. Therefore, if we were to use our rough weighting system[1] as a guide, our view is that John and Laura's rating on climate action is a 2 or 3. The main reason we did not assign a 1 is because climate change is not the sole focus of John and Laura's philanthropic efforts (they are also involved in other issues such as education, health, and criminal justice).

John and Laura also have current investments in the energy sector. Our funding policy focuses on funders' current portfolio rather than past since we believe that is the best indicator as to whether there could be a potential conflict of interest (hence why we would accept funds from Tom Steyer, as you noted above). John and Laura's current portfolio includes a mixture of oil and gas as well as clean energy investments. We could therefore understand if one assigned a rating on his current investments as a 3 or 4.

These initial estimates suggest that John and Laura's product would fall somewhere between a 6 and 12. Clearly, it is complicated. In such cases, we return to the heart of our conflict of interest question: is the donor seeking to support and exploit solar geoengineering as a pretext for delaying reductions in greenhouse gas emissions? From our point of view, John and Laura's leadership roles in climate initiatives, significant donations to climate organizations, and investments in renewable energy demonstrate a real interest in and commitment to reducing greenhouse gas emissions despite their investments in oil and gas. We recognize, however, that our final judgment is subjective and could

[1] We rate the donor's ties to fossil fuels on a 1 to 5 scale, where 1 has no connection with fossil fuels and 5 has nearly all of their current wealth and social connections tied to coal. Then, we rate the donor's commitment to climate from 1 for a donor who has long devoted a majority of their time and resources to climate action to 5 for a donor who has no visible interest in climate. We then take the product of the two ratings, rejecting donors with a multiplicative combined rating that is larger than 10.

certainly be critiqued. We mainly hope this process sheds light on the nuances and complexities of this process in the real world and is useful to others in the future if they choose to adopt such a model.

Question 2: The Committee notes that at least one donor (Bill Gates) has funded other experiments and research on large scale SRM. Do any project funders, whether individuals, foundations, institutions, corporations, affiliates or collaborators stand to benefit financially or politically from the results of this work? The Committee would also be grateful to be informed of the actions that you have taken (or intend to take) to mitigate such potential conflicts of interest.

Response: To our knowledge, no project funders stand to benefit financially or politically from the results of this work.

First, all donations to SGRP are philanthropic, meaning that funders cannot have any *direct* financial return on investment from any gift they give to our program. Their donation is simply that—a charitable gift. And given Harvard's very high level of public visibility, the university is very strict about applying these standards. In fact, this control is at the level of Harvard senior management, so the teams from Harvard's Solar Geoengineering Research Program (SGRP) and/or SCoPEx could not overrule these policies even if we wanted to (which we don't).

Second, funders cannot have any claim over the intellectual property of the program's work. Harvard owns the intellectual property arising from research conducted using university resources (more on this below). This further reinforces the fact that funders cannot have a *direct* financial benefit from the results of our research.

Third, to address *indirect* concerns, we do not accept donations from funders who are seeking to exploit solar geoengineering for personal financial gain in the fossil fuel industry, as noted above. This is why we do not accept donations from corporations, foundations, or individuals if the majority of their current profits or wealth come from the fossil fuel industry unless they can clearly demonstrate that they do not have a conflict of interest and present a strong track record of supporting efforts to address climate change. To mitigate such conflicts of interest, we have implemented the weighting system we outlined in our financial statement.

Question 3. The Committee is advised that there are different kinds of intellectual property that could be generated from SCoPEx, including copyright, patent, trademark, industrial design and geographical indicators. We would welcome a clear statement of the specific principles governing intellectual property generated by the project, and how these principles will apply to members of the SCoPEx project team, its contractors, collaborators and other third parties. As you have stated that all intellectual property from the project will be in the public domain would you consider publishing it under the relevant Creative Commons license structure?

Response: One of SGRP's core principles is to operate in a way that is open access across all activities. As we list publicly on our website, we aim to provide "full transparency with open-access publications and liberal data sharing," and we "discourage patents and any form of IP protection."

If it were possible, SGRP would forbid patenting for any solar geoengineering related technologies it supported. But there is not a legal way to do so. Harvard owns the intellectual property arising from research conducted using university resources, based on Harvard's IP Policy and the individual Participation Agreements faculty and researchers sign. We therefore cannot, for example, force a graduate student working on SCoPEx to not file for a patent. In practice, key SCoPEx personnel have personally committed to not file for patents associated with SCoPEx, including Frank Keutsch, David Keith, Norton Allen, Martin Breitenlechner, John Dykema, Mike Greenberg, Michael Litchfield, Terry Martin, Marco

Rivero, and Yomay Shyur. And Harvard would not practically file to protect or enforce intellectual property against the wishes of the contributing faculty member. Yet, legally, SGRP's hands are tied.

We therefore seek the Advisory Committee's advice as to how we best manage the intellectual property question within the constraints of Harvard's policies, as this will likely be relevant to programs and researchers at other universities.

Similarly, we cannot prevent a third-party contractor from filing for patents since they (not we) own the technology that they create. Importantly, however, we have not and do not expect to contract with a third-party vendor for work that could result in a patent of a core piece of solar geoengineering technology.

Right now, this is easy because nothing on our research radar would have us involved with subcontractors on technologies that would be core to solar geoengineering deployment. For example, in the case of SCoPEx specifically, this is not an issue because any hardware that the balloon vendor develops will not be core to solar geoengineering. It may, for example, be useful to a range of stratospheric balloon flights, including those unrelated to solar geoengineering experiments (if, of course, any new technology is developed at all), but it will not be specific or central to solar geoengineering. This is largely because stratospheric solar geoengineering would most likely be deployed by aircraft, not balloons, if deployed at all.

Hard questions will arise if future research involves a contract with a third-party vendor for work that could result in a patent of a core piece of solar geoengineering technology. We don't know the best path. Perhaps the contract might legally commit the firm to allow for any such technology to undergo rigorous, independent, third party evaluation by multiple entities, including governments and intergovernmental organizations.

Overall, the question of commercial sector engagement is complicated, and we are still forming our own views as we continue to learn more. We certainly welcome critiques and encourage people to read many great publications in this space, including *Intellectual Property Policies for Solar Geoengineering* (Reynolds, Contreras, and Sarnoff, 2018), which provides useful background information on the issues and challenges confronting the use of patents and trade secrets in solar geoengineering technologies.

To us, the central issue with commercial involvement in solar geoengineering is that commercial entities could have divergent interests from the public good. They would, for example, have some interest in hiding risks and presenting benefits that make a technology look better than it might actually be, have the resources to market a technology so that it appears better than another, and have an incentive to accelerate the deployment of solar geoengineering so their technology is used, amongst many other serious concerns.

This is not to say that the commercial sector will not or should not ever be involved in solar geoengineering. Indeed, likely through a procurement structure, there may be areas where private sector innovation can meaningfully contribute, as it has in other areas related to the public good, such as COVID vaccine development. Yet, in the case of vaccines, there is currently a rigorous process in place that allows for independent, third-party testing of the commercial entities' results. The FDA, for example, is overseeing the efficacy and safety of any potential vaccine that could be distributed in the US so that the public can be meaningfully informed of the potential benefits and risks of any particular immunization. There needs to be this level of transparent, independent, rigorous oversight of any private sector technological developments that are patented and are core to solar geoengineering so that governments, scientists, and people around the world can seriously evaluate and understand their risks.

Additional Information: In addition to our above responses, you have sought our assurances that:

1. The Advisory committee will be kept updated on any changes to current Harvard and SCoPEx policy governing the acceptance of philanthropic donations.
2. The Advisory Committee will be informed of the identity of all additional sources and amount of monetary and in-kind support accepted for the SCoPEx experiment.
3. All funding sources will be clearly identified and publicly listed.
4. SCoPEx does not accept resources from anonymous donors.
5. Donations are accepted to provide broad project support, and are without conditionalities that direct the use of such funding to specific project activities.

We can commit to requests numbered 1, 2, 3, and 4, but we cannot commit to number 5.

First, experiments proposed by researchers outside of Harvard may not be housed within a program such as ours (SGRP) and may therefore need to raise funds for the specific project. Researchers at the University of Washington, for example, do not have a formal Solar Geoengineering Research Program at their institution, yet they are raising funds for a proposed, small-scale marine cloud brightening experiment. Why would it be problematic for a research team to raise funds for a specific experiment, assuming they do so in a manner that is transparent, avoids conflict of interest, and follows other principles and guidelines that you recommend?

Second, based on the societal engagement process you have proposed, we may need to work (perhaps in concert with you) to raise funds to support the experiment's engagement process. Hopefully the initial amount of funds SGRP awarded to the Advisory Committee to carry out its work, totaling $335,000, can fund portions of this societal engagement process. But if some of the activities proposed require additional funds, we may need to raise separate funds to specifically support the societal engagement process since SGRP has a limited budget (given its mission to support a range of interdisciplinary research across Harvard's campus). On a positive note, this may not necessarily be a terrible outcome so long as the funds needed are not exorbitant and out of reach of others. In fact, if we can generate philanthropic interest and establish a sustainable funding model for societal engagement activities, it may be useful for future small-scale outdoor experiments looking to adopt such an engagement model.

Appendix

[redacted].

[redacted]

[redacted]

[redacted]

[redacted]

Appendix A-5

Additional Updates from the Research Team

HARVARD John A. Paulson School of Engineering and Applied Sciences

HARVARD Kennedy School JOHN F. KENNEDY SCHOOL OF GOVERNMENT

August 19, 2020

Dear SCoPEx Advisory Committee,

Thank you again for your work on the financial aspects of SCoPEx. On June 11, 2020, we shared our formal response to your request regarding the financials and conflict of interest principles of the experiment. Since that time, we have received a new donation that we wanted to share with you.

On August 6, 2020, Jeffrey T. Haley, Director of the Reflective Earth Foundation, made a donation.

As before, we have provided the amount of Mr. Haley's donation in an Appendix, which can be shared privately with the SCoPEx Advisory Committee with the agreement that the information not be released publicly. This is different from this letter, which can be shared publicly.

Mr. Haley's donation aligns with Harvard's and SGRP's fundraising policies, which we outlined in our prior document. In sum, in addition to Harvard's standard funding policies, SGRP follows two policies:

1. We do not accept anonymous donations.
2. We do not accept donations from corporations, foundations, or individuals if the majority of their profits or wealth come from the fossil fuel industry unless they can clearly demonstrate that they do not have a conflict of interest and present a strong track record of supporting efforts to address climate change.

Please don't hesitate to reach out with questions. I am happy to setup a call to discuss.

Yours,

David Keith
Gordon McKay Professor of Applied Physics, School of Engineering and Applied Sciences (SEAS); and,
Professor of Public Policy, Kennedy School of Government,
Harvard University

HARVARD
John A. Paulson
School of Engineering
and Applied Sciences

HARVARD Kennedy School
JOHN F. KENNEDY SCHOOL OF GOVERNMENT

January 13, 2020

Dear SCoPEx Advisory Committee,

Thank you again for your work on the financial aspects of SCoPEx. Since our June 11, 2020 formal response to your request regarding the financials and conflict of interest principles of the experiment, we have received another new donation that we wanted to share with you.

In late December 2020, the Tansy Foundation made another donation (the foundation donated to SGRP previously, as noted on in our prior response and as listed publicly on our website).

As before, we have provided the amount of the Tansy Foundation's donation in an Appendix, which can be shared privately with the SCoPEx Advisory Committee with the agreement that the information not be released publicly. This is different from this letter, which can be shared publicly.

The Tansy Foundation's donation aligns with Harvard's and SGRP's fundraising policies, which we outlined in our prior document. In sum, in addition to Harvard's standard funding policies, SGRP follows two policies:

1. We do not accept anonymous donations.
2. We do not accept donations from corporations, foundations, or individuals if the majority of their profits or wealth come from the fossil fuel industry unless they can clearly demonstrate that they do not have a conflict of interest and present a strong track record of supporting efforts to address climate change.

Please don't hesitate to reach out with questions. I am happy to setup a call to discuss.

Yours,

David Keith

David Keith
Gordon McKay Professor of Applied Physics, School of Engineering and Applied Sciences (SEAS); and,
Professor of Public Policy, Kennedy School of Government,
Harvard University

Appendix B

Legal Review

Appendix B-1

Scope of Work for Legal Review of Potential Launch in Sweden

Scope of Work

1. Setterwalls Advokatbyra AB (“Setterwalls”) has been retained by Harvard to conduct a legal review in relation to the Project.
2. The Swedish legal aspects of the Project in respect of which Harvard has instructed Setterwalls to provide legal advice are the following:
   a) To verify that SSC has all the necessary regulatory permits and approvals and would be in compliance with Swedish laws in order to carry out the First Phase, limited to the testing of the navigation of one balloon and the gondola to be launched over Sweden into the stratosphere.
   b) As regards the First Phase, to which extent
   (i) Swedish environmental law and/or environmental EU directives and regulations are applicable,
   (ii) regulations on environmental impact assessments are applicable, and
   (iii) potential other relevant Swedish laws are applicable.
   c) To address whether the potential implementation of the Second Phase of the Project including release of material into the stratosphere would in any way affect the conclusions related to the questions in respect of the First Phase.

Appendix B-2

Legal Review Memo from Setterwalls

SETTERWALLS

MEMORANDUM

to

Harvard University

regarding

Project SCoPEx

18 February 2021

SW41029167/4

A. Executive Summary 3
B. Scope of work 4
C. The SCoPEx project 5
C.1 First Phase 5
C.2 Second Phase 6
D. Required permits of SCC 7
D.1 Permit to fly unmanned free balloons 7
D.2 Permit to operate in restricted areas 8
D.3 Permit regarding radio communications 8
D.4 Safety laws and regulations etc. regarding Esrange 8
D.4.1 Compliance to safety laws and regulations 8
D.4.2 Certain geographical safety regulations for Esrange 8
D.5 Conclusions 9
E. Relevant legislation 10
E.1 Introduction 10
E.2 The Space Act 10
E.3 The Environmental Code 11
E.3.1 Environmental Impact Assessments 11
E.4 The Civil Aviation Act 12
E.5 EU regulations and international conventions regarding the environment ratified by Sweden 13
E.6 Conclusions 13
F. Impact on the First Phase due to the potential implementation of the Second Phase 14

SW41029167/4

A. Executive Summary

- Harvard University ("Harvard") is contemplating undertaking the scientific geoengineering project SCoPEx (Stratospheric Controlled Perturbation Experiment) with the aim to advance science on efficiency and risks of solar geoengineering ("the Project"). The Project includes two phases, the First Phase and the Second Phase. The First Phase involves launching a high-altitude balloon with a gondola to test the navigation system. The current tentative plan is to take place in June 2021 and Swedish Space Corporation ("SSC") at Esrange Space Center in Kiruna, Sweden ("Esrange") will manage and provide certain flight services for the test. This has been agreed in an agreement between Harvard and SSC dated 18 December 2020 ("the Agreement"). The test will not include *any* release of aer-osol injections or other materials into the stratosphere.
- The Second Phase, which is yet uncertain when and where to be carried out (if at all carried out) is not subject to the Agreement with SSC and is expected to include the release of approximately 100 – 2,000 g of aerosols into the stratosphere. Currently, the intention is to use calcium carbonate and/or other materials such as sulfates.
- Esrange is owned and operated by SSC. SSC has all the necessary permits and regulatory approvals to carry out the First Phase of the Project pursuant to the Swedish Electronic Communication Act. As Harvard will use a certain satellite telephone instead of a radio transmitter, Harvard will not need to obtain a permit regarding radio communication.
- The Swedish Space Act is not applicable on the Project as it will take place in the stratosphere. Swedish environmental law does not affect the First Phase of the Project and an environmental impact assessment is not required.

SW41029167/4

B. Scope of work

1. Setterwalls Advokatbyrå AB ("Setterwalls") has been retained by Harvard to conduct a legal review in relation to the Project.

2. The Swedish legal aspects of the Project in respect of which Harvard has instructed Setterwalls to provide legal advice are the following:

   a) *To verify that SSC has all the necessary regulatory permits and approvals and would be in compliance with Swedish laws in order to carry out the First Phase, limited to the testing of the navigation of one balloon and the gondola to be launched over Sweden into the stratosphere.*

   b) *As regards the First Phase, to which extent*

      (i) Swedish environmental law and/or environmental EU directives and regulations are applicable,

      (ii) regulations on environmental impact assessments are applicable, and

      (iii) potential other relevant Swedish laws are applicable.

   c) *To address whether the potential implementation of the Second Phase of the Project including release of material into the stratosphere would in any way affect the conclusions related to the questions in respect of the First Phase.*

SW41029167/4

C. The SCoPEx project

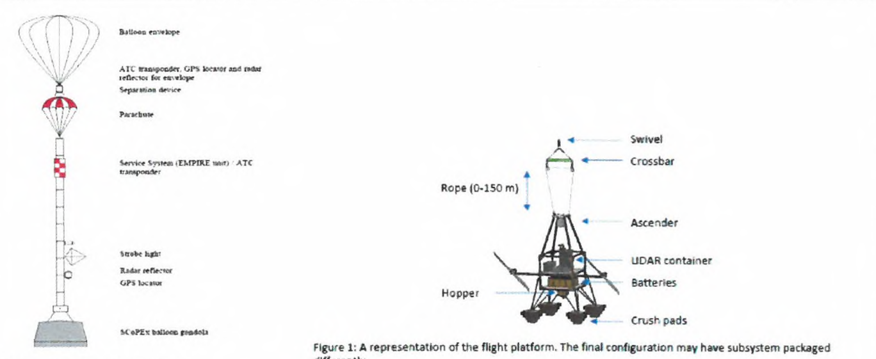
3. The purpose of the Project is to advance understanding of stratospheric aerosols that could be relevant to solar geoengineering, by simulations to provide modellers with experimental results vital to address specific scientific questions. According to the description of the Project, such simulations are the primary tool for estimating the risks and benefits of solar geoengineering. Currently, there is a concern within the Project that limitations of information results in an overestimation of the simulations. Moreover, an advance knowledge on interaction of particles in the stratosphere may increase the expertise on how to mitigate the global warming by preventing solar rays from reaching the earth.[1] The intention is that the Project will consist of two phases, The First Phase and the Second Phase.

C.1 First Phase

4. The First Phase is a flight test where an unmanned free balloon, carrying a gondola with scientific equipment, will be launched to an altitude of 20 - 22 km (which constitutes the stratosphere[2]) from Esrange.

5. Esrange is located in northern Sweden (approximately 45 km east of Kiruna) and is since 1972 owned and operated by SSC, which is wholly-owned by the Swedish State. SSC's operation consist of a *public assignment* combined with a *commercial assignment*. The public assignment is merely to own, operate and develop Esrange. The commercial part includes three business areas: Science & Launch Services, Satellite Ground Network Services and Spacecraft operations & Engineering Services. Stratospheric balloon launches are performed by the Science and Launch Services department and has been conducted at Esrange since 1966. The launched balloons have various scientific or technical instruments on board for research and technical development.

6. The flight platform that will be tested has not flown before. The balloon is owned by Harvard and manufactured by the US company Raven Aerostar. The nominal size and weight of the balloon is 16,766 cubic meters and 176 kg, respectively.



A schematic view of the aerostat configuration.

Figure 1: A representation of the flight platform. The final configuration may have subsystem packaged differently.

---

[1] https://www.keutschgroup.com/scopex (2021-02-07).
[2] https://www.nasa.gov/mission_pages/sunearth/science/atmosphere-layers2.html (2021-02-05).

7. The gondola, also owned by Harvard, has a size of circa 2.5 x 2.5 x 3.5 m, and a weight of around 600 kg. Additional equipment weighing approximately 300 kg will *inter alia* be the parachute, communication equipment, ballast, flight safety system etc. The balloon and gondola including the additional equipment will have a total weight of approximately 1,085 kg, see schematic views of the balloon and gondola above.

8. The purpose of the First Phase launch is purely to test the navigation of the balloon and gondola. No material will be released into the stratosphere. At the end of the flight of the First Phase, the ropes that suspend the gondola from the balloon will be released. The balloon will then deflate and fall to the ground in a different location from the gondola. This platform test is tentatively planned to be performed in June 2021.

C.2 Second Phase

9. The plan of the SCoPEx project in the Second Phase is to release a small amount of 100 g - 2,000 g of material into the stratosphere. Substances that are currently evaluated are calcium carbonate and/or other material such as sulfates. By measuring and observe a small controlled volume of aerosols, the understanding of, *inter alia*, the processes that can reduce or eliminate ozone loss can be improved. However, the performance of the Second Phase is not subject to the Agreement with SSC and is yet to be decided – if it should at all be performed, and if so, where it will be performed - and this legal review will not address a review of the Second Phase (unless specifically mentioned).

SW41029167/4

D. **Required permits of SCC**

We have been instructed: *To verify that SSC has all the necessary regulatory approvals and would be in compliance with Swedish laws in order to carry out the First Phase, limited to the testing of the navigation of one balloon to be launched over Sweden into the stratosphere.*

10. SSC requires the following permits related to First Phase for its operations[3]:

- Permit to fly unmanned free balloons in Sweden and permits for unmanned balloon flights through Finnish and Norwegian airspace
- Permit to operate in restricted areas
- Permit regarding radio communications

D.1 **Permit to fly unmanned free balloons**

11. General operational provisions for the launching of unmanned free balloons are found in the 1944 Chicago Convention on International Civil Aviation Rules according to the ICAO (International Civil Aviation Organization) Annex 2 - Rules of the Air, and according to Regulation (EU) No 923/2012 on Standardized European Rules of the Air ("SERA"), Annex 2, sections 2.1-2.5.

12. Section 2.1 of SERA prescribes that unmanned free balloon must not be operated without permit from the state from which the launch is made. The launching of an unmanned free balloon must not take place within Swedish territory without permit from the Swedish Transport Agency. Flights with balloons from Swedish territory into another country's territory must not take place without a permit from the other country's aviation authority.

13. SSC annually applies for permits to launch stratospheric balloons in Sweden to the Swedish Transport Agency. The most recent permit for 2021 (TSL 2020-7174) dated 13 November 2020 is referring to a certain programme including Project SCoPEx noting 1 – 30 June 2021 for one "*Heavy scientific balloon flight*" and that the air traffic control (ATS) shall be notified at latest seven days in advance of launch.

14. The reason for the permit is as follows (translated from Swedish): "*The requirements contained in Regulation (EC) No 923/2012 SERA (Standardized European Rules of the Air), Annex 2, section 2.1 prescribes that flying with an unmanned free balloon must not be commenced without permit from the state from which the launch is made.*"

15. The Swedish Transport Agency's decision has gained legal force, which means that it cannot be appealed. Harvard does not have to apply for its own permit for the launch of the balloon in the First Phase. In addition, SSC holds relevant permits

[3] It should be noted that SSC most likely hold several other permits that are not related to the First Phase of the SCoPEx project.

SW41029167/4

to fly unmanned balloons in both Norwegian and Finnish airspace between 1 January 2021 and 31 December 2021.[4]

D.2 Permit to operate in restricted areas

16. The operation of SSC is to a large extent governed by SERA. SERA is regulating common aircraft and operational provisions for services and procedures in air traffic. In accordance with SERA and chapter 1 section 4 in the Aviation Regulation (Sw. *Luftfartsförordningen* (2010:770)), the Swedish Transport Agency has authority to determine if an area should constitute a restricted area. A restricted area means that the airspace, due to for example safety reasons, becomes limited and restricted. The Swedish Transport Agency has decided that Esrange shall be a restricted area and granted SSC permission to operate in the area.[5]

D.3 Permit regarding radio communications

17. The Electronic Communications Act (2003:389) (Sw. *lag om elektronisk kommunikation*), prescribes that the use of radio transmitters on aircrafts require a certain permit.[6] The Swedish Post and Telecommunications Agency has confirmed that SSC has been granted such a permit.

18. Pursuant to 130 § in the regulation PTSFS 2020:5, Harvard does not need to apply for a permit when using an Iridium satellite phone instead of a transmitter. Harvard could operate under Iridium's license.

D.4 Safety laws and regulations etc. regarding Esrange

D.4.1 Compliance to safety laws and regulations

19. SSC Proposal section 5.5.1 refers to that SSC must comply with Swedish law and Swedish safety and security regulations applying to all activities at Esrange including the Work Environment Act (1977:1160) (Sw. *Arbetsmiljölagen*) which is the basic general law which defines the framework for provisions concerning occupational safety and health in Sweden. The purpose of the act is to prevent occupational illness and accidents and to otherwise ensure a good work environment.

D.4.2 Certain geographical safety regulations for Esrange

20. The Administrative Board of Norrbotten County decides on safety regulations for the activities at Esrange (Norrbotten County Statute Collection 25 FS 2020:29 A28). Such regulations are based on chapter 3 section 11 of the Public Order Act (1993:1617) (Sw. *Ordningslagen*) and a decision by the Government of 30 June 1972. The safety regulations deal with the geographical security protection area outside Esrange and information to the public, local authorities and reindeers

---

[4] According the SSC SCoPEx 2021 proposal dated 9 December 2020, section 5.2, permits have also been granted by transport authorities in Norway and Finland. This has been verified by the provision of decisions of the Norwegian Civil Aviation Authority dated 13 November 2020 and the Finnish Transport and Communication Agency dated 20 November 2020, respectively.
[5] The Transport Agency has confirmed this by email dated 8 February 2021.
[6] Chapter 3 section 1 of the Electronic Communications Act. In accordance with the preparatory work to the Swedish Aviation Act, a balloon is included in the definition of an "aircraft", government bill 2009/2010 p. 293.

herders to stay out of the area in connection with launching etc. Non-compliance with the regulations may result in fines.

D.5 Conclusions

21. SCC holds the necessary permits to fly unmanned balloons in both Sweden, Norway and Finland. SSC has confirmed that is does not consider it necessary with a permit for any other country in respect of the Project.

22. Harvard does not need to apply for a permit regarding radio communication equipment when using an Iridium satellite telephone instead of a radio transmitter. Harvard could operate under Iridium's license.

SW41029167/4

E. Relevant legislation

We have been instructed to investigate: *As regards the First Phase, to which extent (i) Swedish environmental law and/or environmental EU directives and regulations are applicable, (ii) regulations on environmental impact assessments are applicable, and (iii) potential other relevant Swedish laws are applicable.*

E.1 Introduction

23. Below is a description of Swedish legislation that have been identified as relevant to the Project. It should be noted that the description does not aim to be exhaustive such to include *all* Swedish laws that could to some extent be applicable to the Project. The focus has been environmental laws and laws related to space or aviation.

E.2 The Space Act

24. Given the operation of SSC, the Swedish Space Act (1982:693) (Sw. *Rymdlagen*) ("Space Act") and Space Ordinance (1982:1096) (Sw. *Rymdförordningen*) ("Space Ordinance") should be mentioned. According to section 1 of the Space Act, the act is only applicable on operations in *outer space*. Since SSC does not operate in outer space, the Space Act and the Space Ordinance are not applicable.

25. However, it should be noted that the Swedish government on 2 April 2020 issued a committee directive proposing a review of the Swedish legislation on space activities (Sw. *rymdverksamhet*), which means that a special investigator shall review the Space Act and, if necessary, the adjacent space regulation. The purpose is to achieve a long-term sustainable regulation of space activities in line with international regulations and national security and which creates predictable and favorable conditions for companies, universities and authorities within the space field.[7]

26. Initially, the review were to be submitted to the Swedish Parliament by no later than 1 June 2021, however, the deadline has been extended to 17 September 2021.[8] Once the review has completed, any suggestions will likely be subject to consultation process by which certain authorities and institutions may provide feedback. This means that any amendments to the regulation will not enter into force before the First Phase of the Project, which is expected to be carried out in June 2021.

SW41029167/4

---

[7] The investigator shall *inter alia* (i) provide an opinion if the Space Act should include conditions to obtain a permit for space activities, and whether or not the state should be included in such permit requirement, (ii) examine whether it is appropriate to introduce provisions in the Space Act on the protection of the space environment ant the prevention of the emerge of space debris (Sw. *rymdskrot*), (iii) in the review, take into account Sweden's foreign security and defense interests as well as political interests and obligations under international law, and (iv) assess whether protection of Sweden's security should be regulated in the Space Act.

[8] https://www.regeringen.se/pressmeddelanden/2020/09/rymdlagen-utreds--ny-utredare-blir-goran-lundahl/ (2021-02-04)

E.3 The Environmental Code

E.3.1 Environmental Impact Assessments

27. One of the main issues in respect of which we have been instructed to advice is if the Swedish regulation requires an environmental impact assessment for the First Phase. The main regulatory framework on Swedish Environmental law is the Environmental Code (1998:808) (Sw. *Miljöbalken*) ("the Code") and chapter 6 of the Code deals with environmental impact assessments. This chapter was recently revised with the purpose to further align with the provisions on environmental impact assessments of EU law and certain international conventions.[9] The revised version of chapter 6 entered into force in 2018.

28. The revised chapter 6 contains a clearer division of regulations on environmental impact assessments in respect of the planning and decisions on plans and programs (strategic environmental impact assessments) on the one hand, and actual operations and activities (specific environmental impact assessments) on the other hand.[10] The First Phase of the Project is not a plan nor a program according to the Code but could be considered as *an activity*. Section 20 of chapter 6 of the Code stipulates which operations and activities requiring environmental impact assessment: (i) for a permit according to chapter 7 section 28a (referring to section 27) of the Code (regarding wild birds[11] and certain habitat for wild animals and plants[12]), or (ii) for a permit referred to in chapter 9 (environmentally hazardous activities), or (iii) chapter 11 (water activities) or that requires such permissibility (Sw. *tillåtlighet*) as referred to in chapter 17 (regarding general navigable waterways (Sw. *allmänna farleder*), geological storage of carbon dioxide and facilities for nuclear activities, if the activity or measure can be assumed to have a significant environmental impact.

29. It is clear that the First Phase of the Project does not include (i) and (iii) above, but it should be further investigated if such a requirement could be raised in relation to (ii) above (environmentally hazardous activities) or if it in any other manner constitutes an activity with a significant environmental impact (Sw. *betydande miljöpåverkan*).

30. Given that the First Phase of the Project only includes the launch and deflate of a test balloon and gondola that will be airborne for approximately four to six hours[13], we do not consider that chapter 9 section 1 of the Code is applicable, and

SW41029167/4

---

[9] UNECE Convention on Environmental Impact Assessment in a Transboundary Context ("the Espoo Convention") (see Appendix 1, C) and its protocol, as well as the UNECE Convention on Access to Information, Public Participation in Decision-making and Access to Justice in Environmental Matters ("the Aarhus Convention") (see section Appendix 1, D).
[10] Government bill 2016/17:200 p. 61.
[11] Directive 2009/147/EC of 30 November 2009 on the conservation of wild birds.
[12] Directive 92/43/EEC of 21 May 1992 on the conservation of natural habitats and of wild fauna and flora.
[13] Harvard SCoPEx Balloon Statement of Work Summary AD-1 referred to in SSC Proposal.

in conclusion that the potential implementation of the First Phase does not constitute an operation or activity requiring environmental impact assessment according to section 20 of chapter 6 of the Code.

31. There are also activities which fall within the scope of the EIA directive without being subject to a permit requirement under the Code. Such operations and activities are regulated by section 26a of the Ordinance on Environmentally Hazardous Activities and Health Protection (1999:899) (Sw. *förordningen om miljöfarlig verksamhet och hälsoskydd*). For the activities subject to notification that are listed in this ordinance, the responsible authority shall assess whether the activity needs to be subject to a permit process. Such assessment shall be made on the basis of the operations' or activities' environmental impact and the criteria in section 10-13 of the Environmental Assessment Ordinance (2017:966) (Sw. *miljöbedömningsförordningen*). The criteria in mentioned sections seek to establish if an operation involves a significant environmental impact (Sw. *betydande miljöpåverkan*). For example, section 10 states that a decision should take into consideration the operations' or activities' distinctive features, location and the type and characteristics of the possible environmental effects. Moreover, section 11 states that as regards the distinctive features, the operations' or activities' scope and design as well as if it contributes to cumulative environmental effects together with other operations, should be considered.

32. After having reviewed the criteria in section 10-13 for an activity that constitutes a significant environmental impact, our conclusion is that the First Phase of the Project does not fulfil these criteria, including not constituting an activity with significant environmental impact. Thus, an environmental impact assessment is not required for the First Phase of the Project.

E.4 The Civil Aviation Act

33. The Code interacts with other laws. For example, section 7 of chapter 1 of the Code refers to the Civil Aviation Act (2010:500) (Sw. *luftfartslagen*) in respect of environmentally worthiness of the aircraft.[14] Following from section 1 and 4 of chapter 3 of the Civil Aviation Act, it may be required that an aircraft must be issued with a certificate of airworthiness and a certificate of compliance with environmental standards prior to being used for aviation. However, according to the Swedish Transport Agency, these licenses require that the balloon is registered in the aircraft register. Requirements for registration are primarily placed on other types of manned aircraft than balloons.[15] In light of this, it is not a requirement that Harvard obtain any of these licenses to use the balloon.

SW41029167/4

[15] Cf. Regulation (EU) 2018/1139 of the European Parliament and of the Council of 4 July 2018 on common rules in the field of civil aviation and establishing a European Union Aviation Safety Agency, the Civil Aviation Act, the ordinance (1986:172) regarding the aircraft register and TSFS 2016:40.

E.5 **EU regulations and international conventions regarding the environment ratified by Sweden**

34. Since 1995, Sweden is a member of the EU. As environmental matters are typically cross-border issues, Swedish environmental laws are, with few exceptions, strongly influenced by EU law. As a result, EU regulations is to a large extent implemented into and thus become part of Swedish law. However, an overview of certain EU regulations and international conventions ratified by Sweden with regards to, inter alia, environmental impact assessment, transboundary impact assessments, right to legal access etc., as well as the relevant implementations into Swedish law are described in Appendix 1.

E.6 **Conclusions**

35. The Space Act is not applicable on the First Phase of the Project. There is no requirement that Harvard obtain a license for the balloon and gondola according to the Civil Aviation Act and consequential legislation.

36. The Code is also not applicable on the First Phase of the Project, as there are no environmental impacts such as emissions to air or water. Moreover, the waste such as plastics from the parachute of the gondola or the balloon itself is limited and will be salvaged either by SSC or by partners of SSC. Accordingly, no waste from the First Phase triggers any particular requirements or obligations under the Code. Therefore, it is our conclusion that an environmental impact assessment is not required with regards to the First Phase of the Project.

F. Impact on the First Phase due to the potential implementation of the Second Phase

We have been instructed: *To address whether the potential implementation of the Second Phase of the SCoPEx project including release of material into the stratosphere would in any way affect the conclusions related to the questions in respect of the First Phase.*

37. Section E.3.1 describes under which circumstances an "operation or activity" requires an environmental impact assessment to be established. As there are no hazardous environmental consequences from the First Phase, we have concluded that such an environmental impact assessment is not required for the First Phase of the Project. Although it is noted in the preparatory works to the Code and in Swedish case law that an assessment should include *all parts* of the operation or activity, not only the part requiring a permit, such assessment is limited to the activity at hand.[16] This means that the "project as a whole" should be taken into account, which in our case is the whole of the First Phase.

38. This interpretation is in line with the principles in the Swedish Public Administration Act (2017:900) (Sw. *Förvaltningslagen*), the Administrative Procedure Act (1971:291) (Sw. *Förvaltningsprocesslagen*) and general principles of administrational process. For example, a trial or assessment can typically only cover what the applicant has applied for or intend to do. Only the applicant is in control of the application. Thus, the scope of the trial, application, notification or assessment is limited to the matter at hand.

39. Consequently, in relation to an application for a defined action, as one part of a planned operation, the possible forthcoming actions or further processes if not part of the application cannot be subject to the process as they fall outside the admissible scope. A consequence of this is that any opinions from the public or other authorities can only be considered if falling within the scope of what is being applied for or evaluated. Potential future actions cannot be considered.

40. In conclusion, since the First Phase and the Second Phase are separate independent phases, the First Phase will not be affected by the Second Phase. Accordingly, the assessment of applicable laws and regulations in the First Phase does not affect the fact that SCoPEx may include a later, separate Second Phase.[17]

SW41029167/4

[16] Government bill 2016/17:200 p. 195 and case no. MÖD 2007:50
[17] It should also be noted that the Agreement (between SSC and Harvard) relates only to First Phase.

Stockholm 18 February 2021

Håkan Fohlin   Tove Skärblom

Appendix 1:

Overview of certain EU regulations and international conventions ratified by Sweden

SW41029167/4

SETTERWALLS

Appendix 1

Overview of selected EU regulations and international conventions on the environment ratified by Sweden

A.	**Introduction**	**2**
B.	**Precautionary Principle**	**2**
C.	**Procedural regulations on environmental impact assessment**	**2**
C.1	Espoo Convention	2
C.2	Aarhus Convention	3
C.3	The International Court of Justice	4
D.	**Regulations on Environmental Impact Assessments**	**4**
E.	**Convention on Biological Diversity**	**5**
F.	**Summary**	**5**

SW41029167/4

SETTERWALLS

Introduction

1. The Project relates to solar radiation management ("**SRM**") and in particular stratospheric aerosol injection. SRM is not regulated by any specific international agreement or convention. Although Sweden and the EU are parties to a number of international treaties concerning geoengineering generally, none are dealing with SRM in particular. However, with regards to cross-border impact risks there are international agreements on environmental protection which could be applicable, for example in relation to air pollution control and species and habitat conservation, depending on the nature, size and location of the activities. [18]

2. Below is a summary of EU regulations and international treaties that could be relevant in relation to solar radiation management, with focus on environmental impact assessments. We have reviewed and considered these regulations and treaties in respect of First Phase and concluded that, in our opinion, none are applicable on First Phase.

B. Precautionary Principle

3. Pursuant to Article 191 of the Treaty on the Functioning of the European Union, Union policy on the environment *"is based on the precautionary principle and on the principles that preventive action should be taken ... Effects on the environment should be taken into account at the earliest possible stage in all the technical planning and decision-making processes."* The principle is also referred to in a number of international treaties as well as in Swedish national law.[19] It is common for environmental organizations to refer to the Precautionary Principle, not always as a legal but more of a universal principle to act cautiously.

4. The First Phase does not cause any environmental impact but is merely a test of the navigation system of the balloon. In our opinion there is no relevant environmental caution to be exercised and references to the precautionary principle is irrelevant.

C. Procedural regulations on environmental impact assessment

C.1 Espoo Convention

5. Sweden and EU have ratified the 1991 Espoo Convention on environmental impact assessment in a transboundary context (**"Espoo Convention"**), including its amendments and protocols.[20] The Espoo Convention recognizes that operations in one country can have environmental effects in other countries, and coop-

SW41029167/4

---

[18] Cf. Shepherd J et al. 2009 Geoengineering the climate: science, governance and uncertainty. London, UK: The Royal Society. Page 40. See https://royalsociety.org/topics-policy/publications/2009/geoengineering-climate/

[19] For example, the Rio Declaration and Chapter 2 section 3 of the Code.

[20] https://www.naturvardsverket.se/Miljoarbete-i-samhallet/EU-och-internationellt/Internationellt-miljoarbete/miljokonventioner/Esbokonventionen/ (2021-02-05)

eration around these issues is crucial. Operations that have significant environmental cross-border impact should, as much as possible, be avoided.[21] It also sets out the general obligation of states to notify and consult each other on all major projects under consideration that are likely to have significant adverse cross-border environmental effects. Under Swedish law, this is reflected in section 34 of chapter 6 of the Code.

6. In our opinion the First Phase does not have any environmental impact and as a result the Espoo Convention is not applicable.

C.2 Aarhus Convention

7. The 1998 Aarhus Convention on Access to Information, Public Participation and Decision-making and Access to Justice in Environmental Matters ("**Aarhus Convention**") has been ratified by EU and Sweden. As the title suggests, the Aarhus Convention grants rights to the public on access to environmental information, public participation in environmental decision-making and access to justice on matters concerning the local, national and transboundary environment. It focuses on relations between the public and state authorities.

8. Prior to the introduction of the Code, environmental organizations in Sweden had limited rights to appeal decisions unless they were immediately affected. With the Code entering into force in 1999, environmental organizations received extended rights to appeal decisions on environmental matters. This was essentially an implementation of article 9(2) of the Aarhus Convention into Swedish law.[22]

9. In the Code, this is reflected in section 13 of chapter 16, from which follows that environmental organizations have a right to appeal decisions on permits, permissions and exemptions issued under the Code. To be entitled to appeal, the organization must be a non-profit organization that has been active in Sweden for at least three years, have no less than 100 members (or that may otherwise prove it has public support) etc. In recent case law, environmental organizations have also to some extent been allowed to appeal permits, permissions and exemptions that have been issued under law other than the Code. These rights have been established under the Administrative Procedure Act (Sw. *Förvaltningslagen*), interpreted *in the light* of the Aarhus Convention. As a result, environmental organizations enjoy further extended rights to appeal environmental matters, for example decisions on concessions issued under the Swedish Electricity Act (1997:857) (Sw. *ellagen*).

10. At this point, the launch of the balloon and gondola is not subject to any law requiring a decision in respect of a permit for Harvard. Moreover, in our opinion the First Phase will not have any environmental impact. The potential implementation of the First Phase will not result in any decision by court or authority on which the Code or the Aarhus Convention will be applicable.

---

[21] Government bill 2016/17:200 p. 59.
[22] Government bill. 1997/98:45 s 488.

C.3 The International Court of Justice

11. The International Court of Justice ("**ICJ**"), to which all members of the UN are subject, has established by case law a requirement on states to carry out due diligence on projects with environmental cross-border impact.[23] This also applies in respect of SRM. The ICJ recognized that the accepted practice amongst states amounted to *"a requirement under general international law to undertake an environmental impact assessment where there is a risk that the proposed industrial activity may have a significant adverse impact in a transboundary context, in particular, on a shared resource."* It is argued that the ICJ judgment implies that states have a duty to notify and consult with potentially effected other states.[24]

12. Because First Phase in our opinion does not have any environmental impact, and even less so any cross-border impact, the ICJ case law is not applicable.

D. Regulations on Environmental Impact Assessments

13. With regards to environmental impact assessments for operations or activities, the main EU legislation is the EIA Directive[25]. The directive contains provisions to ensure a systematic assessment is carried out for projects which, due to their nature, size or location, entail a significant environmental impact.[26] This applies to a wide range of public and private projects, which are defined in Annexes I and II of the directive. All projects listed in Annex I are deemed having significant ef-fects on the environment and require an environmental impact assessment, for example long-distance railway lines, airports and installations for the disposal of hazardous waste. In Sweden this corresponds to activities that require a permit under Chapter 9 (environmentally hazardous activities), Chapter 11 (water activi-ties) and related sectorial legislation (Sw. *sektorslagstiftning*), for example roads and railways.

14. For projects listed in Annex II, it is up to the national authorities to decide whether an environmental impact assessment is required or not. This is done by a certain screening procedure, which determines the effects of projects on the basis of certain thresholds/criteria or on a case-by-case assessment. However, the national authorities must take into account the criteria laid down in Annex III.[27]

SW41029167/4

[23] ICJ Judgement of 20 April 2010 Pulp Mills on the River Uruguay (Argentina v. Uruguay)

[24] Bodansky D. 2019 Solar geoengineering and international law. In *Governance of the deployment of solar geoengineering* (eds RN Stavins, RC Stowe), page 121. Cambridge, MA: Harvard Project on Climate Agreements. See https://www.belfercenter.org/publication/governance-deployment-solar-geoengineering (2021-02-11).

[25] Directive 2011/92/EU of 13 December 2011. The first directive 85/337/EEC on environmental impact assessments entered into force in 1985.[25] This directive has been amended three times, in 1997, 2003 and 2009. The EIA Directive of 2011 is a codification of the directive of 1985 and its amendments. The EIA Directive has in turn been amended in 2014 by directive 2014/52/EU.

[26] The EIA Directive was aligned with the Espoo Convention through the amendment 97/11/EC of the EIA Directive, which broadened the scope of the EIA Directive by increasing the type of projects covered and the number of projects requiring mandatory environmental impact assessments.

[27] https://ec.europa.eu/environment/eia/eia-legalcontext.htm (2021-02-03)

15. The First Phase does not qualify as an operation or activity under any of Annex I, II or III of the EIA Directive. As there are no environmental impacts from the First Phase, the EIA Directive is in our opinion not applicable.

E. Convention on Biological Diversity

16. Of special interest is the 1992 Convention on Biological Diversity ("**CBD**") including Sweden and the EU, which in 2010 issued a non-binding decision[28], a moratorium, allowing exemptions for small scale scientific research studies[29];

"that no climate-related geo-engineering activities that may affect biodiversity take place, until there is an adequate scientific basis on which to justify such activities and appropriate consideration of the associated risks for the environment and biodiversity and associated social, economic and cultural impacts, with the exception of small scale scientific research studies that would be conducted in a controlled setting in accordance with Article 3[30] of the Convention, and only if they are justified by the need to gather specific scientific data and are subject to a thorough prior assessment of the potential impacts on the environment."[31]

17. As stated previously, in our opinion the CBD is not applicable on First Phase. However, even if the CBD would be applicable, there is an exception for small scale scientific research studies. The First Phase would qualify as such an exception.

F. Summary

18. There is no international agreement or treaty comparable to the UNCLOS that governs the atmosphere. States have sovereignty over the air space above their territory, from the ground to where the outer space commences.[32] Consequently, the injection of aerosols is subject to the jurisdiction and control of the state in whose air space it is injected into.[33]

19. However, the obligation not to cause significant transboundary harm is recognized in many international treaties, such as CBD, the Espoo Convention and also in Swedish national law. In general, states are not allowed to conduct or permit activities within their territory, or in common spaces such as the high

---

[28] COP 10 Decision X/33 and confirmed 2016 COP 13.

[29] The Project SCoPEx website www.keutschgroup.com/scopex, page 9, refers to that SCoPEx Second Phase does not violate CBD due to the small scale scientific test.

[30] Article 3 states that *"States have, in accordance with the Charter of the United Nations and the principles of international law, the sovereign right to exploit their own resources pursuant to their own environmental policies, and the responsibility to ensure that activities within their jurisdiction or control do not cause damage to the environment of other States or of areas beyond the limits of national jurisdiction."*

[31] It could be noted that the Swedish preparatory work SOU 2020:4 (January 2020) "The pathway to a climate-positive future – strategy and action plan for achieving negative greenhouse gas emissions after 2045" is not addressing SRM specifically but states on page 103: *"Sweden should work to ensure that the decision on the moratorium on geoengineering made at the Tenth meeting of the Conference of the Parties to the Convention on Biological Diversity in Nagoya is amended such that bio-CCS and other non-fossil CCS are not covered by the moratorium."*

[32] The precise point where this limit is reached is not entirely settled as a matter of law, https://royalsociety.org/topics-policy/publications/2009/geoengineering-climate/see Shepherd J et al. page 40. (2021-02-11)

[33] Shepherd J et al. page 40.

SW41029167/4

seas and up to outer space, without considering the interests of other states and the protection of the global environment. Consequently, states are obliged to exercise due diligence in regulating activities under their jurisdiction and control. If an operation or activity have cross-border implications, or is located beyond national jurisdiction (for example space-based techniques for reducing solar radiation) international cooperation on regulation will be necessary.[34]

20. However, the First Phase does not include any environmental harm and does not have any environmental cross-border impacts. Thus, our conclusion is that the regulations and conventions referred to in this appendix are not applicable to the First Phase of the Project.

SW41029167/4

[34] Shepherd J et al. Page 40.

Appendix C

Engineering and Safety Review Documents

Appendix C-1

Letter from Research Team Requesting Authorization for Platform Test

HARVARD
School of Engineering
and Applied Sciences

Frank N. Keutsch
Stonington Professor of Engineering and Atmospheric Sciences
keutsch@seas.harvard.edu

Harvard John A. Paulson School of Engineering and Applied Sciences 12 Oxford Street, Cambridge, MA 02138 USA **Department of Chemistry and Chemical Biology** Phone: 617-495-1878 / Fax: 617-495-4902 **Department of Earth and Planetary Sciences**

November 10, 2020

Dear SCoPEx Advisory Committee,

We hereby request that the advisory committee review our plans for a proposed platform test in June 2021. This test is not the experiment itself, but rather a test of the SCoPEx platform without release of any particles.

This balloon flight would be managed by Swedish Space Corporation (SSC), flying out of northern Sweden. SSC would provide balloon operations including launch, recovery, and safety management and would secure any necessary regulatory approvals.

As mentioned, the goal of this flight is to test the SCoPEx platform. Specifically, we would like to review the gondola's horizontal and vertical control using the ascender system and propellers as well as the power, data, navigation, and communication systems. We would not release any aerosols, nor fly an aerosol injection/release system.

Although there is no release of materials related to solar geoengineering, we will not proceed with this flight without a formal recommendation authorizing the flight from the advisory committee to Harvard management.

We expect to commit to a contract with SCC within the next two weeks, and that contract requires a significant upfront payment. Because of this, it would be very helpful for us to receive an indication from the committee regarding a schedule for reaching a decision on such authorization before we sign the contract. Specifically, could the advisory committee commit to completing a review and reaching a decision (be it positive or negative) about this platform test before the 15th of February 2021?

This would, of course, be contingent on the experiment team providing you with timely access to materials needed for the review.

Thanks for your engagement in this process.

Sincerely,

Frank Keutsch
Stonington Professor of Engineering and Atmospheric Science
Harvard John A. Paulson School of Engineering and Applied Sciences

Department of Chemistry and Chemical Biology
Department of Earth and Planetary Sciences

Appendix C-2

Research Plan Provided by Research Team

HARVARD
School of Engineering
and Applied Sciences

Frank N. Keutsch
Stonington Professor of Engineering and Atmospheric Science
Professor of Chemistry and Chemical Biology
keutsch@seas.harvard.edu

Harvard John A. Paulson School of Engineering and Applied Sciences
Department of Chemistry and Chemical Biology
Department of Earth and Planetary Sciences
12 Oxford Street, Cambridge, MA 02138 USA
Phone: 617-495-1878 / Fax: 617-495-4902

November 10, 2020

Dear SCoPEx Advisory Committee,

In response to a written request by the Advisory Committee, we submit a document for a review of the scientific merits of SCoPEx.

In this document, we first review why existing observations are incapable of addressing questions that SCoPEx will answer. We then give a description of the basic design of the SCoPEx platform and its concept of operations. Finally, we describe the three science goals of SCoPEx, explain how they represent knowledge gaps for stratospheric aerosol injection (SAI), and specify what measurements are needed to enable SCoPEx to provide quantitative answers to these questions.

We do not provide a detailed engineering description of the SCoPEx platform nor of its scientific instrumentation. Nor do we provide a general justification for research on solar radiation modification. Finally, we do not provide a risk management plan, as that plan will be managed in coordination with the balloon operator and will depend on the specifics of the flight location and plan.

We look forward to working with the Committee and will be happy to revise these documents and provide additional materials on request.

Sincerely,

Frank Keutsch
Stonington Professor of Engineering and Atmospheric Science
Harvard John A. Paulson School of Engineering and Applied Sciences
Department of Chemistry and Chemical Biology
Department of Earth and Planetary Sciences

The Stratospheric Controlled Perturbation Experiment (SCoPEx)

Version 1.0

Executive Summary.....2
1. Introduction.....3
2. Observational SAI Research Needs.....5
3. SCoPEx Short Overview.....11
4. SCoPEx Goals.....18
5. Data Management Plan and Dissemination of Results.....30
6. SCoPEx Research Team Biographies.....31
References.....34

Executive Summary

Climate model studies of stratospheric solar radiation modification (SRM) depend, perhaps implicitly, on processes that take place in the near field of an injection plume. This is because materials delivered to the stratosphere by aircraft will form persistent, high aspect-ratio plumes with strong gradients before becoming well mixed, and processes within the plume will alter the large-scale, well-mixed aerosol and chemical properties that are simulated in global atmospheric models. All models ultimately depend on observations, yet we lack experimental data to assess some of the critical transport, microphysical, and chemical processes that directly control aerosol dynamics in the near-field that are important for understanding stratospheric SRM.

The scientific goal of the Stratospheric Controlled Perturbation Experiment (SCoPEx) is to improve process models that will, in turn, reduce uncertainties in global-scale models, thus reducing uncertainty in predictions of important SRM risks and benefits.

SCoPEx addresses questions in stratospheric aerosol injection (SAI) research that observations of existing analogues are incapable of addressing. For example, existing observational data do not include chemistry of alternate geoengineering materials specific to SAI, near-field particle microphysics of injection plumes, and relevant scales of atmospheric transport in the near-field. Yet these are needed to assess processes that control aerosol dynamics in the near field of an injection plume and that allow for the evaluation of alternate SAI materials, i.e., materials other than the naturally existing sulfate aerosol.

We first review why existing observations do not address the questions that SCoPEx will answer. We then give a description of the basic design of the platform and the concept of operations of SCoPEx. Finally, we describe the three specific science goals of SCoPEx, explain how they represent critical knowledge gaps in SAI research, and specify what measurements are needed to enable SCoPEx to provide quantitative answers to these questions. The three specific science goals are improving understanding of (i) turbulent mixing scales, (ii) aerosol microphysics with a focus on alternative SAI materials in the near-field of an injection, and (iii) process level chemical interactions of alternative SAI materials in the stratosphere.

We do not provide a detailed engineering document of the SCoPEx platform or its scientific instrumentation, nor do we provide a justification for the need for research on SRM via SAI in general. Rather, we focus specifically on the merits of SCoPEx itself.

1. Introduction

In this document we focus on the motivation and scientific merit of SCoPEx. We do not provide detailed engineering documentation of the SCoPEx platform or its scientific instrumentation. We also do not provide general justification for the need for research on solar radiation modification (SRM) via stratospheric aerosol injection (SAI), which can be found in many prior documents such as the 1992 NAS report that recommended the US government "Undertake research and development projects to improve our understanding of both the potential of geoengineering options to offset global warming and their possible side effects. This is not a recommendation that geoengineering options be undertaken at this time, but rather that we learn more about their likely advantages and disadvantages" (National Academy of Sciences et al., 1992) or the recent 2015 NAS report (National Research Council, 2015). Rather, we focus specifically on the need for small-scale field experiments such as SCoPEx, and the specific, critical SAI research needs that will be addressed by SCoPEx.

1.1. Role of and Need for Small-Scale Field Experiments

There is a vast array of science and engineering questions that have to be answered to achieve a better understanding of the risks, benefits and feasibility of SAI. The tools and topics that are needed to address these questions range from General Circulation Models (GCMs) all the way to detailed design of instrumentation to monitor or disperse aerosol. SCoPEx addresses a subset of questions that require small-scale field experiments for ground-truthing and that are aimed at improving the ability of models to predict the consequences of SAI.

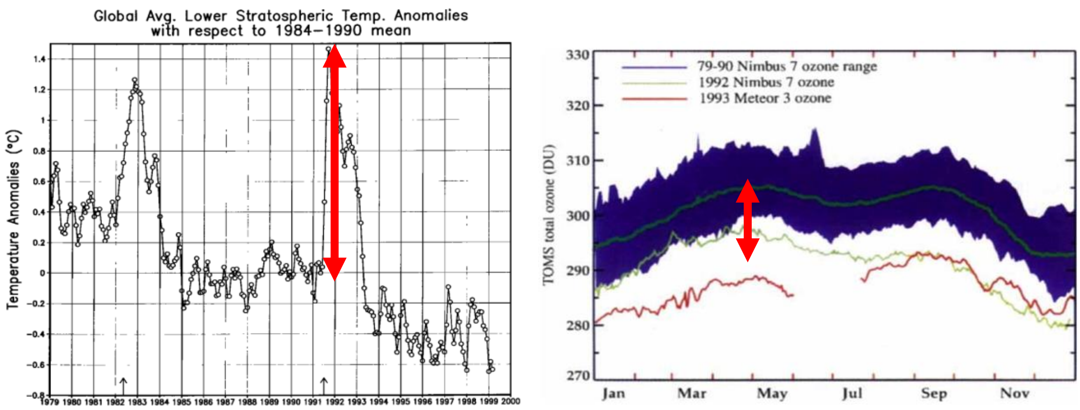


Figure 1: The two most important first-order stratospheric risks from sulfate SAI. The left panel shows stratospheric temperature anomalies from the El Chichon and Mount Pinatubo eruptions on top of background temperatures that are decreasing due to greenhouse gas emissions (Robock, 2000). The dynamical response of the stratosphere from such a short heating pulse likely is different than from sustained heating from longer-term SAI. The right panel shows that in the two years following the Mount Pinatubo reaction total ozone columns were lower than in the 1979-90 average as a result of increase sulfate aerosol surface area. Smaller eruptions also contributed to this. (McCormick et al., 1995)

There are numerous known risks associated with SAI, and SCoPEx focuses primarily on improving understanding of the first-order impacts in the stratosphere, i.e., risks and risk reduction associated with impacts of SAI within the stratosphere. There are many downstream / higher-order risks, e.g., impact on cloud formation as SAI particles leave the stratosphere (Cziczo et al., 2019), impacts on ecosystems via changes in the hydrological cycle (Bala et al., 2008; Russell et al., 2012; Tilmes et al., 2013), or the amount of direct versus diffuse radiation (Gu et al., 2002; Farquhar & Roderick, 2003; Gu et al., 2003). Despite their importance, these impacts are not the direct target of this proposal although many of these are also influenced by stratospheric processes and properties of SAI aerosol. Two first-order risks are at the focus of this work: stratospheric ozone loss and the dynamic response resulting from stratospheric heating as a result of SAI.

Whereas stratospheric ozone chemistry is fairly well understood (World Meteorological Organization, 2019), there are still substantial uncertainties in the understanding and ability to model stratospheric dynamics (Figure 1). For example, models have only recently been able to reproduce the quasi-biennial oscillation without having it imposed (see Butchart et al., 2018 for a discussion of challenges). One approach taken in this work is to evaluate whether there are types of aerosols or methods of aerosol injection that can reduce first-order risks for a given amount of radiative forcing. It stands to reason that a reduction in the first-order stratospheric impacts will reduce downstream and higher-order risks. A case in point is the growing body of work that has been investigating the impacts of stratospheric heating on stratospheric water vapor and the dynamic response on regional climate (Simpson et al., 2019; Ferraro et al., 2015; Richter et al., 2018; Ji et al., 2018). It is important to note that the amount of stratospheric heating for a given material will be primarily driven by the total mass of aerosol, ozone destruction will be driven by the total surface area of aerosol, and the desired radiative forcing will be determined by the amount and size distribution of aerosol. Critically, both the aerosol mass required for a given desired radiative forcing *and* the resulting surface area are tied to this size distribution. Therefore, accurate models of the evolution of the size distribution of injected aerosol are critically needed. In addition, alternate materials with reduced stratospheric heating have to be investigated, as do injection methods for sulfate that minimize stratospheric heating and ozone loss for a given radiative forcing, as this will reduce risks associated with the dynamic response to this first-order perturbation.

2. Observational SAI Research Needs

Most of the rapidly growing body of literature on SAI rests on General Circulation Models (GCMs). We acknowledge the importance of GCM studies, but in the following we focus on research needs that require experiments and observations, and especially questions that can only be answered by conducting perturbative field experiments such as SCoPEx (see supplemental manuscripts Keith et al., 2020 and Floerchinger et al., 2020). In fact, SCoPEx will in the end inform GCMs by providing improved process level information that will be integrated in parameterizations used in GCMs. Below we review existing observational data sets and describe their utility for different SAI approaches, highlighting where they are unable to shed light on critical issues thus motivating studies like SCoPEx.

2.1. Field Experimental Needs for Sulfate SAI

Most studies that have sought to research SAI have assumed the addition of aerosol would take place by means of an injection of gas-phase SO_2, which is ultimately converted to H_2SO_4 and then to sulfate aerosol in the stratosphere on a timescale of approximately one month. The aerosol size distribution from this injection of gas phase precursor must be accurately predicted as it will control the shortwave (SW) scattering properties, the stratospheric lifetime of the aerosol, and ultimately be the driver for the radiative forcing (RF) efficiency per mass of injected sulfate. Some studies, such as Niemeier & Timmreck (2015), have suggested that with higher injection rates of SO_2, the resulting sulfate aerosol would be forced into a larger, coarse-mode size distribution and functionally reach a point of diminishing return. In this diminishing return scenario, the added amount of SW RF achieved per added mass of sulfate decreases exponentially.

Recent work by Pierce et al. (2010), Benduhn et al. (2016), and Vattioni et al. (2019) has highlighted the potential benefits of injecting H_2SO_4 aerosol directly into the accumulation mode (AM), i.e., aerosols with a radius of 0.1–1.0 µm, potentially by emitting H_2SO_4 vapor into an aircraft plume. This work has suggested better control of the resulting aerosol size distribution and thus the radiative forcing per unit mass sulfur injection, which would allow for the design of a system that maximizes the radiative forcing per mass of sulfate in a way that would not have the diminishing returns at high SO_2 injection rates. This would thus minimize the increase in the stratospheric sulfate burden and hence the risk of stratospheric heating which is driven by total mass whereas ozone loss is driven by surface area. While injecting AM–H_2SO_4 may represent the best possible approach for SAI with stratospheric sulfate, there is currently no proven way to introduce vapor phase AM–H_2SO_4 into the stratosphere. As AM–H_2SO_4 has not been studied, perturbative experiments are required to provide observational constraints on the aerosol size distributions predicted by models.

2.2. Field Experimental Needs for Alternate Aerosol Material SAI

Though sulfate aerosol does exist in the background stratosphere and there are some natural analogs of broad stratospheric sulfate injections (volcanic eruptions), it likely is not the optimal candidate for SAI. Alternative aerosol may be most appropriate in order to mitigate SAI risks (Teller et al., 1996; Crutzen, 2006; Ferraro et al., 2011; Ferraro et al., 2015; Weisenstein et al., 2015; Keith et al., 2016; Dykema et al., 2016; Weisenstein et al., 2015). These alternate aerosols could reduce the previously noted two major first-order stratospheric impacts, i.e., changes in ozone and stratospheric heating. Due to the uncertainties in the impacts of stratospheric heating, the study of materials with optical properties that negate stratospheric heating is especially important. Materials such as calcium carbonate ($CaCO_3$), alumina (Al_2O_3), diamond (carbon), and several others, have been proposed as a way to minimize the inherent risks from SAI (Keith et al., 2016; Dykema et al., 2016; Weisenstein et al., 2015; Ferraro et al., 2015; Ferraro et al., 2011; Crutzen, 2006). Although model results of these aerosol species suggest that some of them possess optical properties that make them well suited to be used in a SRM scenario ($CaCO_3$, Al_2O_3, and diamond) (Dykema et al., 2016; Ferraro et al., 2011), the stratospheric aerosol microphysics of these compounds (especially coagulation) is poorly understood. As with AM–H_2SO_4 injections, there is a profound lack of in situ data to assess the ability to model the microphysics of alternative aerosols and the stratospheric chemistry of these materials. This is especially pertinent with respect to changes in ozone, and is exacerbated by the fact that these aerosols have no naturally existing analog in the stratosphere that could be studied. Because early studies suggest that these aerosols show much promise with respect to deploying SAI while mitigating the inherent risks of the deployment, it is imperative to design and execute in situ experiments in order to test our current understanding of the aerosol microphysics and observe the effects of alternative aerosol on the chemical composition and dynamics of the stratosphere.

2.3. Limitations in Existing Analogues

In this section we will review previous in situ studies of stratospheric plume processes, show how those datasets have contributed to our current understanding, and demonstrate the need for experiments such as SCoPEx to inform small-scale models of aerosol microphysics (nucleation and coagulation), plume transport and physical morphology, and chemical properties of new aerosol species that have thus far not been observed in the stratosphere. Because the nature of the injection scenarios (AM–H_2SO_4 or solid aerosols) are so complex compared to natural analogs, new experiments must be designed and implemented to provide observational constraints on our current nearfield modeling framework. Experimental data from carefully targeted small-scale studies would contribute to the development of nearfield-scale models that represent currently uncertain processes in detail.

We note that sub-grid scale processes do not represent the only unknowns in GCMs that are relevant to high-fidelity simulations of SRM scenarios, and that there are many large scale model phenomena which should be further assessed with observational evidence. However, here we focus on the need for in situ data to constrain sub-grid scale processes that can be addressed by SCoPEx and highlight the need for reducing the uncertainty in transport and aerosol dynamics and chemistry at this scale.

2.3.1. Limitations of Solid Rocket Motor Plume Observations

From 1996 to 2000 a number of rocket plumes were observed by high-altitude research aircraft. Generally, these missions involved a research team coordinating stratospheric sampling flights on either the NASA ER-2 or on the NASA WB-57 with coincident rocket launch events from either Cape Canaveral or Vandenberg Airforce Base. These studies sampled plumes from a host of rocket types including Titan IV, Space Shuttle (STS106, STS83, STS85), Delta II, Athena II, and Atlas IIAS.

Plumes were intercepted by the sampling aircraft between 5 and 125 minutes after emission from the rocket motor at stratospheric altitudes ranging from 11 to 19.8km (Voigt et al., 2013). The main science objective of these missions was to assess the stratospheric ozone depletion potential of space exploration by understanding the halogen chemistry occurring as a result of the high-altitude rocket burn. However, in studying the effects on the ozone layer, this era of stratospheric sampling provided a unique set of plume measurements to study nearfield processes of chemical injections into the stratosphere.

While measuring the plumes from the Titan IV rocket (as a part of the United States Airforce Rocket Impacts on Stratospheric Ozone (RISO) Campaign) and attempting to develop a plume chemistry model to solve for the Cl_2 concentration in a rocket plume as it evolves shortly after its emission, Ross et al. (1997) noted the many assumptions that had to be made about the plume morphology in order to simulate the mixing and diffusion that the rocket plume had with the surrounding stratosphere. Their model solved for the Cl_2 concentration of a circular nighttime plume as it expanded in diameter along an isentropic surface. Subsequent aircraft measurements showed that plumes contained more than twice the predicted concentration of Cl_2 despite the plume being intercepted during the day time (when the Cl_2 reservoir should be somewhat depleted by the photolysis reaction $Cl_2 + h\nu \rightarrow 2Cl$), suggesting that there may be an error in the assumption of a circular plume morphology on the short transport time scales observed in this study ($\sim 28 min$).

Ross went on to publish a second study as a part of the RISO project in 1999, this time looking to quantify the size distribution of alumina aerosols emitted from the rocket engines which contained particulate alumina (Al_2O_3) (Ross et al.,1999). They compared measured aerosol size distributions from the WB-57F plume interceptions to results from an aerosol coagulation model and highlighted a massive discrepancy. The model predicted a much smaller aerosol size distribution with 1-10% of the aerosol mass being in the smallest (0.005μm) mode and the aircraft observed only fractions (<0.05%) of the model estimate in that same small mode. At the same time, over 99% of the aerosol mass sampled by the aircraft was found in the coarsest mode (2 μm), which the model was unable to predict. It is most likely that the model used in Ross et al. (1999) did not well account for the effects of ion mediated nucleation as described by Yu & Turco (1997). However, the data from Ross et al. (1999) was some of the first in situ data to highlight the uncertainty in stratospheric aerosol coagulation models. Alumina aerosol, as well as other solid aerosols, in contrast to liquid sulfate aerosol, have since been investigated as a candidate for use in SAI (Weisenstein et al., 2015). Therefore, it is imperative that we understand the chemical, coagulation, and accumulation properties of these and other solid aerosols in a stratospheric environment.

2.3.2. Limitations of Previous Stratospheric Aircraft Wake Crossing Observations

We can look to the few times high-altitude aircraft wake plumes have been sampled in situ for another example of stratospheric plume measurements. In the early 1990s the popularity and capability of the Concorde spurred discussions of a large fleet of High Speed Civil Transport (HSCT) aircraft that would operate in the lower stratosphere between 16 and 23 km. Scientists became concerned with the effects of high-altitude aircraft and high-altitude supersonic aircraft on stratospheric ozone destruction via the creation of a large NOx source in the lower stratosphere. NASA then launched several field campaigns using the ER-2 to study the exhaust profiles of high-altitude aircraft. In 1992 NASA commissioned the Stratospheric Photochemistry Aerosols and Dynamics Expedition (SPADE) to look at the effects of HSCTs. As a part of SPADE the ER2 sampled its own plume on several occasions by making a hairpin turn and heading into its original path, therefore measuring its own wake

(Figure 2). SPADE resulted in at least 11 published studies and some of these can inform us about the mixing and aerosol dynamics that may be relevant to an SAI scenario (Stolarski & Wesoky, 1993).

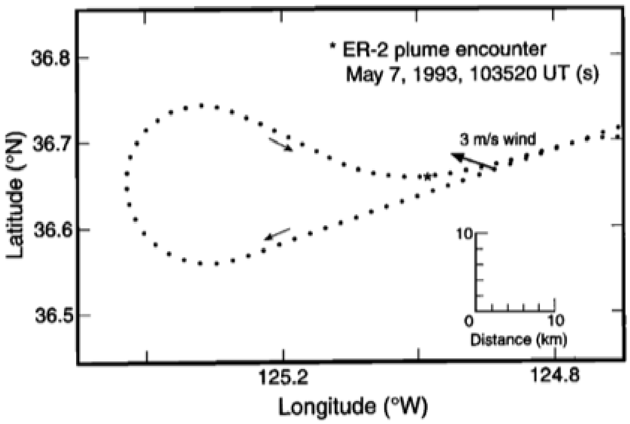


Figure 2: Shows the ER-2 flight track on a typical wake crossing trajectory (adapted from Fahey et al. 1995).

Fahey et al. (1995a) described measurements made of condensation nuclei (CN) present in the ER-2's exhaust plume from the emission of aerosol carbon and of sulfur compounds during one of its SPADE wake crossing events. Because the main focus of this study was to quantify the emission indices (EIs) of various compounds measured by the ER-2 that may have ozone depletion implications, they focused mainly on gas phase compounds. However, for the three wake crossings that the study focused on, they observed large variability in their EI measurements for CN. They noted that this is likely due to differences in mixing history of the encountered air parcels and noted that a full explanation of CN coagulation required more in-depth study and further measurements (Fahey et al, 1995b).

In another study published by Fahey et al. (1995b), they used a similar wake crossing technique to measure the exhaust of the Concorde aircraft and developed an aerosol coagulation model to predict particle formation and size as a function of the time since emission from the aircraft. The coagulation model was initialized at the observed conditions from the one-hour old Concord transect. The results from this model estimated that from 0 to 10 hr since emission from the engine, the mean particle diameter remained fairly constant at 0.06 μm before growing exponentially to a factor of 3 times its initial value over the next 1,000hr. The model predicted exponential mean particle diameter growth continuing right until the of the simulation at 1,000 hr (Fahey et al., 1995a).

Yu & Turco (1997) attempted to model the observed aerosol plume during the Concorde wake crossings with the goal of determining the driving factor for the large aerosol size distributions observed by the ER-2 in the exhaust which had not yet been explained by models. Yu proposed that aerosol formation was being aided by ion-mediated nucleation (IMN), that is, charged particles formed by chemi-ionization processes within the aircraft engines provide charged centers (H_2SO_4 [S(VI)]) around which molecular clusters rapidly coalesce. "The resulting charged micro-particles exhibit enhanced growth due to condensation and coagulation aided by electrostatic effects" (Yu & Turco, 1997). It is likely that IMN is the reason previous particle coagulation modeling of solid rocket motor plumes had overestimated the amount of aerosol in the small size ranges when compared to the in situ data, though this has not since been tested. Because of these effects, and the fact that specific size distributions of aerosol are desired to obtain the optimal radiative forcing effects for SAI (nominally smaller than observed in rocket or aircraft plumes), we must understand the aerosol nucleation and coagulation dynamics in an unperturbed stratosphere.

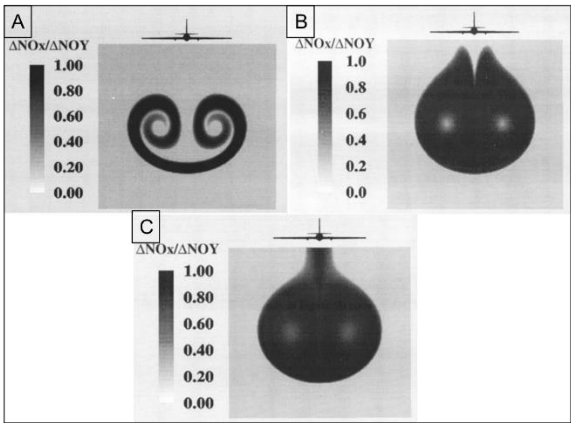


Figure 3: Shows the chemical and morphological evolution of an ER-2 plume during SPADE at 1.7 km (A), 4.8 km (B), and 7.9 km (C). (adapted from Anderson et al. (1996))

As a part of the SPADE project, Anderson et al. (1996) computed the flow field and chemical kinetics of the ER-2 aircraft exhaust using the Aerodyne Research Inc. UNIWAKE model. Their calculations address the effects of complex plume morphology on in-plume chemistry as a function of dilution time since emission from the aircraft engine. They showed that the plume morphology is highly variable out to about 5 km post emission Figure 3 and estimated that the stability of the wing vortex pair begins to break up at roughly 20 km post emission. Although this study was completed in the mid 1990s, it is still one of the only studies that attempts to compute nearfield chemistry within a dynamic stratospheric plume. However, particles were not considered as part of this study.

2.3.3. Limitations of Stratospheric Wake Crossings

Previous stratospheric plume studies of solid rocket motors and aircraft wake crossings have laid the foundation for our understanding of stratospheric plume chemical, aerosol, and mixing dynamics on transport scales of 0→100 km. These studies highlight the types of processes we must be aware of when considering the logistics of SAI. However, the violent initial conditions of engine exhaust plumes (such as temperatures of 700K, IMN) make it difficult to relate these observations to other systems. Because the engines drive the mixing and transport in the nearfield, and the ionic injection conditions of the plume create electrostatic forces that introduce complex nucleation affinities (IMN), understanding individual parameters can become analogous to finding a needle in a haystack. Moreover, because the radiative properties of any stratospheric aerosol that may be used for SRM depend on the diameter of the particle, we must understand the coagulation of that aerosol in the nearfield after the injection, which means that we must also understand the plume morphology that dictates the concentrations of that aerosol. Currently there have been no in situ data gathered that help us understand nearfield aerosol nucleation and plume dynamics in the absence of a very disruptive source. These conditions are necessary to understand as SAI may require that we mitigate the effect of IMN in order to obtain an aerosol size distribution that is small enough to provide the desired radiative properties.

2.3.4. Limitations of Naturally Occurring Analogs

Another source of useful in situ data on plume dynamics in the stratosphere can be found in literature addressing the fate and transport of convective overshooting events that often occur at the top of a Mesoscale Convective Complex (MCC). These events drive brief airmass exchange with the troposphere and often end up resulting in a plume-like parcel of tropospheric air being injected into the stratosphere.

Measurements of convective systems and upper troposphere-lower stratosphere exchange, as a means to interrogate stratospheric plume transport, have provided valuable in situ datasets that help us understand mid-field (10 to >1000 km) plume dynamics in the lower stratosphere. Similar to convective overshooting events, volcanic eruptions have provided an immense amount of in situ data that has informed us about regional and even global transport of stratospheric injections (Robock, 2000). Although their data are applicable in some sense to the transport of an SAI plume after its initial injection, the turbulent nature of a convective storm makes it difficult to measure these events at points near their injection source. Additionally, the storm conditions themselves dramatically complicate the system in the lower stratosphere such that is difficult to see through the effects of the induced turbulence in the nearfield. Indeed, an important limitation of these type of natural analogs is the spatial extent of their perturbation, which does not allow for near-field observations analogous to that of a point source. This also arises from the violent nature of these events which does not allow airborne platforms, such as the ER-2, to sample the initial conditions of the injection. We also note that volcanic eruptions are limited in their utility to evaluate dynamic response to stratospheric heating from sulfate aerosol, as they represent a perturbative pulse rather than the long-term heating one would expect from SAI.

In addition, these natural analogues provide extremely limited ability to study alternate materials, although organic and mineral dust aerosol injections into the lowermost stratosphere have been documented from convective overshoots. However, the complexity of the massive perturbations of both gas- and particle-phase preclude a study focusing on the impact on stratospheric composition and aerosol evolution that would result from SAI of a single material.

3. SCoPEx Short Overview

This section provides a brief overview of the engineering and operational aspects of SCoPEx. We first describe the platform, the instruments, and the concept of operations before describing the rationale for the overall SCoPEx design choices.

3.1. SCoPEx Platform

The SCoPEx gondola (Figure 4) is a balloon-born new research platform being developed at Harvard by the engineering and science staff within the Anderson/Keith/Keutsch laboratory group. The development builds on four decades of stratospheric research on aircraft, balloon, and rocket platforms that has focused on understanding the environmental chemistry of the ozone layer. The SCoPEx experiment was first described by Dykema et al. (2014). While many details of the design have changed, that paper still succinctly describes the advantages of choosing a balloon born platform over an aircraft, particularly for studying perturbations like solar geoengineering, and several of the limits of laboratory experiments that that could be addressed in a perturbative experiment like SCoPEx.

The gondola has three primary features: the frame, the ascender, and the propellers. The aluminum and carbon fiber frame contains two decks and a ballast hopper for coarse altitude control. One deck is primarily dedicated to platform support (power and flight control) and one deck is primarily dedicated to instruments. At the top of the gondola is an ascender and rope which allows the distance between the bottom of the balloon train and the gondola to vary from 0 to 150 m, which provides fine altitude control of the gondola. The ascender has been developed and tested by Atlas (Chelmsford, MA) building on their previous hardware in collaboration with the Harvard engineering team. The propellers serve two purposes: to create a well-mixed volume of air where observations of the aerosols and perturbed gas-phase can be made, and to reposition the gondola within the evolving aerosol plume. While the trajectory of the balloon and gondola system will be dictated by the balloon, the propellers allow for repositioning relative to the prevailing winds.

The ascender makes it impossible to have cables and other physical connections between the flight operations equipment and the gondola. Thus, the platform will handle its own communications and power. The SCoPEx platform will be powered using 28 V and 100 V DC power supplies which will power all operations on the platform including the propellers, ascender, and instruments. Elements of the flight platform are listed in Table 1. The gondola flight, flight safety, recovery parachute, and recovery operations will be managed by the balloon operator (in contrast to the SCoPEx team itself). Because the absolute velocity and distance capability of the gondola are so small compared to balloon drift, the trajectory will be determined by the balloon operator as if it was a passive nonpowered payload. During operations, the detailed float altitude will be jointly managed by the balloon operator via control of the balloon vents and the Harvard team via control of the ballast and ascender.

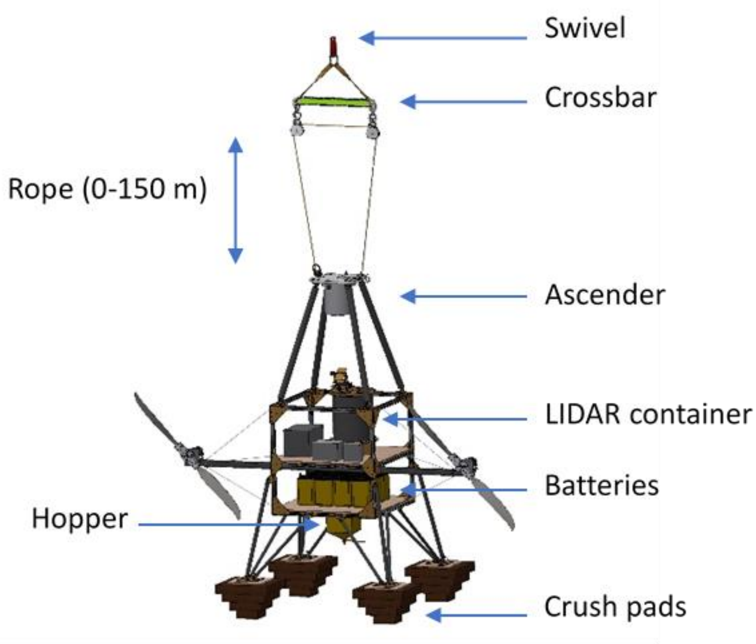


Figure 4: A representation of the SCoPEx flight platform. The final configuration may have subsystems packaged differently.

Parameter	**Description**
Total mass (Frame, all subsystems, hopper with ballast)	600 kg
Interface to balloon	Crosby 5-S-2 jaw & jaw swivel
Ascender	13 mm diameter rope Range of motion: 0-150 m Max speed: 10 m/min
Gondola propulsion	Twin propellers, 1.88 m diameter 32 N thrust each Max airspeed: 3 m/s
Power	28 V and 100 V DC power supplies with 24 MJ and 10 MJ total energy when fully charged
Communications	Satellite phone for communication between ground equipment and payload
Maximum termination shock	10 g

Table 1: Elements of the SCoPEx flight platform.

3.2. Instruments for First Science Flights (Science Goals 1 and 2)

The proposed instruments for the first science flight, addressing science Goals 1 and 2, are listed in Table 2. The corresponding science goals that motivate their inclusion are detailed in Section 4.

Measurement	Instruments	Rationale	Corresponding Science Goal
Wind speed measurement	Wind pendulum	Gondola and plume movement relative to balloon	Platform operation
Meteorology	Commercial off-the-shelf instrument	Temperature and pressure measurement throughout the flight	1, 2, 3
Wind turbulence	Constant temperature anemometer	Stratospheric mixing and modeling evolution of aerosol size distribution	1, 2
Particle dispersal	Solid Aerosolizer	Injects monodispersed particles for measurement and study	2, 3
Plume tracking	LIDAR	Tracking plume and navigation back into plume	2, 3
Particle sizer	POPS	Aerosol size distribution measurement for comparison with microphysics models of near-field evolution	2, 3
Light Scattering	Radiometer	Comparison of aerosol scattering with model prediction	2

Table 2: Instruments for first SCoPEx science flight.

Wind Pendulum: Understanding differential wind speed measurements between the balloon and payload will be important for plume evolution relative to the balloon trajectory and navigating the payload back into the plume. Commercial equipment to measure wind speed is typically not designed for the low densities found in the stratosphere. SCoPEx will therefore use a pendulum-based instrument and model to extract wind speed measurements. A camera will track a pendulum bob with high surface area and low mass, light enough to be perturbed by low winds in the stratosphere. Using the location and tilt data from the payload and a 3-dimentail kinetic model, the wind speed will be extracted from photos of the pendulum bob.

Commercial Meteorology Instrument: Commercial off-the-shelf instruments will be used for meteorological measurements on SCoPEx. They will record pressures and temperatures of the ambient stratosphere.

Constant Temperature Anemometer: A constant temperature anemometer (CTA) uses convective cooling caused by air flowing across a heated thin wire to measure flow velocity. LITOS (Leibniz-Institute Turbulence Observations in the Stratosphere) (Gerding et al., 2009; Theuerkauf et al., 2010) used such a measurement to study stratospheric turbulence up to 29 km. LITOS consisted of a 5 μm diameter and 1.25 mm long tungsten wire CTA and a 16 bit ADC with 2000 samples per second to collect measurements with a vertical resolution of 2.5 mm at 5 m/s ascent speed. The anemometer data was analyzed by performing a spectral analysis on the voltage signal to retrieve the spectral slope of the observed variation. A similar instrument will be used on SCoPEx to measure stratospheric turbulence. Air flow around the device will be simulated using CFD tools. The CFD runs will provide a means to identify key flow characteristics that drive sensor performance (sensitivity and accuracy), and to drive detailed sensor design.

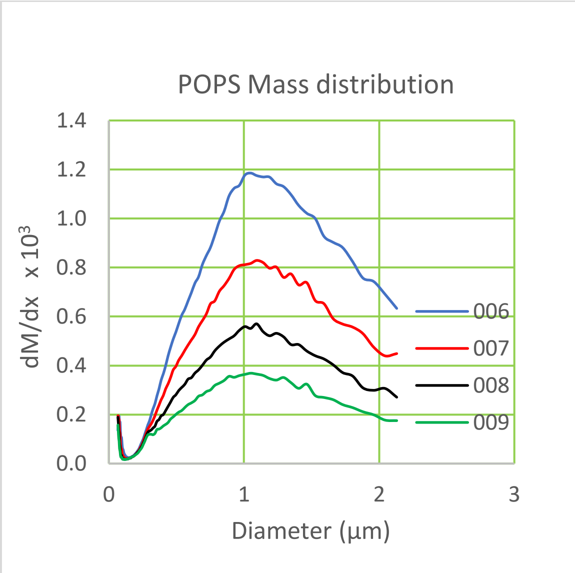


Figure 5: Successive measurements of sprayed $CaCO_3$ using an optical particle spectrometer. 006-009 indicate numbered time intervals spaced 4 minutes apart with 006 being the earliest measurement. $CaCO_3$ was sprayed using a 200 µm nozzle. In this laboratory experiment there was no significant variation in the shape of the distribution over time. (personal communication A Neukermans and team)

Solid Aerosolizer: The solid particle aerosolizer has been developed by a team lead by Armand Neukermans. For SCoPEx, the goal is to spray roughly monodisperse ~0.5 µm diameter precipitated calcium carbonate powder, the first candidate for solid SAI, through a 1-2 mm nozzle using the expansion of powder suspended in high pressure liquid CO_2. The aerosolizer would use a 1:4 weight ratio of $CaCO_3$ to CO_2. For 1 kg of $CaCO_3$ this would require a 5-7 L pressurized container. This concept has already been demonstrated in the lab. Figure 5 shows successive measurements of sprayed $CaCO_3$ with a size distribution centered at 1 µm diameter. Measurements were taken every 4 minutes using POPS (see below). In this case, total particle count decreased over time but there was no significant variation in the shape of the size distribution.

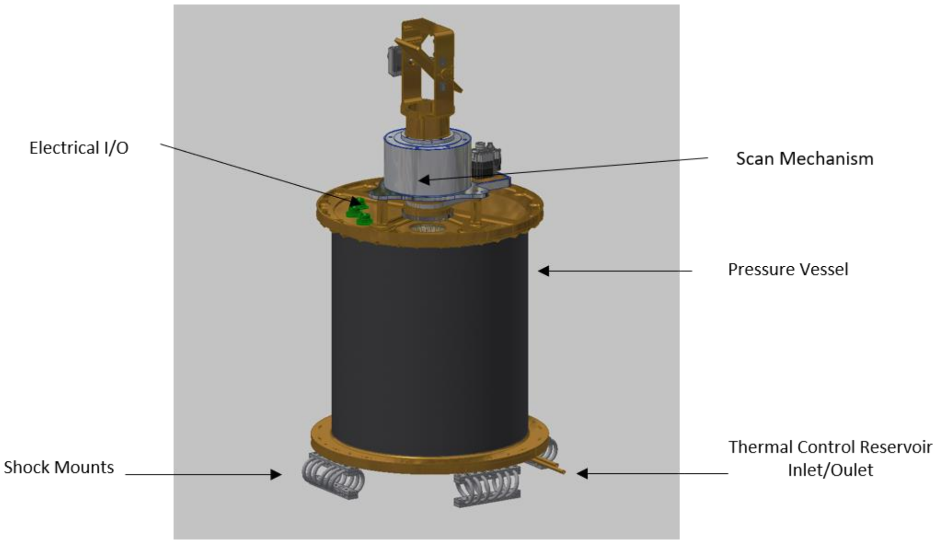


Figure 6: LIDAR pressure vessel provides safe storage and operating environment and support equipment.

LIDAR: The LIDAR is used to track the plume and allow navigation back into it. The core of the LIDAR system is an off-the-shelf eye-safe visible LIDAR, purchased from Sigma Space (now owned and operated by Droplet Measurement Technologies). This LIDAR produces 4 µJ pulses of 532 nm light at a repetition rate of 532 nm. The light that is backscattered by molecules and aerosols is collected by an 80 mm telescope and detected with a high-speed, high-sensitivity photodiode.

We have integrated this LIDAR in a pressure vessel (Figure 6) to provide a near-1 atm pressure environment with adequate temperature stability to ensure safe operation of the LIDAR at float altitude and safe storage on launch, ascent, descent, and recovery. This pressure vessel includes equipment for electrical and mechanical support, including command, data handling, and shock mounting. The LIDAR requires a scan capability to search the nearby atmosphere for the extent and geometry of the plume. The tilt and pan functions of the scan capability allows the LIDAR to be scanned over a set of angles that define the plausible location of the plume.

Portable Optical Particle Spectrometer (POPS): The POPS instrument will provide the aerosol size distribution measurements for studying aerosol formation and agglomeration. POPS is a light-weight instrument that directly samples the aerosol. It was built by and provided to SCoPEx through a collaboration with NOAA. The particles are illuminated with a 405 nm diode laser and the scattered light is collected onto a photomultiplier tube. The particle size is determined by the intensity of the scattered light. It has both the detection limit and size range (0.13 – 3 µm) to measure background stratospheric aerosol, which is more than sufficient for SCoPEx needs (Gao et al., 2016).

The Keutsch Group has already developed and extensively characterized a POPS instrument in preparation for the NASA-EVS3 Dynamics and Chemistry of the Summer Stratosphere field campaign on board the NASA-ER2, for which Keutsch is the deputy-PI. The POPS instrument tests include extensive thermal vacuum chamber characterizations to ensure operation under harsh stratospheric conditions. Compared to the ER-2, operation for SCoPEx will be simpler due to the insignificant air speed of the balloon and a much simpler operational pressure regime (on the ER-2 there is a large range of external pressures for both sampling and exhaust).

Radiometer: The aerosol plume can also be detected using a narrowband, narrow field of view radiometer with azimuthal/zenith pointing capability. The relationship between measurements of scattered solar radiation and the physical characteristics of atmospheric aerosols has been studied for more than two decades. Sky scanning measurements at multiple wavelengths between 300 nm and 1200 nm have been obtained using robotically pointed ground-based spectral radiometers deployed worldwide (Holben et al., 1998). The theory of these measurements has been refined and validated as a function of viewing geometry to provide a strong basis for inferring aerosol microphysics from radiometer data (Torres et al., 2014). The success of these approaches has motivated the development of compact sky scanning radiometers suitable for deployment on unsteady platforms like unmanned aerial vehicles (UAVs) and SCoPEx. One such design, reported by NOAA (Murphy et al., 2016), measures at 4 wavelengths (460 nm, 550 nm , 670 nm, and 860 nm) with a field of view of 0.006 sr (equivalent to 2.5° half-angle) and a circular limiting aperture of 1.1 mm diameter. A radiometer like this one deployed on SCoPEx would be capable of observing a SCoPEx plume, based on Golja et al. (2020), formed by a 0.1 g s^{-1} injection of calcite from a distance of 200 m with an approximate signal-to-noise ratio of 6000 for a 1 ms signal accumulation.

3.3. Instruments for Future Science Flights (Science Goal 3)

The additional instruments listed in Table 3 are candidates for future SCoPEx flights beyond the initial science flight, i.e., addressing science goal 3. They have not yet been adapted to fly on the SCoPEx platform. Instrument choices will be refined based on experiences in the first science flights. The corresponding science goals that motivate their inclusion are detailed in Section 4.

Measurement	Candidate Instrument	Rationale	Corresponding science goal
Aerosol composition	Drum Sampler	Collecting aerosols for offline analysis	3
Water Vapor	IR Absorption or Frost Point	H_2O outgassing of platform, Influence on coagulation and heterogenous chemistry	2, 3
Atmospheric trace gas concentrations (ex: HCl, NOx)	Spectroscopic trace gas instruments	For measuring concentrations of various atmospheric trace gases before and after addition of solid ASI material	3

Table 3: Potential instrument for future SCoPEx science flights.

Aerosol Composition: Aerosol composition can be analyzed via the collection of aerosol with a drum sampler followed by offline analysis in the laboratory using standard offline methods. Aerosol sampling has been done numerous times aboard stratospheric platforms.

Water Vapor: Gas-phase water vapor measurements are important as relative humidity likely has a large impact on the heterogeneous reactivity of solid SAI material. The balloon and gondola can outgas significant amounts of water and thus an initial experiment will characterize how long, if at all, this outgassing perturbs the SCoPEx plume. As mentioned previously, the goal of SCoPEx is to ideally minimize the perturbation to only the introduction of calcium carbonate. Water vapor measurements are common on many stratospheric platforms.

Hydrogen Chloride: HCl can be measured via infrared absorption spectroscopy. The Anderson group at Harvard, which shares a laboratory with the Keutsch group, has developed a stratospheric HCl instrument and thus has extensive experience with the design of stratospheric HCl instrumentation. In addition, the Keutsch group has designed multiple spectroscopic trace gas measurements. The much lower air speeds of the balloon compared to aircraft favor the design of an open path system, which eliminates the notorious wall effects that can make HCl measurements challenging.

NOx: For NOx there exist a number of good instrumentation options. Recently, a compact NO-LIF instrument has been designed that has spectacular detection limits in the low ppt range, more than sufficient for the needs of SCoPEx. The instrument is a close analogue of the fiber-laser based formaldehyde LIF instrument that the Keutsch Group developed, so there is a high degree of expertise available for such an instrument. There are also sensitive cavity enhanced techniques available usually in the visible range of the spectrum.

3.4. SCoPEx Concept of Operations

Flights will proceed in the following manner. The payload would be launched with the ascender retracted such that there is minimal distance between the crossbar and platform. Once the balloon reaches the float altitude, the rope will be let out through the ascender such that there is 100 m between the crossbar and platform. The platform will then be ready to perform experiments and execute maneuvers. Figure 7 illustrates a proposed flight maneuver. The platform will initially travel in a straight line laying out a plume, after which it will maneuver back through the plume to make measurements. During these maneuvers the ascender can be used to fine tune the altitude of the platform and instruments. Several series of such maneuvers can be performed within each flight. At the conclusion of the experiments the ascender retracts the rope before the descent.

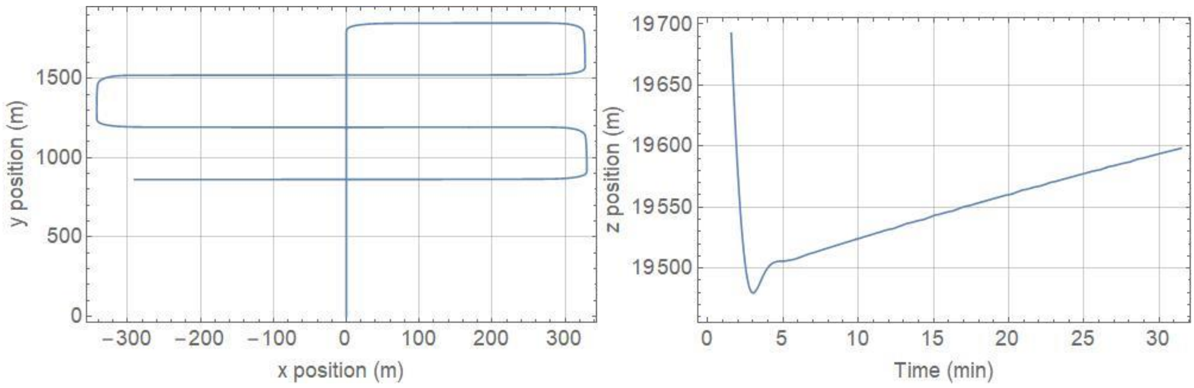


Figure 7: (left) A top down view of the proposed flight maneuvers over a 35-minute window. x and y are in the horizontal plane. The platform begins at (0,0). (right) The vertical position expected without any ascender or hopper vertical trimming over the same 35-minute platform maneuver.

4. SCoPEx Goals

In this section we describe the three long-term SCoPEx science goals. For each goal we describe the scientific problem, the need for SCoPEx, and the measurements required. The first phase of science flights targets the first two science goals. The design of the flights for the third goal will be informed by an understanding of the evolution of particle size distribution in the plume and the plume size. Thus, if later stage science flights move forward, they will be refined based on the results of the first science flights and the most up-to-date knowledge within the solar geoengineering and stratospheric science research communities.

4.1. Goal 1: Measurements of Turbulence for Small-Scale Mixing

4.1.1. The Importance of Plume-Scale Turbulence

Stratospheric turbulence influences the evolution of aerosol distribution from plume to regional to global scale. The mixing of air masses (of differing composition) in the stratosphere is a combination of two processes (Nakamura, 1996; Schoeberl & Bacmeister, 1993). The first process is strain, the distortion of streamline flow that brings air masses of differing composition adjacent to one another (Prather& Jaffe, 1990). Sometimes this is also referred to as “stirring” (Haynes, 2005). The second process occurs when air masses of differing composition are transported across the streamlines. This second process is the true “mixing” process.

In the stratosphere, mixing ultimately occurs because of molecular diffusion. This happens at the length scale of molecular viscosity. It is accelerated by turbulence, which can dramatically enhance the rate at which differing air masses are deformed to small enough spatial scales for molecular diffusion to mix them efficiently. Stratospheric turbulence is, however, highly intermittent (Vanneste, 2004). Understanding the mechanisms of stratospheric turbulence production is essential to understanding the spatial inhomogeneity and effective rate of mixing on spatial scales of 10-500 m (Schneider et al., 2017).

An understanding of this role of turbulence is of interest to stratospheric science because studies suggest that more accurate representations of mixing influence tracer distributions (Hoppe et al., 2014). Measurements of long-lived tracers are the strongest observational constraint on the stratospheric age of air, a key measure of the stratospheric large-scale circulation. Turbulence also modifies the character of kinetic energy fluxes. The magnitude and variability of these energy fluxes determine the rate of frictional dissipation in the atmosphere. This dissipation is represented in global models by a damping parameter and is the primary determinant of the mesoscale atmospheric kinetic energy spectrum. The uncertainty in kinetic spectrum is important to the understanding of the large-scale circulation of the middle atmosphere (Jablonowski & Williamson, 2011).

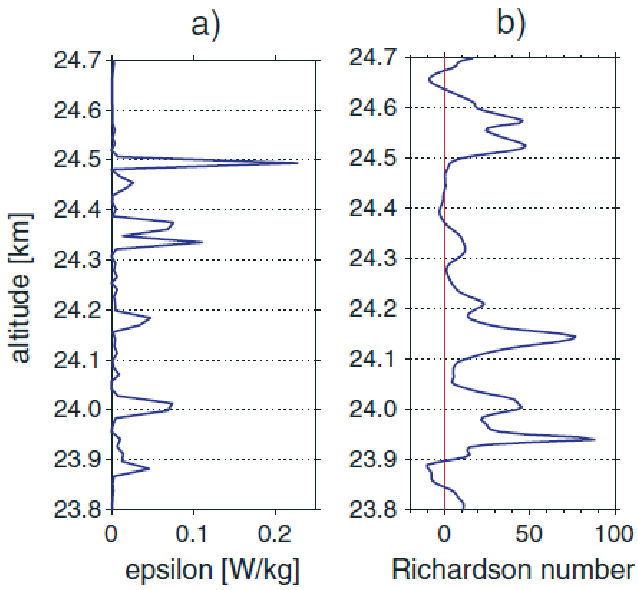


Figure 8: LITOS balloon-borne high-speed anemometer measurements reveal that models of atmospheric turbulence do not explain observed stratospheric turbulence. Physical models predict that a low Richardson number (buoyancy/shear ratio) implies turbulence, but high values of epsilon (turbulent dissipation) should be correlated with low Richardson number, which is not observed. (Haack et al., 2014)

Physical models predict that a low buoyancy/shear ratio (Richardson number) implies turbulence, and that high values of turbulent dissipation should be correlated with low Richardson number (Figure 8). However, recent balloon born measurements during the LITOS campaign did not agree with this, with numerous instances of high values of turbulent dissipation occurring at high Richardson numbers (Haack et al., 2014). As detailed above, both the impact of turbulence on mixing and the associated dissipation of energy are important for general stratospheric science. The point at which viscous fluid forces dominate atmospheric motion is the point where atmospheric motions become purely statistical and is called the dissipation scale. At this scale, models no longer require computationally expensive deterministic modeling. Furthermore, these viscous forces are also responsible for the dissipation of turbulent kinetic energy. Therefore, measurements which resolve the winds at the dissipation scale will allow numerical models to realistically close the atmospheric kinetic energy budget, an important metric of model fidelity.

4.1.2. Importance of Small-Scale Mixing for SAI and SCoPEx

From an SAI and SCoPEx perspective, plume-scale turbulence influences the frequency of collisions of monomer particles within the SCoPEx plume, which determines the rate of formation of fractal, larger aggregates. While Van der Waals forces finally determine whether particles that collide stick together and remain as a fractal aggregate (Sukhodolov et al., 2018), the collision rate is a critical quantity in determining total coagulation rate. Therefore, it is essential to know the frequency of collisions. This frequency is controlled by the wind variability at small spatial scales, i.e., the power spectrum. Intuitively, inertial forcing of particles by wind is much stronger than thermal forcing (e.g. Boltzmann distribution of velocity for ~1 µm particles at ~220 K). Fractal aggregates have a shorter lifetime in the stratosphere and are less effective at scattering light on a per mass basis (Weisenstein et al., 2015), so being able to model the formation rate of fractal aggregates is an important aspect of SAI, especially with alternate SAI materials.

Improved knowledge of collision rates from wind measurements will allow for the selection of the appropriate mathematical representation of particle coagulation, the coagulation kernel. An accurate kernel is essential for numerical models to correctly simulate aerosol microphysical processes that determine the size distribution and residence time of solid aerosol particles. Adding wind and turbulence measurements to the SCoPEx payload will therefore address the major sources of uncertainty in aerosol microphysics under real atmospheric conditions, which include small-scale fluid flow, particle composition, and humidity.

4.1.3. Experimental Methods to Measure Turbulence in the Stratosphere

Multiple technologies are possible to achieve wind measurements with the necessary spatial resolution under stratospheric conditions. Current state of the art options include pitot tubes (with high sensitivity micro-pirani pressure sensors), hot wire anemometers, and acoustic anemometers. An existing stratospheric program has utilized hot wire anemometers to make measurements that are a close analog to what is necessary for SCoPEx. The program developed LITOS (Leibniz-Institute Turbulence Observations in the Stratosphere), an instrument which made measurements of stratospheric turbulence up to 29 km (Gerding et al., 2009; Theuerkauf et al., 2011). The LITOS instrument has undergone significant calibration and has been compared against radiosondes (Schneider et al., 2015). One drawback of its deployment on a balloon has been the contamination of its wind measurements due to the influence of the balloon's wake. In contrast, SCoPEx is engineered so that the wind environment of the instrument payload is well separated from the balloon wake when SCoPEx is traveling horizontally. For this reason, SCoPEx could provide significantly more data per flight at a chosen float altitude. In this way, SCoPEx and LITOS would be very complementary. The horizontal flight path of SCoPEx, combined with measurements of the wind power spectrum, would provide an excellent complement to the LITOS observations, which are only obtained along a vertical profile. These power spectra obtained by SCoPEx would contribute to improved micrometeorology understanding relevant both to stratospheric aerosol injection and to fundamental atmospheric science.

Additionally, air flow through the turbulence instrument will be simulated using CFD tools. The CFD runs will provide a means to identify key flow characteristics that drive sensor performance (sensitivity and accuracy) and detailed sensor design. This application of the SCoPEx platform would therefore constitute a nonperturbative means to obtain necessary turbulence measurements that have, to date, eluded the scientific community. This information is important for understanding stratospheric dynamics, including the response to climate change or stratospheric heating from SAI. As no injection of particles is needed, these could be among the first scientific measurements to be conducted.

4.2. Goal 2: Evaluation of Aerosol Microphysics of AM-Sulfate and Alternative SAI Materials

One of the goals for which there are insufficient observational analogues is the near-field evolution of particles injected from a point source in the stratosphere. Specifically, observations of the temporal and spatial evolution of the aerosol size distribution (number and volume) of solid, alternate SAI materials or AM-H_2SO_4 injected form a point source can only be compared with plume model predictions via a perturbative experiment such as SCoPEx. In the following we describe a plume model by Golja et al. (2020) specifically designed for SCoPEx. We also explain the results from the model and the SCoPEx experimental approach for comparing observations with model results.

4.2.1. Plume Model

Golja et al. (2020) incorporated the SCoPEx design features in their model to study the injection of a solid aerosol and vapor-phase sulfuric acid from a balloon payload. To provide observations relevant to SAI, SCoPEx needs to produce downstream aerosols with radii within the range of roughly 0.2 to 1.0 µm. For calcium carbonate, the objective is to maintain a high fraction of the aerosol in monomer form, while for sulfate an ideal distribution would have a peak diameter of 0.6 µm (Dykema et al., 2016). The generation of largely smaller than ideal particles, while imperfect for assessing radiative efficiency relevant to SAI, does not serve to increase particle sedimentation rates within the plume. Such smaller sizes may, however, result in a larger surface to volume ratio, which can strongly influence stratospheric composition as heterogeneous chemistry is directly related to surface area. Distributions centered on small particle sizes in the near field may, however, continue to evolve beyond the domain of the study.

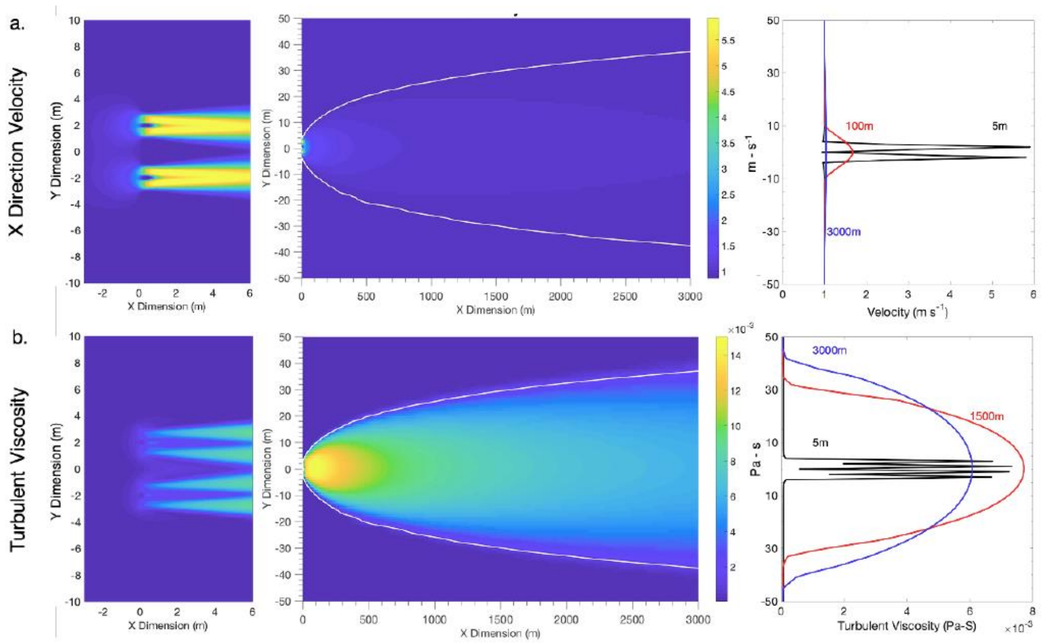


Figure 9 : ANSYS Fluent Velocity and Turbulence Fields. Shown above are the steady state x-direction velocity, u, and turbulent viscosity fields generated by ANSYS Fluent. Left panels show the genesis of disruptions to background X direction flow of 1 ms^{-1}, where propeller features are imposed at locations of 0,2) and (0,-2) meters. The center panel shows the entire domain, from 0 to 3 km, where the imposed red line contours 1 ms^{-1} in plot A, and contours 10% of the absolute maximum turbulent viscosity in plot B. Note Y direction scaling differs between the center and left panels. The right panel shows cross sections of velocity (A) and turbulent viscosity (B) through the Y plane at varying X locations. (Golja et al. 2020)

The velocity and turbulent viscosity fields from Fluent are shown in Figure 9. These fields form the basis of the simulation environment and are instructive in achieving an understanding of SCoPEx and the perturbation it achieves. Peaks in the x-direction velocity, u, are found directly downstream from the modeled propeller centers with an absolute maximum value of 6.3 ms^{-1}. By 1500 m downstream from the inlet locations, the velocity is reduced to the imposed background flow of 1 ms^{-1}. Turbulent viscosity, used as a measure of particle mixing with background air, exhibits a narrow distribution of peak values ~10 m downstream from simulated propellers. With increasing distance downstream, the turbulent velocity spatial distribution widens, attaining a full width half maximum (FWHM) of 60 m by 1500 m downstream. The wake of the balloon itself is not visible, as it is sufficiently far from the payload to avoid wake crossing/interaction. Additionally, this simulation assumes a laminar stratospheric background flow, neglecting the potential impacts of breaking gravity waves.

For SCoPEx, precipitated calcium carbonate powder with roughly monodisperse size distribution centered at ~0.5 mm diameter will be aerosolized using the expansion of powder suspended in high pressure CO_2 through a 1-2 mm nozzle (see description in Section 3). The model injects aerosol as a 3D gaussian distribution of mass flux into the model grid, where the size of that distribution represents the scale of which the high velocity jet from the nozzle mixes with ambient air. The model considered two injection scenarios: scenario 1 (S1), a single point injection between the propellers; and scenario 2 (S2), injection from the center of each propeller. The model plume diameter at 3 km is, however, insensitive to the injection scenario for injection of both AM-H_2SO_4 and calcium carbonate. This suggests that injection at or between the propellers does not significantly alter the characteristics of the particles' experienced velocity field, and scenario S1 is the one selected for testing the model of plume evolution on SCoPEx. This is also important for the SCoPEx experiment as it necessitates only one sprayer that can be more easily placed in the equipment gondola.

4.2.2. Modelled Mass Injection Rate Dependence of Aerosol Size Distribution

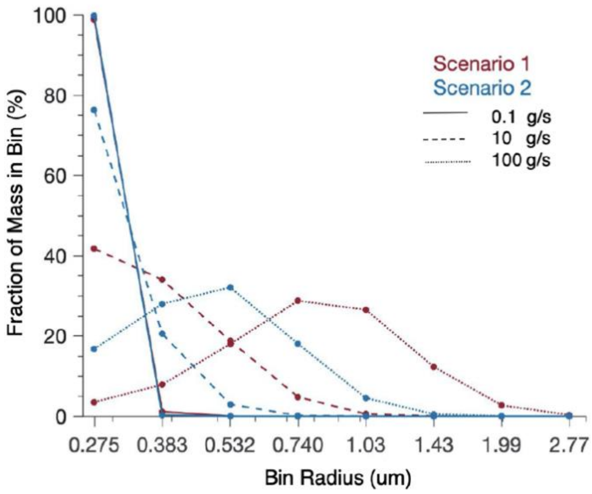


Figure 10: Calcium carbonate aerosol size distributions. Fraction of total mass in each sectional bin where the x-axis markers represent the central radius of each sectional size bin. These distributions represent the percent of total aerosol mass in the final 100 m of the plume across the full domain. Results are shown for three injection rates, 0.1 g s^{-1}, 10 gs^{-1}, and 100 gs^{-1}, for injection scenario 1 (red) and 2 (blue). (Golja et al. 2020)

Mass injection rates of 0.1, 10, and 100 g s^{-1} (0.36, 36, and 360 kg hr^{-1}) were used to test the influence of initial particle number density on the final plume aerosol size distribution. Although some of these are high, their use in the model is instructive as it can answer how different a short burst of high injection rate (much less than an hour) is from a slower but longer injection for the same total mass. Increasing calcium carbonate injection rates from 0.1 to 100 g s^{-1} reduces the share of monomer particles and increases undesired multi-monomer fractal aggregates. Figure 10 shows calcium carbonate's size distribution in the final 100 m of the modeled plume, i.e., the percent in each bin for the three different injection rates of 0.275 μm radius particles. The low calcium carbonate injection rate of 0.1 g s^{-1} is the most desirable, maintaining 99% of the total mass in the final 100 m of the plume in monomer form. Increasing mass injection rate to 10 g s^{-1} and 100 g s^{-1}, with an S1 injection, shifts peak mass loading to favor particles of radii 0.5 and 0.75 um, respectively, corresponding to fractal "dimers" and "trimers".

Golja et al. (2020) also evaluated whether, in addition to the very sensitive in-situ optical particle counting aerosol size distribution instrument which originally was designed to measure background stratospheric aerosol size distributions (Murphy et al., 2016), the plumes could also be detected optically via scattered light. It should be emphasized that this does not refer to measurements from the ground but rather from close to the plume, e.g., when the equipment gondola is in close vicinity to the plume. Measuring the scattering from one view angle gives the product of the scattering phase at that angle and the scattering efficiency. This is closely related to the radiative forcing, but it does not uniquely determine the radiative forcing. By measuring at multiple angles, we could obtain enough information to quantify the radiative forcing. For example, we could measure from the side and below to obtain the forward scatter fraction, then calculate backscatter by flux conservation.

In the model, the extinction optical depth was calculated using Mie scattering theory and vertically integrating down columns in the y-z plane. Figure 11 shows the relative optical thickness of a sulphate and calcite aerosol plume formed via scenario 1 with an injection rate of 0.1 g s^{-1}. Calcite exhibits greater optical thickness by an order of magnitude at 550 nm, with an average value of 8.6×10^{-4} and maximum of 0.014 across the domain, as compared to sulphate, with an average of 9.4×10^{-5} and maximum 0.001. From these values, Golja et al. calculated that we expect adequate SNR to confidently detect the plume with a fast-scanning radiometer via the solar radiation it scatters. This calculation assumed an altitude of 21 km, solar elevation angle of 60°, an observing instrument situated on the payload gondola, and the gondola 200 m away from the edge of the plume and 1 km downstream of the termination of a scenario 1 type injection of calcite aerosol. Details of this calculation can be found in Golja et al. (2020).

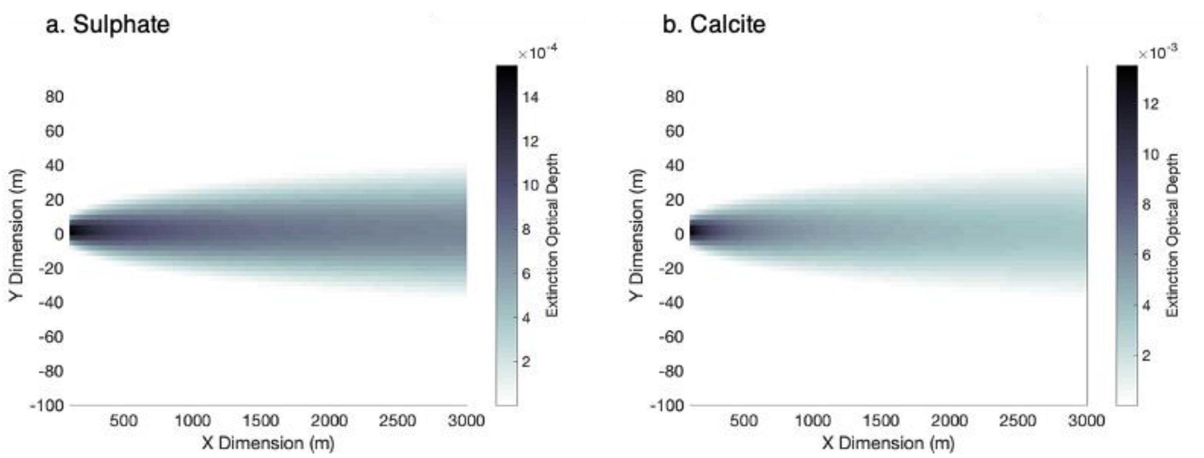


Figure 11: Extinction optical depth integrated vertically through all columns in the plume from 100-3000 m. Plots a and b show results for 0.1 gs^{-1} injections of condensable H_2SO_4 and calcite, respectively. The resulting number density of calcite aerosol is 490 cm^{-3} on the centerline at a downstream distance of 1000 m, predominantly as monomers. Aerosol optical depths were derived from Mie scattering theory at 550 nm, using refractive indices for sulphate and calcite stated in Dykema et al. (2016). (Golja et al. 2020)

4.2.3. SCoPEx Experimental Design and Analysis of Plume Evolution

For this goal, SCoPEx will follow the standard concept of operations, first spraying calcium carbonate at an injection rate suggested by the model analysis. It is desirable to maximize the contrast with the background stratosphere, both with respect to the aerosol concentration and the potential resulting chemical changes, while also maintaining calcium carbonate as monodisperse aerosol. To this end, additional models will be run at injection rates between 0.1 and 10 gs^{-1}. Based on these results, an injection rate will be chosen for the actual SCoPEx experiment. In addition to the basic components of the SCoPEx platform (gondola, ascender, propulsion, power, flight computer, communication, and wind), the calcium carbonate sprayer as well as the LIDAR and POPS instrument are critical for this science goal; without these components, there would not be a way to make and find the plume or measure the aerosol size distribution. While the turbulence measurement from goal 1 is desirable, it is, at least initially, not necessary. Similar studies of AM-H_2SO_4 injection would also be extremely useful. Our current plan is to conduct these after the calcium carbonate injection studies, as initially calcium carbonate is easier to handle than sulfuric acid and its precursors (see next section for motivation of calcium carbonate).

The aerosol size distribution measurements will be compared with the model predictions. In combination with turbulence measurements, discrepancies between the observed and modeled aerosol size distributions can be used to identify issues within the aerosol microphysical scheme or highlight misrepresentations of the velocity and turbulence field of the payload. The results of these studies will provide critical observational constraints on the aerosol microphysics and plume evolution of an injection with solid particles. It will be unique data that is ideal for testing the model of plume evolution as SCoPEx does not have to address problems resulting from the much more violent injection regime associated with injection from airplanes. Clearly, such studies are also needed, but SCoPEx represents a feasible and compelling first step in a sequence of new studies that more comprehensively investigate the aerosol microphysics of point source injections.

4.3. Goal 3: Evaluation of Process Level Chemical Models of Stratospheric Chemistry of Sulfate and Alternative SAI Materials

4.3.1. Need for Alternative SAI Materials

As previously discussed, the two largest first-order stratospheric risks of SAI with sulfate aerosol are ozone depletion and stratospheric heating. For sulfate aerosol the relative magnitude of these two risks can be adjusted if the size distribution can be controlled, e.g., via the AM-H_2SO_4 approach. It is worth noting that the impact on stratospheric ozone may be greatly reduced in the future if reactive halogen concentrations are lower. In contrast, the impact of stratospheric heating will not change. This represents a risk with a poorer understanding of its consequences, which makes it highly desirable to minimize stratospheric heating and resulting dynamic response. Therefore, it is important to investigate alternative SAI materials.

The properties of the "ideal" SAI material is (i) no absorption of radiation, i.e., purely scattering aerosol both fresh and aged, (ii) chemically inert, i.e., no direct impact of this material on stratospheric composition, and (iii) minimal down-stream effects, i.e., no impact on cirrus or other clouds, no environmental impact on deposition on the ground, etc. In reality, it is unlikely that a material with no impacts exists and rather the question is which materials can minimize these impacts. There have been a number of studies investigating

SAI materials in this context. High refractive index materials have been suggested as they reduce the mass of material that have to be lofted (Ferraro et al., 2015; Ferraro et al., 2011; Pope et al., 2012; Keith et al., 2016; Dai et al., 2020; Weisenstein et al., 2015). This largely cost-driven perspective is not a motivation for our work. In contrast, one of the goals of SCoPEx is to decrease the uncertainty in SRM models that use calcium carbonate SAI. The rationale for the choice of calcium carbonate as well as the approach to evaluate some of these risks is described in the following sections.

4.3.2. Unreactive Alternative SAI Materials

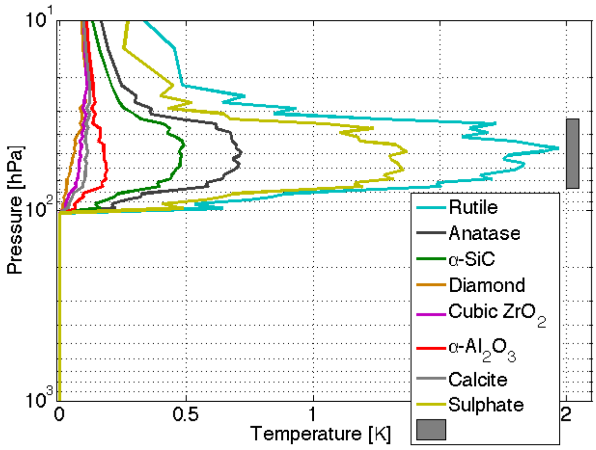


Figure 12: Comparison of stratospheric heating for different materials. Diamond has the lowest impact, although cubic zirconia and calcite are very similar. Sulfate and rutile result in much larger heating. (Dykema et al., 2016)

Diamond is probably the material with the best properties for SAI from a purely stratospheric perspective. Diamond has no absorption features in the solar or terrestrial spectrum and thus triggers the minimal possible dynamical response Figure 12. In addition, diamond should have ideal chemical properties. Hydrogen-terminated diamond surfaces are extremely inert and hydrophobic, precluding the ozone destroying chemistry initiated on sulfuric acid surfaces. The surface itself is also resistant to concentrated sulfuric acid. Exposure to OH radicals would probably slowly make the surface more hydrophilic. From a purely stratospheric perspective the only first-order risk of diamond would be increased ozone loss from the increased sulfuric acid surface area resulting from coagulation with background sulfate aerosol.

4.3.3. Reactive Alternative SAI Materials: The Case for Calcium Carbonate

Although the impact on cloud properties and the risk to Earth's surface from deposition of SAI diamond is likely very low, it could be preferable to have a material that dissolved easily in water, hence not persisting for long times outside of the stratosphere. It would also be preferable to have a material that is naturally abundant at Earth's surface. In addition, it would be ideal to overcome increased ozone loss due to coagulation by using a reactive aerosol. We therefore propose calcium carbonate as a prototype alternate SAI material for the following reasons: First, its optical properties are nearly equal to diamond and stratospheric heating and resulting dynamic response should be negligible compared to sulfate (Figure 12). Second, carbonates are typically quite reactive with acids, especially with concentrated sulfuric acid (Figure 13). Hence, calcium carbonate will neutralize upon coagulation with sulfate aerosol eliminating the acidic surfaces resulting from coagulation of diamond and sulfate aerosol. Of course, the reactivity of calcium carbonate also makes model predictions with calcium carbonate more complex. The evolution of chemical and optical aerosol properties has to be modeled over its stratospheric lifetimes. One of the key research questions that SCoPEx will help address is whether the reactivity of calcium carbonate and the evolution of its chemical and optical properties and those of the surrounding gas-phase correspond to the detailed hypothesis laid out below. To this end, SCoPEx will compare observations of the chemical evolution of calcium carbonate, as well as the gas-phase, with those of a model based on known properties of calcium carbonate and recent laboratory experiments (Dai et al., 2020). This will provide a real-world evaluation of kinetic parameters, such as heterogeneous uptake coefficients derived from the laboratory studies, that will enable GCMs to include reliable parameterizations of the stratospheric impacts of calcium carbonate SAI.

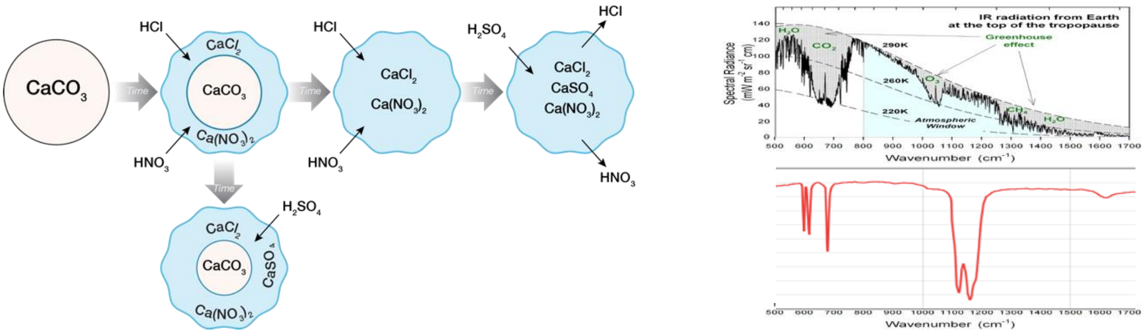


Figure 13: The left panel shows schematic of potential chemical reactivity of calcium carbonate in the stratosphere. The right panel shows the atmospheric windows in the terrestrial infrared (top) as well as the infrared absorption spectrum of calcium sulfate (bottom). The position of the 1150 cm^{-1} sulfate in part explains the stratospheric heating effect of sulfuric acid.

4.3.3.1. Optical Properties

Based on well-established chemistry, the reaction of sulfuric acid aerosol with calcium carbonate can be assumed to go to completion, i.e., be reagent limited. The optical properties of calcium sulfate in the terrestrial infrared are similar to those of sulfuric acid with only slight differences in relative band intensities and wavelengths (Figure 13 right hand inset). This is important as it implies that there will be no large first-order changes in stratospheric heating from changing background sulfuric acid to calcium sulfate. There are higher order impacts due to slight differences in the absorption of sulfuric acid, which has some liquid water compared to calcium sulfate. There are also numerous forms of calcium sulfate (anhydrite, bassanite, gypsum, etc.). However, the resulting differences are much smaller than introducing an absorbing material via SAI.

4.3.3.2. Chemical Properties

Predicting the evolution of the chemical properties of calcium carbonate under stratospheric conditions is more challenging. It is certain that calcium carbonate does not have the same heterogeneous reactions that activate ozone destroying substances as sulfuric acid. Figure 13 shows a schematic of the expected reactivity. Calcium carbonate is expected to react with acidic substances neutralizing them, forming salts and carbon dioxide. These acid neutralizing reactions can deplete gas-phase HNO_3, HCl, etc. There are a large number of ozone destroying catalytic cycles involving NOx, chlorine and other halogens, which are altitude (and latitude) dependent. NOx can be produced via HNO_3 photolysis and lost via heterogeneous reaction of N_2O_5. It participates both in ozone destroying catalytic cycles and is important for deactivation of ozone destroying halogen radicals. Thus, knowledge of the heterogeneous reaction rates of numerous substances with calcium carbonate are required to predict the impact it will have on stratospheric composition.

However, until the recent study by Dai et al. in our laboratory, no heterogeneous chemistry studies of calcium carbonate under stratospheric conditions had been conducted, to our knowledge, although there exists a rich data set under tropospheric conditions (Dai et al., 2020). This work, as well as the work of Dai et al., highlights that reactive solid aerosols are indeed more complex than liquid sulfuric acid: The authors observed moderate initial uptake of the gas-phase acids HCl and HNO_3 on fresh calcium carbonate, as the dry stratospheric conditions already make uptake coefficients lower than under typical tropospheric conditions. An additional large difference to liquid aerosol is that the surface of the solid calcium carbonate passivates, drastically reducing the uptake coefficients of HCl and HNO_3. Hence, based on the Dai et al. laboratory study, calcium carbonate rapidly becomes effectively unreactive with respect to uptake of these gas-phase acids, an important finding that confirms calcium carbonate as a good candidate as alternate SAI material. In addition, calcium carbonate particles are abundant at Earth's surface due to windblown mineral dust. And the small calcium carbonate SAI particles should dissolve rapidly in water. This does not exclude risks associated with the deposition of calcium carbonate SAI particles or impacts on clouds (Cziczo et al., 2019). However, due to its abundance at the Earth's surface, there already exists a large knowledge base for its environmental impacts in contrast to, e.g., diamond. Further laboratory work is required to study especially the $ClONO_2$ + HCl and N_2O_5 hydrolysis reactions on fresh and aged calcium carbonate. However, the existing results prepare the stage for studying them in the real stratospheric environment as outlined below. Figure 14 shows results of the AER 2-D chemistry-transport-aerosol model for annual average ozone column changes of calcium carbonate SAI compared to a control for 2040. Ignoring the passivation of calcium carbonate (thk-ind) results in increases in ozone columns from calcium carbonate SAI whereas the inclusion of passivation can either result in very little ozone column change or losses in the Southern Hemisphere, depending how the $ClONO_2$+HCl is parameterized. Either of the two, more realistic, passivation scenarios result in significantly lower ozone loss than the equivalent amount of sulfate SAI, consistent with the hypothesis.

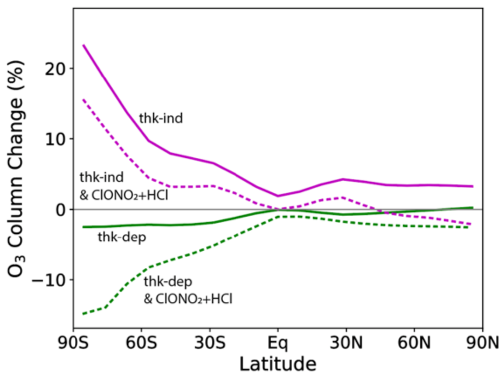


Figure 14: Shows the role of passivation and the heterogeneous $ClONO_2$+HCl reaction on ozone column change using the AER 2-D model taken from Dai et al. 2020. Inclusion of this reaction with the same rate as measured for Al_2O_3 results in a substantial reduction in ozone for scenarios including, thk-ind, or excluding passivation, thk-dep.

4.3.4. Need for SCoPEx Calcium Carbonate Plume Studies

One of the challenges for alternate SAI aerosol is the lack of materials such as calcium carbonate in the stratosphere. The only way to then study these materials in the actual stratosphere is via deliberate stratospheric injection of a small amount of these materials. In environmental studies, including stratospheric studies, it is not possible to rely purely on laboratory studies. For example, flights on the NASA ER-2 into the polar vortex over Antarctica provided the ability to test whether laboratory-derived reaction mechanisms were able to capture real-world ozone destruction chemistry. Without these flights, the level of confidence in the model predictions would have been much lower, and for good reason. It is not clear that a given experimental setup in the laboratory can faithfully capture the entire complexity of the real stratosphere; only field observations are able to provide this. For a number of natural stratospheric processes, remote observations can provide important information in addition to in situ aircraft or balloon. However, these are only possible when large-scale phenomena are at work.

Since there are no natural calcium carbonate plumes in the stratosphere that would even allow for in situ observations, intentional injection is necessary to perform these studies. Calcium carbonate injections will allow SCoPEx to provide invaluable observations as it will quantitively test the mechanisms determined in the laboratory. As stated above, there is a need for more laboratory studies, however, there is good reason to proceed with the planning of SCoPEx calcium carbonate experiments. First, by the time of the first injection experiments, additional studies should have been conducted. In addition, N_2O_5 uptake coefficients used in the model are likely a very good estimation as similar values have been found for different solid materials, e.g., Al_2O_3 and SiO_2 (Molina et al., 1997). In addition, even with these additional lab determined mechanisms, the same type of experiments as proposed here will still have to be conducted, as we expect these reactions to not make a significant difference. In other words, they will not be a deciding factor about the viability of calcium carbonate as an alternate SAI material. Only field experiments will help shed insight into these questions. In summary, there is a critical need for evaluating not just the aerosol microphysics (goal 2) but also the stratospheric chemistry of calcium carbonate due to the promise it holds as a lower risk SAI material.

4.3.5. SCoPEx Experimental Design and Analysis of Chemical Calcium Carbonate Plume Evolution

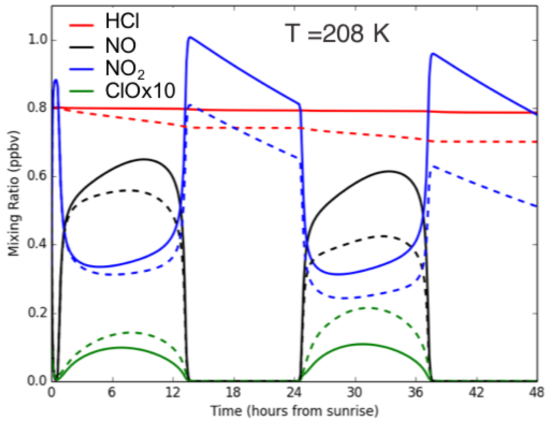


Figure 15: Solid lines: background 2µm^2 cm^{-3} sulfate 5ppm$_v$ H_2O. Dashed lines: plume 15µm^2 cm^{-3} sulfate 10 ppm$_v$ H_2O.

The experiments will again follow the standard concept of operations as under goal 2. In order to determine optimal injection rates, we will include chemical reactions in the plume model, updated with the newest mechanisms available at that time. Figure 15 shows the evolution of an air mass perturbed by a sulfate aerosol injection over multiple days, i.e., significantly longer than the initial SCoPEx experiments. Significant changes in HCl and NOx can be observed already over short time periods and these are easily detectable with existing instrumentation. For this science goal, it is desirable to measure aerosol composition and size distribution as well as key gas-phase chemical species, especially HCl, NOx and water. Therefore, this science goal requires a much larger set of instruments. In addition, the equivalent model to Figure 15 for calcium carbonate is informed by the results of science goal 2. The work of Dai et al. provides kinetic parameters needed for this model, and reactions for which there are no laboratory data to date are parameterized using close analogues and conditions, e.g., $ClONO_2$ + HCl are parameterized using the results for alumina (and silica) from Molina et al. (1997). One key question is whether the changes in HCl and NOx will indeed be smaller for calcium carbonate than those for sulfate shown in the figure above, which would confirm the hypothesis for calcium carbonate as a potential alternate SAI material.

In summary, SCoPEx experiments using calcium carbonate injections will provide a unique evaluation as to whether calcium carbonate indeed is an alternate SAI material that could substantially reduce risk from SAI compared to sulfate. Follow-up studies will be needed. For example, improved chemical and aerosol microphysics models will provide improved models of the chemical and physical evolution of calcium carbonate, which likely will motivate specific laboratory investigations. These will provide information for SCoPEx studies using “stratospherically aged” calcium carbonate as precursor for injection that can then be used to compare whether the laboratory mechanisms of this aged calcium carbonate agree with that found in the real stratospheric environment.

5. Data Management Plan and Dissemination of Results

Products of the research. The data generated during this project consists of meteorological, navigational, telemetry, and a variety of instrumentation data, in particular aerosol size distributions as well as chemical composition data during later science flights. In addition, there will be model data on plume chemical evolution.

Access to data, data sharing practices, and policies and dissemination of results. Data relevant for scientific analysis will be made public within 60 days of the end of flight. This taw data will be made public with appropriate warnings that it has not undergone QA/QC. The email address of users will be recorded so that they can be automatically notified when revised versions become available. Based on previous experiences with stratospheric airborne campaigns, this is typically 6-15 months after the flight depending on the type of data, e.g., the amount of calibration and data workup required. We have chosen to make raw data available rapidly—going far beyond what is typical for stratospheric science missions—because of the public scrutiny of SCoPEx and because of the broad commitment to Open Access data principles articulated by Harvard's Solar Geoengineering Research Program which is funding SCoPEx.

Principal Investigators (PI) and their groups have an excellent track record with presenting their work at major national and international conferences and workshops. All data that go into key analyses and figures in the group's publications will be made publicly available via the PI's group website. All publications resulting from this project will be posted on the PI's webpage (https://projects.iq.harvard.edu/keutschgroup/publications). Preprints of manuscripts submitted for publication as well as the underlying data will also be posted on Harvard's Dash manuscript repository. Publications will be made in open access formats.

Archiving of data. All data acquisition/storage computers in the PI's group are automatically backed up daily, both wirelessly to a server elsewhere on campus, and/or to a cloud server. Both of these processes ensure that data will not be lost and enable rapid access to the data. The file naming system used for all software (which includes the date of the experiment) ensures straightforward retrieval and use of archived data. Group laptops are also backed up daily, ensuring that analyzed data are archived as well.

6. SCoPEx Research Team Biographies

Frank Keutsch was born in Tübingen, Germany and received his Diplom in chemistry from the Technische Universität München, Germany, under the supervision of Vladimir E. Bondybey in 1997. He received his PhD in physical chemistry from the University of California at Berkeley in 2001. His graduate research was conducted under the direction of Richard J. Saykally and focused on vibration−rotation−tunneling spectroscopy and hydrogen-bond-breaking dynamics in water clusters. After working on stratospheric chemistry in the Department of Chemistry and Chemical Biology at Harvard University under the direction of James G. Anderson, he started his independent academic career in 2005 at the University of Wisconsin-Madison. He then moved to his current position as Stonington Professor of Engineering and Atmospheric Science at Harvard University in the Paulson School of Engineering and Applied Sciences and the Department of Chemistry and Chemical Biology and he has held numerous visiting professor positions. Keutsch Group research combines laboratory and field experiments with instrument development to investigate fundamental mechanisms of anthropogenic influence on atmospheric composition within the context of impacts on climate, humans and the environment. Keutsch's main focus has been on understanding how *unintentional* emissions of pollutants such as nitrogen oxides, sulfur dioxide, and hydrocarbons have changed key chemical pathways controlling ozone and particulate matter, two key pollutants affecting human health and climate. Keutsch has been the PI of numerous research grants for this research and currently is the deputy-PI for the NASA-EVS3 DCOTSS campaign. Keutsch has also been focusing on improving the understanding of how *intentional* emissions within the context of stratospheric solar radiation modification could impact the protective stratospheric ozone layer and stratospheric dynamics and climate, and how known risks can be better quantified or reduced. He is currently the PI of SCoPEx. Keutsch has received awards for his teaching, which spans a wide range of courses including introductory chemistry, engineering design and atmospheric chemistry.

David Keith has worked near the interface between climate science, energy technology, and public policy since 1991. He received his B.Sc. in physics from the University of Toronto in 1986 and received his PhD in experimental physics from the Massachusetts Institute of Technology in 1991 under the supervision of David Prichard. He took first prize in Canada's national physics prize exam, won MIT's prize for excellence in experimental physics, and was one of TIME Magazine's Heroes of the Environment. David is Professor of Applied Physics at the Harvard School of Engineering and Applied Sciences and Professor of Public Policy at the Harvard Kennedy School, and founder of Carbon Engineering, a Canadian company developing technology to capture CO_2 from ambient air to make carbon-neutral hydrocarbon fuels. Best known for his work on the science, technology, and public policy of solar geoengineering, David led the development of Harvard's Solar Geoengineering Research Program, a Harvard-wide interfaculty research initiative. His work has ranged from the climatic impacts of large-scale wind power to an early critique of the prospects for hydrogen fuel. David's hardware engineering work includes the first interferometer for atoms, a high-accuracy infrared spectrometer for NASA's ER-2, and the development of Carbon Engineering's air contactor and overall process design. On SCoPEx, he is the faculty lead for platform design and engineering. David teaches science and technology policy, climate science, and solar geoengineering. He has reached students worldwide with an edX energy course. David is author of >200 academic publications with total citation count of >15,000. He has written for the public in op-eds and *A Case for Climate Engineering*. David splits his time between Cambridge, Massachusetts and Canmore, Alberta.

Norton Allen is Head Software Engineer for the Anderson, Keith, and Keutsch groups in the Harvard John A. Paulson School of Engineering and Applied Sciences. Working closely with electrical and mechanical engineering, he is responsible for the design and deployment of software for data acquisition and control on all flight instruments. He has successfully deployed over two dozen instruments and supported field deployments in locations around the world. He received an AB cum laude from Harvard College, studying math, applied math, computer science, and physics.

John Dykema is a Project Scientist at the Harvard John A. Paulson School of Engineering and Applied Sciences and the LIDAR principal investigator on SCoPEx. His main interests are atmospheric radiation and remote sensing instrumentation, with an emphasis on development of novel, compact LIDARS for trace gas and aerosol measurement. John earned his AB in physics from UC Berkeley and his PhD in applied physics from Harvard University, where his dissertation focused on developing a new airborne infrared sounder that was a prototype for a climate-focused atmospheric radiation mission. He is participating in the NASA DCOTSS mission as the principal investigator for the POPS optical particle counter and as a member of the DCOTSS aerosol science subgroup. He also collaborates with several external organizations in designing and simulating new LIDAR prototypes, incorporating emerging laser and optical technology. John leads the engineering development and data analysis for the SCoPEx LIDAR and works on the radiative and micrometeorological science aspects of the SCoPEx mission.

Mike Greenberg is the Lead Optical-Mechanical engineer for the Anderson, Keith, and Keutsch groups in the Harvard John A. Paulson School of Engineering and Applied Sciences. He is responsible for the mechanical development and implementation of flight and laboratory based instrumentation, equipment packaging, documentation, and platform integration. Working closely with the electrical, software, and science team members, he has over 20 years of experience developing, delivering, and supporting designs and has been on more than a dozen airborne campaigns with the ER-2, WB-57, and DA-42 aircraft platforms and with stratospheric balloons. Mike received a BSME from Tufts University and a MSME from Stanford University. His additional work experiences include time spent Argonne National Laboratory and The Raytheon Company.

Michael Litchfield is the Senior Engineering Lead for Climate Research in the Anderson, Keith, Keutsch groups at the Harvard John A. Paulson School of Engineering and Applied Sciences and the engineering lead on the SCoPEx Flight Platform development program. He and the rest of the engineering team are focused on taking high level SCoPEx flight platform requirements through the design, fabrication, assembly, test, and validation processes. Michael earned his BS and MS degrees in Electrical Engineering specializing in controls and communications systems at Worcester Polytechnic Institute. Prior to joining the lab to assume this role, Michael worked for over 30 years in industry across 5 start-ups leading their various engineering teams in bringing first products to market where those markets included; X-ray Semiconductor Lithography, 3D Ultrasound

Medical imaging , X-ray 2D Projection / 3D CT Airport Baggage Security Imaging, and 4D (3D movies) mmWave Personnel Security imaging.

Craig Mascarenhas is a mechanical engineer for the Anderson, Keith, and Keutsch groups in the Harvard John A. Paulson School of Engineering and Applied Sciences. He is responsible for the mechanical design and integration of instrumentation, equipment packaging, and aerodynamic analysis of flight systems. He has previously been involved in instrument design for airborne campaigns with the ER-2 and stratospheric balloons. Craig received a BASc from the University of Toronto and an SM from MIT. His additional work experiences include engineering roles in the nuclear, biotech, and hydro-power industries.

Terry Martin is an electronics technician with the Anderson, Keith, and Keutsch research groups. She has worked on electrical build up and documentation of numerous scientific experiments over the course of the 42 years she has been with the group and is presently helping with the electronic assembly and wiring of the SCoPEx instrument.

Marco Rivero is a senior Electrical Engineer in the Anderson, Keith, and Keutsch groups in the Harvard John A. Paulson School of Engineering and Applied Sciences. As such, he has been primarily involved in the electrical engineering design, fabrication, and testing of the SCoPEx platform and payload instrumentation since inception. Marco holds a BS in Microelectronic Engineering from Rochester Institute of Technology and a MS in Electrical Engineering from Tufts University. During his 25 years with the group, Marco has been involved in the electronics and systems design of 14 airborne instruments and supported their deployment in over 20 NASA national and international field campaigns; most recently, a HCl instrument deployment out of NASA's Columbia Scientific Balloon Facility in Fort Sumner NM in August of 2018.

Yomay Shyur is a Postdoctoral Fellow at the Harvard John A. Paulson School of Engineering and Applied Sciences and a project manager and project scientist on SCoPEx. She leads technical project coordination, works on science instrument design and analysis, and assists with platform engineering tasks. Yomay earned her BA in physics from Wellesley College and her PhD in physics from the University of Colorado Boulder, where her dissertation focused on developing new experimental methods of manipulating cold molecules using high-voltage electrodes and laser detection techniques.

References

Anderson, M. R., Miake-Lye, R. C., Brown, R. C., & Kolb, C. E. (1996). Calculation of exhaust plume structure and emissions of the ER 2 aircraft in the stratosphere. *Journal of Geophysical Research: Atmospheres*, *101*(D2), 4025–4032. https://doi.org/10.1029/95JD02366

Bala, G., Duffy, P. B., & Taylor, K. E. (2008). Impact of geoengineering schemes on the global hydrological cycle. *Proceedings of the National Academy of Sciences*, *105*(22), 7664–7669. https://doi.org/10.1073/pnas.0711648105

Balluch, M. G., & Haynes, P. H. (1997). Quantification of lower stratospheric mixing processes using aircraft data. *Journal of Geophysical Research: Atmospheres*, *102*(D19), 23487–23504. https://doi.org/10.1029/97JD00607

Benduhn, F., Schallock, J., & Lawrence, M. G. (2016). Early growth dynamical implications for the steerability of stratospheric solar radiation management via sulfur aerosol particles. *Geophysical Research Letters*, *43*(18), 9956–9963. https://doi.org/10.1002/2016GL070701

Butchart, N., Anstey, J. A., Hamilton, K., Osprey, S., McLandress, C., Bushell, A. C., Kawatani, Y., Kim, Y.-H., Lott, F., Scinocca, J., Stockdale, T. N., Andrews, M., Bellprat, O., Braesicke, P., Cagnazzo, C., Chen, C.-C., Chun, H.-Y., Dobrynin, M., Garcia, R. R., … Yukimoto, S. (2018). Overview of experiment design and comparison of models participating in phase 1 of the SPARC Quasi-Biennial Oscillation initiative (QBOi). *Geoscientific Model Development*, *11*(3), 1009–1032. https://doi.org/10.5194/gmd-11-1009-2018

Conway, J. P., Bodeker, G. E., Waugh, D. W., Murphy, D. J., Cameron, C., & Lewis, J. (2019). Using Project Loon Superpressure Balloon Observations to Investigate the Inertial Peak in the Intrinsic Wind Spectrum in the Midlatitude Stratosphere. *Journal of Geophysical Research: Atmospheres*, *124*(15), 8594–8604. https://doi.org/10.1029/2018JD030195

Crutzen, P. J. (2006). Albedo Enhancement by Stratospheric Sulfur Injections: A Contribution to Resolve a Policy Dilemma? *Climatic Change*, *77*(3), 211. https://doi.org/10.1007/s10584-006-9101-y

Cziczo, D. J., Wolf, M. J., Gasparini, B., Münch, S., & Lohmann, U. (2019). Unanticipated Side Effects of Stratospheric Albedo Modification Proposals Due to Aerosol Composition and Phase. *Scientific Reports*, *9*(1), 18825. https://doi.org/10.1038/s41598-019-53595-3

Dai, Z., F. Keutsch, D. Weisenstein, and D. W. Keith. (2020). Experimental reaction rates constrain estimates of ozone response to calcium carbonate geoengineering, *Manuscript submitted for publication.*

Dykema, J., Keith, D., Anderson, J. G., & Weisenstein, D. (2014). Stratospheric controlled perturbation experiment (SCoPEx): A small-scale experiment to improve understanding of the risks of solar geoengineering. *Philosophical Transactions of the Royal Society A*, *372*. https://doi.org/10.1098/rsta.2014.0059

Dykema, J., Keith, D., & Keutsch, F. (2016). Improved aerosol radiative properties as a foundation for solar geoengineering risk assessment. *Geophysical Research Letters*. http://onlinelibrary.wiley.com/doi/10.1002/2016GL069258/full

Fahey, D. W., Keim, E. R., Boering, K. A., Brock, C. A., Wilson, J. C., Jonsson, H. H., Anthony, S., Hanisco, T. F., Wennberg, P. O., Miake-Lye, R. C., Salawitch, R. J., Louisnard, N., Woodbridge, E. L., Gao, R. S., Donnelly, S. G., Wamsley, R. C., Del Negro, L. A., Solomon, S., Daube, B. C., … Chan, K. R. (1995a). Emission Measurements of the Concorde Supersonic Aircraft in the Lower Stratosphere. *Science*, *270*(5233), 70–74.

Fahey, D. W., Keim, E. R., Woodbridge, E. L., Gao, R. S., Boering, K. A., Daube, B. C., Wofsy, S. C., Lohmann, R. P., Hintsa, E. J., Dessler, A. E., Webster, C. R., May, R. D., Brock, C. A., Wilson, J. C., Miake-Lye, R. C., Brown, R. C., Rodriguez, J. M., Loewenstein, M., Proffitt, M. H., … Chan, K. R. (1995b). In situ observations in aircraft exhaust plumes in the lower stratosphere at midlatitudes. *Journal of Geophysical Research: Atmospheres*, *100*(D2), 3065–3074. https://doi.org/10.1029/94JD02298

Farquhar, G. D., & Roderick, M. L. (2003). Pinatubo, Diffuse Light, and the Carbon Cycle. *Science*, *299*(5615), 1997–1998. https://doi.org/10.1126/science.1080681

Ferraro, A. J., Charlton-Perez, A. J., & Highwood, E. J. (2015). Stratospheric dynamics and midlatitude jets under geoengineering with space mirrors and sulfate and titania aerosols. *Journal of Geophysical Research: Atmospheres*, *120*(2), 414–429. https://doi.org/10.1002/2014JD022734

Ferraro, A. J., Highwood, E. J., & Charlton-Perez, A. J. (2011). Stratospheric heating by potential geoengineering aerosols. *Geophysical Research Letters*, *38*(24). https://doi.org/10.1029/2011GL049761

Gao, R. S., Telg, H., McLaughlin, R. J., Ciciora, S. J., Watts, L. A., Richardson, M. S., Schwarz, J. P., Perring, A. E., Thornberry, T. D., Rollins, A. W., Markovic, M. Z., Bates, T. S., Johnson, J. E., & Fahey, D. W. (2016). A light-weight, high-sensitivity particle spectrometer for PM2.5 aerosol measurements. *Aerosol Science and Technology*, *50*(1), 88–99. https://doi.org/10.1080/02786826.2015.1131809

Gerding, M., Theuerkauf, A., & Suminska, O. (2009). Balloon-borne hot wire anemometer for stratospheric turbulence soundings. *Proceedings of the 19th ESA Symposium on European Rocket and Balloon Programmes and Related Research, SP-671*.

Golja, C. M., L. W. Chew, J. A. Dykema, and D. W. Keith. (2020). Aerosol Dynamics in the Near Field of the SCoPEx Stratospheric Balloon Experiment, *Manuscript submitted for publication.*

Gu, L., Baldocchi, D. D., Wofsy, S. C., Munger, J. W., Michalsky, J. J., Urbanski, S. P., & Boden, T. A. (2003). Response of a Deciduous Forest to the Mount Pinatubo Eruption: Enhanced Photosynthesis. *Science*, *299*(5615), 2035–2038. https://doi.org/10.1126/science.1078366

Gu, L., Baldocchi, D., Verma, S. B., Black, T. A., Vesala, T., Falge, E. M., & Dowty, P. R. (2002). Advantages of diffuse radiation for terrestrial ecosystem productivity. *Journal of Geophysical Research: Atmospheres*, *107*(D6), ACL 2-1-ACL 2-23.

https://doi.org/10.1029/2001JD001242
Haack, A., Gerding, M., & Lübken, F.-J. (2014). Characteristics of stratospheric turbulent layers measured by LITOS and their relation to the Richardson number. *Journal of Geophysical Research: Atmospheres*, *119*(18), 10,605-10,618. https://doi.org/10.1002/2013JD021008
Haynes, P. H. (2005). Transport and Mixing in the Atmosphere. In W. Gutkowski & T. A. Kowalewski (Eds.), *Mechanics of the 21st Century* (pp. 139–152). Springer Netherlands. https://doi.org/10.1007/1-4020-3559-4_8
Holben, B. N., Eck, T. F., Slutsker, I., Tanré, D., Buis, J. P., Setzer, A., Vermote, E., Reagan, J. A., Kaufman, Y. J., Nakajima, T., Lavenu, F., Jankowiak, I., & Smirnov, A. (1998). AERONET—A Federated Instrument Network and Data Archive for Aerosol Characterization. *Remote Sensing of Environment*, *66*(1), 1–16. https://doi.org/10.1016/S0034-4257(98)00031-5
Homeyer, C. R., Bowman, K. P., Pan, L. L., Atlas, E. L., Gao, R.-S., & Campos, T. L. (2011). Dynamical and chemical characteristics of tropospheric intrusions observed during START08. *Journal of Geophysical Research: Atmospheres*, *116*(D6). https://doi.org/10.1029/2010JD015098
Hoppe, C. M., Hoffmann, L., Konopka, P., GrooÃ, J.-U., Ploeger, F., Günther, G., Jöckel, P., & Müller, R. (2014). The implementation of the CLaMS Lagrangian transport core into the chemistry climate model EMAC 2.40.1: Application on age of air and transport of long-lived trace species. *Geoscientific Model Development*, *7*(6), 2639–2651. https://doi.org/10.5194/gmd-7-2639-2014
Jablonowski, C., & Williamson, D. L. (2011). The Pros and Cons of Diffusion, Filters and Fixers in Atmospheric General Circulation Models. In P. Lauritzen, C. Jablonowski, M. Taylor, & R. Nair (Eds.), *Numerical Techniques for Global Atmospheric Models* (pp. 381–493). Springer. https://doi.org/10.1007/978-3-642-11640-7_13
Ji, D., Fang, S., Curry, C. L., Kashimura, H., Watanabe, S., Cole, J. N. S., Lenton, A., Muri, H., Kravitz, B., & Moore, J. C. (2018). Extreme temperature and precipitation response to solar dimming and stratospheric aerosol geoengineering. *Atmospheric Chemistry and Physics*, *18*(14), 10133–10156. https://doi.org/10.5194/acp-18-10133-2018
Keith, D., Weisenstein, D., Dykema, J., & Keutsch, F. (2016). Stratospheric Solar Geoengineering without Ozone Loss. *Proceedings of the National Academy of Sciences*. http://www.pnas.org/content/113/52/14910.full
Kravitz, B., Robock, A., Boucher, O., Schmidt, H., Taylor, K. E., Stenchikov, G., & Schulz, M. (2011). The Geoengineering Model Intercomparison Project (GeoMIP). *Atmospheric Science Letters*, *12*(2), 162–167. https://doi.org/10.1002/asl.316
Legras, B., Pisso, I., Berthet, G., & Lefèvre, F. (2005). Variability of the Lagrangian turbulent diffusion in the lower stratosphere. *Atmospheric Chemistry and Physics*, *5*(6), 1605–1622. https://doi.org/10.5194/acp-5-1605-2005
Määttänen, A., Merikanto, J., Henschel, H., Duplissy, J., Makkonen, R., Ortega, I. K., & Vehkamäki, H. (2018). New Parameterizations for Neutral and Ion-Induced Sulfuric

Acid-Water Particle Formation in Nucleation and Kinetic Regimes. *Journal of Geophysical Research: Atmospheres*, *123*(2), 1269–1296. https://doi.org/10.1002/2017JD027429

Maruca, B. A., Marino, R., Sundkvist, D., Godbole, N. H., Constantin, S., Carbone, V., & Zimmerman, H. (2017). Overview of and first observations from the TILDAE High-Altitude Balloon Mission. *Atmospheric Measurement Techniques*, *10*(4), 1595–1607. https://doi.org/10.5194/amt-10-1595-2017

McCormick, M. P., Thomason, L. W., & Trepte, C. R. (1995). Atmospheric effects of the Mt Pinatubo eruption. *Nature*, *373*(6513), 399–404. https://doi.org/10.1038/373399a0

Molina, M. J., Molina, L. T., Zhang, R., Meads, R. F., & Spencer, D. D. (1997). The reaction of ClONO2 with HCl on aluminum oxide. *Geophysical Research Letters*, *24*(13), 1619–1622. https://doi.org/10.1029/97GL01560

Murphy, D. M., Telg, H., Eck, T. F., Rodriguez, J., Stalin, S. E., & Bates, T. S. (2016). A miniature scanning sun photometer for vertical profiles and mobile platforms. *Aerosol Science and Technology*, *50*(1), 11–16. https://doi.org/10.1080/02786826.2015.1121200

Nakamura, N. (1996). Two-Dimensional Mixing, Edge Formation, and Permeability Diagnosed in an Area Coordinate. *Journal of the Atmospheric Sciences*, *53*(11), 1524–1537. https://doi.org/10.1175/1520-0469(1996)053<1524:TDMEFA>2.0.CO;2

National Academy of Sciences, National Academy of Engineering, & Institute of Medicine. (1992). *Policy Implications of Greenhouse Warming: Mitigation, Adaptation, and the Science Base*. The National Academies Press. https://doi.org/10.17226/1605

National Research Council. (2015). *Climate Intervention: Reflecting Sunlight to Cool Earth*. The National Academies Press. https://doi.org/10.17226/18988

Niemeier, U., & Timmreck, C. (2015). What is the limit of climate engineering by stratospheric injection of SO2? *Atmospheric Chemistry and Physics; Katlenburg-Lindau*, *15*(16), 9129. http://dx.doi.org.ezp-prod1.hul.harvard.edu/10.5194/acp-15-9129-2015

Orsolini, Y., Simon, P., & Cariolle, D. (1995). Filamentation and layering of an idealized tracer by observed winds in the lower stratosphere. *Geophysical Research Letters*, *22*(7), 839–842. https://doi.org/10.1029/95GL00389

Phoenix, D. B., Homeyer, C. R., & Barth, M. C. (2017). Sensitivity of simulated convection-driven stratosphere-troposphere exchange in WRF-Chem to the choice of physical and chemical parameterization. *Earth and Space Science*, *4*(8), 454–471. https://doi.org/10.1002/2017EA000287

Pierce, J., Weisenstein, D., Heckendorn, P., Peter, T., & Keith, D. (2010). Efficient formation of stratospheric aerosol for climate engineering by emission of condensible vapor from aircraft. *Geophysical Research Letters*, *37*. https://doi.org/10.1029/2010GL043975

Pope, F. D., Braesicke, P., Grainger, R. G., Kalberer, M., Watson, I. M., Davidson, P. J., & Cox, R. A. (2012). Stratospheric aerosol particles and solar-radiation management. *Nature*

Climate Change, *2*(10), 713–719. https://doi.org/10.1038/nclimate1528
Prather, M., & Jaffe, A. H. (1990). Global impact of the Antarctic ozone hole: Chemical propagation. *Journal of Geophysical Research: Atmospheres*, *95*(D4), 3473–3492. https://doi.org/10.1029/JD095iD04p03473
Rauthe, M., Gerding, M., & Lübken, F.-J. (2008). Seasonal changes in gravity wave activity measured by lidars at mid-latitudes. *Atmospheric Chemistry and Physics*, *8*(22), 6775–6787. https://doi.org/10.5194/acp-8-6775-2008
Richter, J., H., Tilmes, S., Glanville, A., Kravitz, B., MacMartin, D. G., Mills, M. J., Simpson, I. R., Vitt, F., Tribbia, J. J., & Lamarque, J.-F. (2018). Stratospheric Response in the First Geoengineering Simulation Meeting Multiple Surface Climate Objectives. *Journal of Geophysical Research: Atmospheres*, *123*(11), 5762–5782. https://doi.org/10.1029/2018JD028285
Robock, A. (2000). Volcanic eruptions and climate. *Reviews of Geophysics*, *38*(2), 191–219. https://doi.org/10.1029/1998RG000054
Ross, M. N., Ballenthin, J. O., Gosselin, R. B., Meads, R. F., Zittel, P. F., Benbrook, J. R., & Sheldon, W. R. (1997). In-situ measurement of Cl2 and O3 in a stratospheric solid rocket motor exhaust plume. *Geophysical Research Letters*, *24*(14), 1755–1758. https://doi.org/10.1029/97GL01592
Ross, M. N., Whitefield, P. D., Hagen, D. E., & Hopkins, A. R. (1999). In situ measurement of the aerosol size distribution in stratospheric solid rocket motor exhaust plumes. *Geophysical Research Letters*, *26*(7), 819–822. https://doi.org/10.1029/1999GL900085
Russell, L. M., Rasch, P. J., Mace, G. M., Jackson, R. B., Shepherd, J., Liss, P., Leinen, M., Schimel, D., Vaughan, N. E., Janetos, A. C., Boyd, P. W., Norby, R. J., Caldeira, K., Merikanto, J., Artaxo, P., Melillo, J., & Morgan, M. G. (2012). Ecosystem Impacts of Geoengineering: A Review for Developing a Science Plan. *AMBIO*, *41*(4), 350–369. https://doi.org/10.1007/s13280-012-0258-5
Schneider, A., Gerding, M., & Lübken, F.-J. (2015). Comparing turbulent parameters obtained from LITOS and radiosonde measurements. *Atmospheric Chemistry and Physics*, *15*(4), 2159–2166. https://doi.org/10.5194/acp-15-2159-2015
Schneider, Andreas, Wagner, J., Söder, J., Gerding, M., & Lübken, F.-J. (2017). Case study of wave breaking with high-resolution turbulence measurements with LITOS and WRF simulations. *Atmospheric Chemistry and Physics; Katlenburg-Lindau*, *17*(12), 7941–7954. https://doi.org/10.5194/acp-17-7941-2017
Schoeberl, M. R., & Bacmeister, J. T. (1993). Mixing Processes in the Extra Tropical Stratosphere. In M.-L. Chanin (Ed.), *The Role of the Stratosphere in Global Change* (pp. 135–152). Springer. https://doi.org/10.1007/978-3-642-78306-7_5
Simpson, I. R., Tilmes, S., Richter, J. H., Kravitz, B., MacMartin, D. G., Mills, M. J., Fasullo, J. T., & Pendergrass, A. G. (2019). The Regional Hydroclimate Response to Stratospheric Sulfate Geoengineering and the Role of Stratospheric Heating. *Journal of Geophysical Research: Atmospheres*, *124*(23), 12587–12616.

https://doi.org/10.1029/2019JD031093

Söder, J., Gerding, M., Schneider, A., Dörnbrack, A., Wilms, H., Wagner, J., & Lübken, F.-J. (2019). Evaluation of wake influence on high-resolution balloon-sonde measurements. *Atmospheric Measurement Techniques*, *12*(8), 4191–4210. https://doi.org/10.5194/amt-12-4191-2019

Sparling, L. C., & Bacmeister, J. T. (2001). Scale dependence of tracer microstructure: PDFs, intermittency and the dissipation scale. *Geophysical Research Letters*, *28*(14), 2823–2826. https://doi.org/10.1029/2000GL012781

Stolarski, R. S., & Wesoky, H. L. (1993). *The atmospheric effects of stratospheric aircraft*. NASA. http://ntrs.nasa.gov/search.jsp?R=19930013868

Sukhodolov, T., Sheng, J.-X., Feinberg, A., Luo, B.-P., Peter, T., Revell, L., Stenke, A., Weisenstein, D. K., & Rozanov, E. (2018). Stratospheric aerosol evolution after Pinatubo simulated with a coupled size-resolved aerosol–chemistry–climate model, SOCOL-AERv1.0. *Geoscientific Model Development*, *11*(7), 2633–2647. https://doi.org/10.5194/gmd-11-2633-2018

Teller, E., Wood, L., & Hyde, R. (1996). *Global warming and ice ages: I. prospects for physics based modulation of global change* (UCRL-JC-128715; CONF-9708117-). Lawrence Livermore National Lab., CA (United States). https://www.osti.gov/biblio/611779-global-warming-ice-ages-prospects-physics-based-modulation-global-change

Theuerkauf, A., Gerding, M., & Lübken, F.-J. (2011). LITOS – a new balloon-borne instrument for fine-scale turbulence soundings in the stratosphere. *Atmospheric Measurement Techniques*, *4*(1), 55–66. https://doi.org/10.5194/amt-4-55-2011

Tilmes, S., Fasullo, J., Lamarque, J.-F., Marsh, D. R., Mills, M., Alterskjær, K., Muri, H., Kristjánsson, J. E., Boucher, O., Schulz, M., Cole, J. N. S., Curry, C. L., Jones, A., Haywood, J., Irvine, P. J., Ji, D., Moore, J. C., Karam, D. B., Kravitz, B., … Watanabe, S. (2013). The hydrological impact of geoengineering in the Geoengineering Model Intercomparison Project (GeoMIP). *Journal of Geophysical Research: Atmospheres*, *118*(19), 11,036-11,058. https://doi.org/10.1002/jgrd.50868

Tilmes, S., Richter, J. H., Kravitz, B., MacMartin, D. G., Mills, M. J., Simpson, I. R., Glanville, A. S., Fasullo, J. T., Phillips, A. S., Lamarque, J.-F., Tribbia, J., Edwards, J., Mickelson, S., & Ghosh, S. (2018). CESM1(WACCM) Stratospheric Aerosol Geoengineering Large Ensemble Project. *Bulletin of the American Meteorological Society*, *99*(11), 2361–2371. https://doi.org/10.1175/BAMS-D-17-0267.1

Torres, B., Dubovik, O., Toledano, C., Berjon, A., Cachorro, V. E., Lapyonok, T., Litvinov, P., & Goloub, P. (2014). Sensitivity of aerosol retrieval to geometrical configuration of ground-based sun/sky radiometer observations. *Atmospheric Chemistry and Physics*, *14*(2), 847–875. https://doi.org/10.5194/acp-14-847-2014

Vanneste, J. (2004). Small-Scale Mixing, Large-Scale Advection, and Stratospheric Tracer Distributions. *Journal of the Atmospheric Sciences; Boston*, *61*(22), 2749–2761.

Vattioni, S., Weisenstein, D., Keith, D., Feinberg, A., Peter, T., & Stenke, A. (2019). Exploring accumulation-mode H2SO4 versus SO2 stratospheric sulfate geoengineering in a sectional aerosol–chemistry–climate model. *Atmospheric Chemistry and Physics*, *19*.

Vehkamäki, H., Kulmala, M., Napari, I., Lehtinen, K. E. J., Timmreck, C., Noppel, M., & Laaksonen, A. (2002). An improved parameterization for sulfuric acid–water nucleation rates for tropospheric and stratospheric conditions. *Journal of Geophysical Research: Atmospheres*, *107*(D22), AAC 3-1-AAC 3-10. https://doi.org/10.1029/2002JD002184

Voigt, C., Schumann, U., Graf, K., & Gottschaldt, K.-D. (2013). Impact of rocket exhaust plumes on atmospheric composition and climate — an overview. *Progress in Propulsion Physics*, *4*, 657–670. https://doi.org/10.1051/eucass/201304657

Weisenstein, D., Keith, D., & Dykema, J. (2015). Solar geoengineering using solid aerosol in the stratosphere. *Atmospheric Chemistry and Physics*, *15*, 11835–11859. https://doi.org/10.5194/acp-15-11835-2015

World Meteorological Organization, United States, National Oceanic and Atmospheric Administration, United States, National Aeronautics and Space Administration, United Nations Environment Programme, & European Commission. (2019). *Scientific assessment of ozone depletion: 2018.*

Yu, F., & Turco, R. P. (1997). The role of ions in the formation and evolution of particles in aircraft plumes. *Geophysical Research Letters*, *24*(15), 1927–1930. https://doi.org/10.1029/97GL01822

Supplemental Materials

Dai, Z., F. Keutsch, D. Weisenstein, and D. W. Keith. (2020). Experimental reaction rates constrain estimates of ozone response to calcium carbonate geoengineering, *Manuscript submitted for publication.*

Dykema, J., Keith, D., Anderson, J. G., & Weisenstein, D. (2014). Stratospheric controlled perturbation experiment (SCoPEx): A small-scale experiment to improve understanding of the risks of solar geoengineering. *Philosophical Transactions of the Royal Society A*, *372*. https://doi.org/10.1098/rsta.2014.0059

Dykema, J., Keith, D., & Keutsch, F. (2016). Improved aerosol radiative properties as a foundation for solar geoengineering risk assessment. *Geophysical Research Letters*. http://onlinelibrary.wiley.com/doi/10.1002/2016GL069258/full

Floerchinger, C., J. Dykema, D. Keith, F. Keutsch. (2020). A Need For In Situ Observations to Inform Nearfield Plume Transport and Aerosol Dynamics as well as Chemistry of Alternate Geoengineering Materials in the Stratosphere. *Letter to the National Academy for Science.*

Golja, C. M., L. W. Chew, J. A. Dykema, and D. W. Keith. (2020). Aerosol Dynamics in the Near Field of the SCoPEx Stratospheric Balloon Experiment, *Manuscript submitted for publication.*

Keith, D., F. Keutsch, C. Floerchinger. (2020). Empirical methods to reduce uncertainty about solar geoengineering. *Submitted as public input to the National Academy Committee on Climate Intervention Strategies that Reflect Sunlight to Cool Earth.*

Appendix C-3

Advisory Committee Response to Research Team Request

Review of the Engineering Integrity and Safety of SCoPEx

Obligations/Issues for the Advisory Committee

SCoPEx has an obligation to submit a 'Risk Management'[1] to be reviewed by the AC. In Frank's letter of November 10th, SCoPEx is requesting (a) *review* and (b) *authorization* of the test scheduled for June 21st in Sweden.

The AC considers that it would be imprudent to authorize the test without a '*technical soundness review*' of the platform, as part of its due diligence. Further, such review could not be objectively undertaken in the absence of certain engineering details. Consequently, we welcome your timely response to the following:

1. Has SCoPEx research team identified any *potential risks and/or matters relating to safety* associated with the engineering flight?
2. Has SCoPEx undertaken a risk assessment of the engineering launch? If affirmative, we are requesting a copy of same and a proposed mitigation plan.
3. Please describe the process by which the balloon and gondola will be returned safely and intact to the ground following completion of the engineering test.
4. Can Swedish Space Corporation *safely abort the launch without posing a danger to people and structures on the ground and retrieve the gondola,* in the unlikely event that something appears to be going wrong, or has actually gone wrong?
5. What degree of control does Swedish Space Corporation have over times when and the locations where the balloon and gondola return to land? Are there particular areas where this usually occurs?
6. Does the gondola crash land? Is there a risk that if it does, batteries or other equipment will ignite?
7. Has Swedish Space Corporation experienced any incidents where its balloons or gondolas have caused damage or injury on the ground?
8. Please expand on other potential fire hazards posed by the battery powering the balloon.
9. Can you outline the specific potential risks/safety issues associated with each phase of the operation, i.e. during launch/ascent, descent and retrieval?
10. Are there factors in the April 2010 balloon launch accident in Alice Springs, Australia that we should be concerned about for the upcoming planned launch? Specifically, are there lessons worth noting that may be relevant to SCoPEx? For example, NASA's own accident report noted the following, *inter alia*:

    i. Weather conditions were acceptable for launch and there were no technical problems BUT
    ii. "..in the course of our investigation, we found surprisingly few documented procedures for balloon launches".
    iii. "No one considered the launch phase to be a potential hazard."
    iv. There were some 25 causes identified as potential reasons for the accident, including "…insufficient risk analysis, government oversight and public safety issues".

Background Information Extracted from Technical Document:

[1] Refer to document 'Scientific and Technical Merit Review Process'.

- “SCoPEx focuses primarily on improving understanding of the first-order impacts in the stratosphere, i.e., risks and risk reduction associated with impacts of SAI within the stratosphere".
- The technical document describes the role/purpose of each element of the gondola, but (intentionally) omits the engineering details of these components.
- The technical document describes the ascent phase of the launch but does not describe how the balloon and gondola will be returned safely to the ground.
- The gondola has three primary features: the frame, the ascender, and (off the shelf, repurposed airboat) propellers.
- The gondola will be powered by 28 V and 100 V DC power supplies which will power all operations on the platform including the propellers, ascender and instruments.
- Total mass (frame, all subsystems, hopper with ballast) is 600 kg .
- Platform Test Flight will be conducted to test the *durability and maneuverability* of the gondola.

Appendix C-4

Research Team Response to Questions from Advisory Committee

HARVARD
School of Engineering
and Applied Sciences

Frank N. Keutsch
Stonington Professor of Engineering and Atmospheric Science
Professor of Chemistry and Chemical Biology
keutsch@seas.harvard.edu

Harvard John A. Paulson School of Engineering and Applied Sciences
Department of Chemistry and Chemical Biology
Department of Earth and Planetary Sciences
12 Oxford Street, Cambridge, MA 02138 USA
Phone: 617-495-1878 / Fax: 617-495-4902

January 17, 2021

Dear SCoPEx Advisory Committee,

In response to your written request, we submit a document that provides the details you have requested for the technical soundness review of the platform test. These responses were written in consultation with Swedish Space Corporation.

We appreciate the Committee's review of the platform test's engineering integrity and safety and will be happy to provide additional information on request.

Sincerely,

Frank Keutsch
Stonington Professor of Engineering and Atmospheric Science
Harvard John A. Paulson School of Engineering and Applied Sciences
Department of Chemistry and Chemical Biology
Department of Earth and Planetary Sciences

Responses to Review of the Engineering Integrity and Safety of SCoPEx

Question 1. Has SCoPEx research team identified any *potential risks and/or matters relating to safety* associated with the engineering flight?

Response: The SCoPEx research team has identified the following potential physical risks associated with building, testing, and transporting equipment and personnel to the engineering flight:

- Personnel traveling (car crashes, illness)
- Lifting and moving heavy objects (load crushing injuries, falls, physical strain)
- Batteries and electronics (electrocution, explosion, burn)
- Propellers (strike risk from improper operation)
- Crane (load crushing injuries, structure tipping).

The launch and flight services, systems and materials for the launch (helium gas, flight train, parachute), and payload recovery by helicopter will all be provided by Swedish Space Corporation (SSC). Risk management associated with these aspects of the flight will fall under their purview.

Question 2. Has SCoPEx undertaken a risk assessment of the engineering launch? If affirmative, we are requesting a copy of same and a proposed mitigation plan.

Response: No, but we are working with Harvard Environmental Health & Safety to conduct a job hazard analysis to evaluate and manage the risks detailed in the bullet point list in question 1. We would be happy to share hazard analysis with the committee when it is available.

Question 3. Please describe the process by which the balloon and gondola will be returned safely and intact to the ground following completion of the engineering test.

Response: After a sufficient float period is acquired, the termination of the flight will be performed. The payload and flight systems will be separated from the balloon envelope and descend to the ground with a parachute designed to maintain a decent velocity of ~ 4-6 m/s. The balloon envelope will return to the ground separately. SSC will provide recovery of the payload. Recovery will be performed after the flight using a helicopter. The payload will normally be back at Esrange within 24 hours.

Question 4. Can Swedish Space Corporation *safely abort the launch without posing a danger to people and structures on the ground and retrieve the gondola,* in the unlikely event that something appears to be going wrong, or has actually gone wrong?

Response: The launch will take place at Esrange Space Center, which is a restricted site for third persons. Thus, problems during a launch attempt would be kept in an area where no third persons are present. The personnel involved in the launch are positioned so they will not be harmed if something goes wrong.

The balloon launch will take place at the Balloon Launch Area at Esrange Space Center, which is surrounded by infrastructure at the facility. Though it is highly unlikely that something would hit any buildings or equipment on the base, it is not impossible. All infrastructure is insured in case something happens. SSC also has third party insurance in the highly unlikely case that any third party would be hurt during any phase of the balloon operation.

The recoverability of the gondola is dependent on the type of launch failure. If, for example, the balloon and gondola has been launched and is flying at a low altitude, and then a balloon burst occurs, the parachute may not have time to inflate and decrease the landing speed of the payload resulting in substantial damage to the gondola. If the balloon bursts on the spool the gondola is secure on the launch vehicle. If the balloon has a problem at higher altitude the gondola would descend with the parachute.

As the precise landing spot cannot be determined, due to wind drift of the parachute in the end, there may be damage to the gondola when it lands.

Question 5. What degree of control does Swedish Space Corporation have over times when and the locations where the balloon and gondola return to land? Are there particular areas where this usually occurs?

Response: SSC will command the cut-down of the gondola and balloon via radio link so that is done to a high degree of control. The landing spot for the gondola will first be predicted with the help of a trajectory analysis looking at the winds before the launch. After launch, and during the flight, the actual position and thus the predicted landing spot will be continuously monitored. However, the exact landing spot will not be known due to deviation between forecast and real wind. We expect the real landing spot to be within some kilometers of the predicted landing spot.

Question 6. Does the gondola crash land? Is there a risk that if it does, batteries or other equipment will ignite?

Response: The gondola descends to the ground via a parachute and will have a velocity of 4-6 m/s. Crush pads mounted under the legs of the payload are designed to decrease the shock of the landing impact. The structure has been designed to withstand a 10 g load of the full payload mass (600 kg) even under conditions were only one leg makes initial contact with the ground.

The batteries are mounted at the center of the lower deck away from edges of the platform such that they will not experience a direct impact with the ground. We anticipate the batteries will have expended ~ 75% of their stored energy prior to initiating descent. We further anticipate that the containment method used to house the batteries will safely isolate them from the landing shock even in the event of a crash landing. The batteries will be housed in an array of boxes and strapped down with a cargo net. Each insolated metal battery box will provide additional isolation from landing shock and the other batteries.

The cell chemistry is Lithium Nickel Manganese Cobalt (LiNiMnCo). This chemistry was selected after evaluating energy, power, and safety consideration of various battery chemistries. An overview and more information on various Lithium-ion battery technologies can be found in

Miao et al, Energies 2019, 12(6), in particular, Figure 4 present a comparison of several different chemistries.

The cells are from AA Portable Power Corp INR-26650-5000 and we are using package model numbers PR-CU-R635-14S3P and PR-CU-R635-8S3P housed inside aluminum cans with an electronic monitoring system. The engineering flight will carry enough power, with some additional overhead, to complete the mission. It will not carry the full energy capacity required for a science flight. Thermal profiling of the battery system under full load discharge rates over a time period consistent with the engineering flight plan indicates the batteries will remain well within their temperature ratings throughout the flight. During ascent and descent, the batteries will be electrically isolated from their loads (with the exception of the flight computer and ground communication radio).

Besides the batteries there are no additional energy sources on the payload.

Question 7. Has Swedish Space Corporation experienced any incidents where its balloons or gondolas have caused damage or injury on the ground?

Response: No, during the more than 200 launches that SSC has performed, there has been no damage or injury either during launch, flight, or landing.

Question 8. Please expand on other potential fire hazards posed by the battery powering the balloon.

Response: Thermal runaway is the main fire hazard. Other potential fire hazards could be overheating of payload components and should be mitigated by the monitoring system. A 2017 FAA report discusses the fire hazard of lithium batteries. Figure 13 shows the onset of thermal runaway from LiNiMnCo (C-long-sized) cells to be 200 °C. Our preliminary laboratory tests indicate during max loading the max temperature of the battery pack is 82 °C. The report found that LiNiMnCo is a moderate battery choice. The LiNiMnCo batteries were more likely to have the cell eject its contents which prevents heat propagation between cells, but this result is dependent on how the cells are packaged together. Our packaging and mounting configuration will aid in compartmentalizing battery cells and reduce damage propagation in the event of damage.

Question 9. Can you outline the specific potential risks/safety issues associated with each phase of the operation, i.e., during launch/ascent, descent and retrieval?

Response:

At launch: Unexpected wind direction or speed change; faulty balloon; pressurized gases and system; ESD; on-base radio interference

During ascent: Faulty balloon (hole in envelope); too much or little lifting gas

At float: Wind prognosis not accurate (trajectory difference)

Cut-down and landing: Planned landing spot in an area with safety concerns; cut-down does not execute as expected; wind drift under parachute higher than anticipated; gondola structure not strong enough for forces during cut-down

Question 10. Are there factors in the April 2010 balloon launch accident in Alice Springs, Australia that we should be concerned about for the upcoming planned launch? Specifically, are there lessons worth noting that may be relevant to SCoPEx? For example, NASA's own accident report noted the following, *inter alia*:

i. **Weather conditions were acceptable for launch and there were no technical problems BUT**
ii. **"...in the course of our investigation, we found surprisingly few documented procedures for balloon launches".**
iii. **"No one considered the launch phase to be a potential hazard."**
iv. **There were some 25 causes identified as potential reasons for the accident, including "…insufficient risk analysis, government oversight and public safety issues".**

Response: As it happens, Keutsch, Keith, and Dykema of the SCoPEx team heard details of this accident at one of the very first meetings we had with a NASA balloon expert, long before we started talking to commercial balloon operators. Our impression is that the Alice Springs accident has encouraged balloon operators to rethink launch risks and improve procedures.

We are satisfied that SSC has had full access to the NASA investigation regarding the 2010 Alice Springs accident, and it has been discussed internally at SSC and together with NASA safety to reduce the chance of similar accidents at Esrange. For example, the mechanical safety system at SSC has been changed due to the incident. In addition, SSC has its certified procedures for balloon launches (ISO 9001:2000).

Flight Platform Test

Context: SCoPEx will use a new flight platform that has not flown before. There are significant technical challenges in developing it as an operational vehicle independent of the challenges of the actual solar geoengineering experiment. Only a few propelled balloon systems have flown in the stratosphere. SWRI's HiSentinel and High Altitude Airship were both high velocity (>15 m/sec) airships. Balloon operators have told us about a few other simple gondolas with propellers that are analogous to SCoPEx but we do not have access to details of those systems.

Flight Platform Test will test the capabilities of this new platform to perform future outdoor solar geoengineering experiments but will not carry systems for releasing or measuring particles. Instead, it is a development test flight aimed at verifying operation of the platform, controls, and communications.

Harvard will operate the gondola which will hang from a zero-pressure stratospheric balloon with an approximate volume of 350K cu ft (90 ft inflated height and 90 ft inflated diameter). A balloon vendor will be responsible for all launch, flight, termination, and recovery operations. The flight platform test will have a float duration of 4-6 hours at an altitude of 65000 ft.

Objective: To test the flight platform (Figure 1) and subsystems for powering, running, and communicating with the flight platform. A successful platform test means demonstrating:

(a) Heading control stability
(b) The ability to drive at the design velocity of 3 m/s and to cover >3 km of distance
(c) The ability to fly a path such as given in Figure 2, and to fly through an arbitrary reference point with accuracy of 10 m
(d) The ability to ascend and descend the rope system as commanded, and to hold an absolute altitude within 10 m over 30 minutes
(e) Reliable operation of the flight computer and power systems with component temperatures not going beyond their limits.

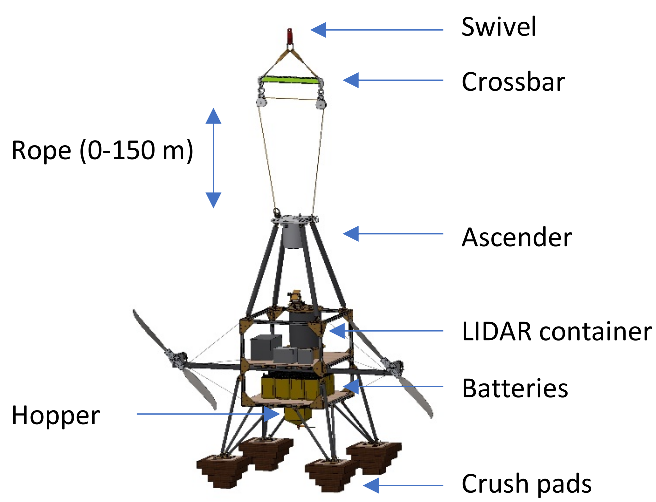


Figure 1: A representation of the flight platform. The final configuration may have subsystem packaged differently.

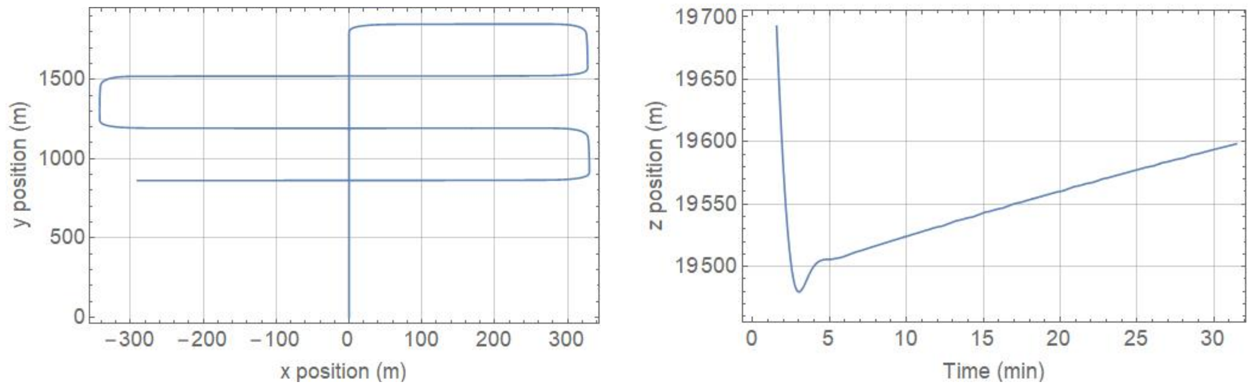


Figure 2: (left) A top down view of the proposed flight maneuvers over a 35-minute window. x and y are in the horizontal plane. (right) The vertical position expected without any ascender or hopper vertical trimming over the same 35-minute platform maneuver.

Systems included:

System	**Description**
Gondola	An aluminum and carbon fiber frame with a ballast hopper for coarse altitude control. The hopper hardware and communication will be under Harvard control and the actions will be managed by the balloon operator. Total mass of frame, all mounted systems, and hopper: 600 kg
Ascender	For fine altitude control using an ascender built by Atlas Devices. 10 mm diameter, 300+ m length rope. Range of motion: 0-150 m. Max speed: 10 m/min
Propulsion	For repositioning the payload. Max airspeed: 3 m/s Twin propellers from Sensenich, 1.88 m diameter, 1500 RPM, 32 N each MDM-5000 brushless servo motors from Montevideo Technology 1543 W rated power, 12 NM peak torque
Power	28 V and 100 V DC power supplies will power all systems on the Harvard payload. Each 28 V (100 V) battery box has a capacity of 60 Ah (15 Ah). There will be 4 (2) battery boxes for the 28 V (100 V) supply giving a total energy of 24 MJ (10 MJ) when fully charged.
Flight computer	Raspberry Pi 4 will run an interrupt driven real time software architecture that has been developed through many Harvard atmospheric research missions. The flight computer will receive commands, supervise onboard components, log data, and run an algorithm to operate the propellers.
Communication	2 Global star GSP-1700 satellite phones, 9600 bps will facilitate communication between ground equipment and the payload. Ground control software will display incoming data from the payload and communicate with the payload.
Wind	A Styrofoam sphere wind pendulum which takes data using a Raspberry Pi camera will be used to collect data on relative winds.
LIDAR Container	The instrument container will fly with no instrument inside. A resistive heating element will be placed inside, and heat load data will be collected

Subsystems not a part of the platform test: Sprayer, POPS, LIDAR and other scientific instruments

Appendix C-5

Reviewer Feedback and Responses

Technical Review Feedback and Responses

To protect the integrity of the review and encourage candid feedback on the research plan, we decided to keep reviewers' comments confidential between the Advisory Committee and Research Team. We have therefore summarized the major themes along with responses from the SCoPEx Research Team that captures all of the feedback we received.

Weather Conditions

What are the limits on the weather conditions at altitude for launching the balloon for safe operation? Can you provide a wind profile and bounds that allow a successful test?

Response from SCoPEx Research Team: The trajectory analysis that SSC conducted was very promising. They analyzed data from 2018 and 2019 and did not find any days in June that would prohibit a launch. While some days provided <6 hours of flight time, there were no days with winds unsuitable for launch. Certain wind conditions may mean that we do not achieve the desired float time. However, even a 4-hour flight would provide significant time to test the platform. Our two-week launch window should provide ample opportunities for appropriate weather and launch conditions. SSC is responsible for all safety aspects of evaluating the weather and launch conditions.

The wind and relative wind shear between the balloon and platform will affect the repositioning abilities of the payload. Since the platform test aims to test the maneuvering capabilities of the platform and it will not be searching for a plume, the absolute position of the payload is not a determining factor in the success of the engineering flight. The goal of the platform test is to demonstrate the capabilities of the platform. Wind data from this first flight will inform future flights and future flight plans. These requirements will be narrower for future science flights. Initial models have shown that the propellers can move the payload-balloon system up to 3 m/s horizontally, at an altitude of 65,000 ft, under wind conditions typically encountered in flight and based on historical data.

Potential for Human Error

Human operational or preparatory errors can happen. What measures will you take to reduce the risk of human error?

Response: We concur. There is no way to eliminate such errors, but we believe there are some institutional practices that make them less likely. We look to lessons from high-rise liability organizations drawn from the work of Gene Rochlin and collaborators. In particular, we will focus on making sure that everyone feels empowered to speak out when they see problems and on review processes that assume accidents will happen.

Altitude Control

How will the research team maintain a relatively level flight during the test period?

Response: The initial Statement of Work proposal specified an altitude of 65,000 ft. While this is a target altitude for science flights, it is not a firm requirement for the platform test. Our altitude requirements can be relaxed for the engineering flight. The platform will test the capabilities of the ascender, propulsion system, navigation system, power system, and flight computer, which can all be accomplished at higher altitudes. The ultimate target altitude for the flight will be selected by SSC, taking into account their flight train design. This is a part of the flight services provided by SSC and they have a strong and successful track record. Speed and altitude data will be available from our GNSS unit. The unit was selected for its high precision and accuracy. The GNSS unit and communication system data rate are sufficient for this application.

For the science flights, the altitude will be controlled via a vent valve on the balloon and a ballast hopper. To simplify the system for the platform test, the first flight will be conducted without the vent valve at the top of the balloon. This limits the achievable altitude precision on this flight. Improved altitude control is a technical specification that will be developed in future flights.

Propulsion

Are the propellers for this flight selected and tuned for this pressure altitude?

Response: The propellers were selected and optimized for 65,000 ft, however, they can be run at atmospheric pressure (at a lower RPM) or at higher altitudes (with lower thrust). 65,000 ft is a target for the science flights but this does not preclude the platform from safely operating at other altitudes. The feedback loop will have to be tuned for different altitudes. We have modeled propeller performance with a computational fluid dynamics study, which provides initial input to tune the feedback loop.

Ascender

Previous flights of this kind have experienced issues with the ascender mechanism when exposed to significant changes in temperature and pressure, leading to issues retracting the mechanism before termination. How will the research team prevent the ascender from getting stuck if the payload hangs lower than intended from the balloon or twists in the event of early termination? How can the balloon land safely in this event scenario?

Response: The SCoPEx team worked with Atlas Devices to identify an existing ascender unit that could be modified for stratospheric operation. It is rated for a 10 g termination load and has been modified to operate under stratospheric conditions. It has been specified to lift our 600 kg payload, survive the parachute shock load at termination, and have an operating temperature range of -80 C to 50 C. Additional testing of the structure can be conducted in our cold, low-pressure chamber. The double rope path was selected in order to provide a mechanical advantage and keep the forces under the load limit for the standard Atlas unit. A spreader bar was added to the design to minimize the chance of the rope twisting up. We judged that this was a lower overall program risk than working with Atlas to develop a new design that could handle the full 600 kg payload on one rope.

As part of the concept of operations the payload will be retracted before the flight is terminated. We agree that one risk is that the payload does not retract properly. This could be due to the system twisting up or an issue with the ascender. We have discussed the possible need to terminate the flight while the payload is fully extended. While this is not desired, SSC has not found any safety issues with this configuration.

If at termination the payload is hanging lower than intended, no attempt will be made to adjust the position at the last minute. The flight could be terminated with the payload in the fully or partially extended position for two reasons: early termination means there is not enough time to retract the payload or there is a concern with the performance of the rope or ascender and the SCoPEx team chooses not to retract the payload.

If the payload descends in the extended position, the gondola will land a bit before the parachute. As the extended rope and then parachute reach the ground, there is a chance for the payload to be dragged by the parachute due to the prevailing winds. Dragging could damage pieces of the structure, particularly the propellers and booms extending from the structure. However, due to the forested areas of northern Scandinavia, the payload is more likely to be caught in a tree which would prevent it from being dragged along the ground.

Materials

What considerations have been made for ensuring proper operation of the mechanical features of the mechanisms? Are the cables, grease, gear systems, pumps, motors, and bearings tested to handle extreme temperatures and higher pressure? What is the rationale for using composite landing legs?

Response: The ascender rope is a static rope made from Technora, an aramid fiber with high strength and chemical resistance properties. It also has appropriate thermal properties. A variety of ropes have been tested with the ascender and several were ruled out because they were not stiff enough or the sheath did not work well with the ascender jaws.

Care has been taken to select components, grease, and epoxies appropriate for the cold flight temperatures. Specifically, the contract with Atlas devices is for a specially modified ascender for a stratospheric flight. Additional testing of specific components can be conducted in our cold, low-pressure chamber or sent out to testing facilities.

The only pump on the payload will be a part of the POPS (Portable Optical Particle Spectrometer), a light-weight instruments which was designed with aerial platforms in mind. Any failure from this pump would not pose a safety risk to the rest of the payload.

Bonding was selected because machining carbon fiber components is both difficult and weakens the material. Instead, the attachment points were selected to be bonded ferrules. The specific epoxy was selected for its strength and cold temperature properties.

Carbon fiber was selected for the landing legs in order to build a lighter weight structure. The modular design of the payload means that any carbon fiber components damaged upon landing can be replaced before the next flight. The propellers and legs are considered to be consumable parts.

Appendix D

Scientific Merit Review Documents

Appendix D-1

Revised Research Plan

The Stratospheric Controlled Perturbation Experiment (SCoPEx)

Version 2.0

Executive Summary 2
1. Introduction 3
2. Observational SAI Research Needs 5
3. SCoPEx Short Overview 11
4. SCoPEx Goals 18
5. Data Management Plan and Dissemination of Results 30
6. SCoPEx Research Team Biographies 31
References 34

Executive Summary

Climate model studies of stratospheric solar radiation modification (SRM) depend, perhaps implicitly, on processes that take place in the near field of an injection plume. This is because materials delivered to the stratosphere by aircraft will form persistent, high aspect-ratio plumes with strong gradients before becoming well mixed, and processes within the plume will alter the large-scale, well-mixed aerosol and chemical properties that are simulated in global atmospheric models. All models ultimately depend on observations, yet we lack experimental data to assess some of the critical transport, microphysical, and chemical processes that directly control aerosol dynamics in the near-field that are important for understanding stratospheric SRM.

The scientific goal of the Stratospheric Controlled Perturbation Experiment (SCoPEx) is to improve process models that will, in turn, reduce uncertainties in global-scale models, thus reducing uncertainty in predictions of important SRM risks and benefits.

SCoPEx addresses questions in stratospheric aerosol injection (SAI) research that observations of existing analogues are incapable of addressing. For example, existing observational data do not include chemistry of alternate geoengineering materials specific to SAI, near-field particle microphysics of injection plumes, and relevant scales of atmospheric transport in the near-field. Yet these are needed to assess processes that control aerosol dynamics in the near field of an injection plume and that allow for the evaluation of alternate SAI materials, i.e., materials other than the naturally existing sulfate aerosol.

We first review why existing observations do not address the questions that SCoPEx will answer. We then give a description of the basic design of the platform and the concept of operations of SCoPEx. Finally, we describe the three specific science goals of SCoPEx, explain how they represent critical knowledge gaps in SAI research, and specify what measurements are needed to enable SCoPEx to provide quantitative answers to these questions. The three specific science goals are improving understanding of (i) turbulent mixing scales, (ii) aerosol microphysics with a focus on alternative SAI materials in the near-field of an injection, and (iii) process level chemical interactions of alternative SAI materials in the stratosphere.

We do not provide a detailed engineering document of the SCoPEx platform or its scientific instrumentation, nor do we provide a justification for the need for research on SRM via SAI in general. Rather, we focus specifically on the merits of SCoPEx itself.

1. Introduction

In this document we focus on the motivation and scientific merit of SCoPEx. We do not provide detailed engineering documentation of the SCoPEx platform or its scientific instrumentation. We also do not provide general justification for the need for research on solar radiation modification (SRM) via stratospheric aerosol injection (SAI), which can be found in many prior documents such as the 1992 NAS report that recommended the US government "Undertake research and development projects to improve our understanding of both the potential of geoengineering options to offset global warming and their possible side effects. This is not a recommendation that geoengineering options be undertaken at this time, but rather that we learn more about their likely advantages and disadvantages" (National Academy of Sciences et al., 1992) or the recent 2015 and 2021 NAS reports (National Research Council, 2015; National Academies of Science, Engineering, and Medicine, 2021) with the latter explicitly recommending small-scale field experiments under specific conditions. Rather, we focus specifically on the need for small-scale field experiments such as SCoPEx, and the specific, critical SAI research needs that will be addressed by SCoPEx.

1.1. Role of and Need for Small-Scale Field Experiments

There is a vast array of science and engineering questions that have to be answered to achieve a better understanding of the risks, benefits and feasibility of SAI. The tools and topics that are needed to address these questions range from General Circulation Models (GCMs) all the way to detailed design of instrumentation to monitor or disperse aerosol. SCoPEx addresses a subset of questions that require small-scale field experiments for ground-truthing and that are aimed at improving the ability of models to predict the consequences of SAI.

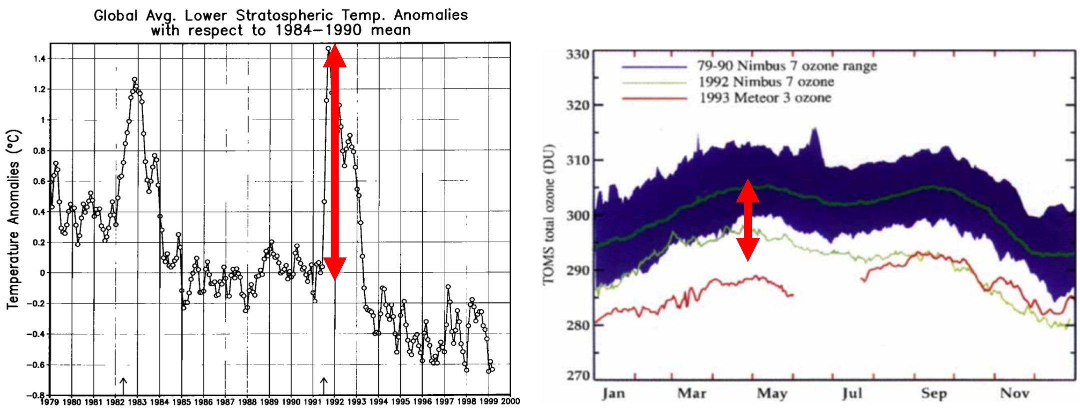


Figure 1: The two most important first-order stratospheric risks from sulfate SAI. The left panel shows stratospheric temperature anomalies from the El Chichon and Mount Pinatubo eruptions on top of background temperatures that are decreasing due to greenhouse gas emissions (Robock, 2000). The dynamical response of the stratosphere from such a short heating pulse likely is different than from sustained heating from longer-term SAI. The right panel shows that in the two years following the Mount Pinatubo reaction total ozone columns were lower than in the 1979-90 average as a result of increase sulfate aerosol surface area. Smaller eruptions also contributed to this. (McCormick et al., 1995)

There are numerous known risks associated with SAI, and SCoPEx focuses primarily on improving understanding of the first-order impacts in the stratosphere, i.e., risks and risk reduction associated with impacts of SAI within the stratosphere. There are many downstream / higher-order risks, e.g., impact on cloud formation as SAI particles leave the stratosphere (Cziczo et al., 2019), impacts on ecosystems via changes in the hydrological cycle (Bala et al., 2008; Russell et al., 2012; Tilmes et al., 2013), or the amount of direct versus diffuse radiation (Gu et al., 2002; Farquhar & Roderick, 2003; Gu et al., 2003). Despite their importance, these impacts are not the direct target of this proposal although many of these are also influenced by stratospheric processes and properties of SAI aerosol. Two first-order risks are at the focus of this work: stratospheric ozone loss and the dynamic response resulting from stratospheric heating as a result of SAI.

Whereas stratospheric ozone chemistry is fairly well understood (World Meteorological Organization, 2019), there are still substantial uncertainties in the understanding and ability to model stratospheric dynamics (Figure 1). For example, models have only recently been able to reproduce the quasi-biennial oscillation without having it imposed (see Butchart et al., 2018 for a discussion of challenges). One approach taken in this work is to evaluate whether there are types of aerosols or methods of aerosol injection that can reduce first-order risks for a given amount of radiative forcing. It stands to reason that a reduction in the first-order stratospheric impacts will reduce downstream and higher-order risks. A case in point is the growing body of work that has been investigating the impacts of stratospheric heating on stratospheric water vapor and the dynamic response on regional climate (Simpson et al., 2019; Ferraro et al., 2015; Richter et al., 2018; Ji et al., 2018). It is important to note that the amount of stratospheric heating for a given material will be primarily driven by the total mass of aerosol, ozone destruction will be driven by the total surface area of aerosol, and the desired radiative forcing will be determined by the amount and size distribution of aerosol. Critically, both the aerosol mass required for a given desired radiative forcing *and* the resulting surface area are tied to this size distribution. Therefore, accurate models of the evolution of the size distribution of injected aerosol are critically needed. In addition, alternate materials with reduced stratospheric heating have to be investigated, as do injection methods for sulfate that minimize stratospheric heating and ozone loss for a given radiative forcing, as this will reduce risks associated with the dynamic response to this first-order perturbation.

2. Observational SAI Research Needs

Most of the rapidly growing body of literature on SAI rests on General Circulation Models (GCMs). We acknowledge the importance of GCM studies, but in the following we focus on research needs that require experiments and observations, and especially questions that can only be answered by conducting perturbative field experiments such as SCoPEx (see supplemental manuscripts Keith et al., 2020 and Floerchinger et al., 2020). In fact, SCoPEx will in the end inform GCMs by providing improved process level information that will be integrated in parameterizations used in GCMs. Below we review existing observational data sets and describe their utility for different SAI approaches, highlighting where they are unable to shed light on critical issues thus motivating studies like SCoPEx.

2.1. Field Experimental Needs for Sulfate SAI

Most studies that have sought to research SAI have assumed the addition of aerosol would take place by means of an injection of gas-phase SO_2, which is ultimately converted to H_2SO_4 and then to sulfate aerosol in the stratosphere on a timescale of approximately one month. The aerosol size distribution from this injection of gas phase precursor must be accurately predicted as it will control the shortwave (SW) scattering properties, the stratospheric lifetime of the aerosol, and ultimately be the driver for the radiative forcing (RF) efficiency per mass of injected sulfate. Some studies, such as Niemeier & Timmreck (2015), have suggested that with higher injection rates of SO_2, the resulting sulfate aerosol would be forced into a larger, coarse-mode size distribution and functionally reach a point of diminishing return. In this diminishing return scenario, the added amount of SW RF achieved per added mass of sulfate decreases exponentially.

Recent work by Pierce et al. (2010), Benduhn et al. (2016), and Vattioni et al. (2019) has highlighted the potential benefits of injecting H_2SO_4 aerosol directly into the accumulation mode (AM), i.e., aerosols with a radius of 0.1–1.0 µm, potentially by emitting H_2SO_4 vapor into an aircraft plume. This work has suggested better control of the resulting aerosol size distribution and thus the radiative forcing per unit mass sulfur injection, which would allow for the design of a system that maximizes the radiative forcing per mass of sulfate in a way that would not have the diminishing returns at high SO_2 injection rates. This would thus minimize the increase in the stratospheric sulfate burden and hence the risk of stratospheric heating which is driven by total mass whereas ozone loss is driven by surface area. While injecting AM–H_2SO_4 may represent the best possible approach for SAI with stratospheric sulfate, there is currently no proven way to introduce vapor phase AM–H_2SO_4 into the stratosphere. As AM–H_2SO_4 has not been studied, perturbative experiments are required to provide observational constraints on the aerosol size distributions predicted by models.

2.2. Field Experimental Needs for Alternate Aerosol Material SAI

Though sulfate aerosol does exist in the background stratosphere and there are some natural analogs of broad stratospheric sulfate injections (volcanic eruptions), it likely is not the optimal candidate for SAI. Alternative aerosol may be most appropriate in order to mitigate SAI risks (Teller et al., 1996; Crutzen, 2006; Ferraro et al., 2011; Ferraro et al., 2015; Weisenstein et al., 2015; Keith et al., 2016; Dykema et al., 2016; Weisenstein et al., 2015). These alternate aerosols could reduce the previously noted two major first-order stratospheric impacts, i.e., changes in ozone and especially stratospheric heating. Due to the uncertainties in the impacts of stratospheric heating, the study of materials with optical properties that negate stratospheric heating is especially important. Materials such as calcium carbonate ($CaCO_3$), alumina (Al_2O_3), diamond (carbon), and several others, have been proposed as a way to minimize the inherent risks from SAI (Keith et al., 2016; Dykema et al., 2016; Weisenstein et al., 2015; Ferraro et al., 2015; Ferraro et al., 2011; Crutzen, 2006). Although model results of these aerosol species suggest that some of them possess optical properties that make them well suited to be used in a SRM scenario ($CaCO_3$, Al_2O_3, and diamond) (Dykema et al., 2016; Ferraro et al., 2011), the stratospheric aerosol microphysics of these compounds (especially coagulation) is poorly understood. As with AM–H_2SO_4 injections, there is a profound lack of in situ data to assess the ability to model the microphysics of alternative aerosols and the stratospheric chemistry of these materials. This is especially pertinent with respect to changes in ozone, and is exacerbated by the fact that these aerosols have no naturally existing analog in the stratosphere that could be studied. Because early studies suggest that these aerosols show much promise with respect to deploying SAI while mitigating the inherent risks of the deployment, it is imperative to design and execute in situ experiments in order to test our current understanding of the aerosol microphysics and observe the effects of alternative aerosol on the chemical composition and dynamics of the stratosphere.

2.3. Limitations in Existing Analogues

In this section we will review previous in situ studies of stratospheric plume processes, show how those datasets have contributed to our current understanding, and demonstrate the need for experiments such as SCoPEx to inform small-scale models of aerosol microphysics (nucleation and coagulation), plume transport and physical morphology, and chemical properties of new aerosol species that have thus far not been observed in the stratosphere. Because the nature of the injection scenarios (AM–H_2SO_4 or solid aerosols) are so complex compared to natural analogs, new experiments must be designed and implemented to provide observational constraints on our current nearfield modeling framework. Experimental data from carefully targeted small-scale studies would contribute to the development of nearfield-scale models that represent currently uncertain processes in detail.

We note that sub-grid scale processes do not represent the only unknowns in GCMs that are relevant to high-fidelity simulations of SRM scenarios, and that there are many large scale model phenomena which should be further assessed with observational evidence. However, here we focus on the need for in situ data to constrain sub-grid scale processes that can be addressed by SCoPEx and highlight the need for reducing the uncertainty in transport and aerosol dynamics and chemistry at this scale.

2.3.1. Limitations of Solid Rocket Motor Plume Observations

From 1996 to 2000 a number of rocket plumes were observed by high-altitude research aircraft. Generally, these missions involved a research team coordinating stratospheric sampling flights on either the NASA ER-2 or on the NASA WB-57 with coincident rocket launch events from either Cape Canaveral or Vandenberg Airforce Base. These studies sampled plumes from a host of rocket types including Titan IV, Space Shuttle (STS106, STS83, STS85), Delta II, Athena II, and Atlas IIAS.

Plumes were intercepted by the sampling aircraft between 5 and 125 minutes after emission from the rocket motor at stratospheric altitudes ranging from 11 to 19.8km (Voigt et al., 2013). The main science objective of these missions was to assess the stratospheric ozone depletion potential of space exploration by understanding the halogen chemistry occurring as a result of the high-altitude rocket burn. However, in studying the effects on the ozone layer, this era of stratospheric sampling provided a unique set of plume measurements to study nearfield processes of chemical injections into the stratosphere.

While measuring the plumes from the Titan IV rocket (as a part of the United States Airforce Rocket Impacts on Stratospheric Ozone (RISO) Campaign) and attempting to develop a plume chemistry model to solve for the Cl_2 concentration in a rocket plume as it evolves shortly after its emission, Ross et al. (1997) noted the many assumptions that had to be made about the plume morphology in order to simulate the mixing and diffusion that the rocket plume had with the surrounding stratosphere. Their model solved for the Cl_2 concentration of a circular nighttime plume as it expanded in diameter along an isentropic surface. Subsequent aircraft measurements showed that plumes contained more than twice the predicted concentration of Cl_2 despite the plume being intercepted during the day time (when the Cl_2 reservoir should be somewhat depleted by the photolysis reaction $Cl_2 + h\nu \rightarrow 2Cl$), suggesting that there may be an error in the assumption of a circular plume morphology on the short transport time scales observed in this study ($\sim 28 min$).

Ross went on to publish a second study as a part of the RISO project in 1999, this time looking to quantify the size distribution of alumina aerosols emitted from the rocket engines which contained particulate alumina (Al_2O_3) (Ross et al.,1999). They compared measured aerosol size distributions from the WB-57F plume interceptions to results from an aerosol coagulation model and highlighted a massive discrepancy. The model predicted a much smaller aerosol size distribution with 1-10% of the aerosol mass being in the smallest (0.005μm) mode and the aircraft observed only fractions (<0.05%) of the model estimate in that same small mode. At the same time, over 99% of the aerosol mass sampled by the aircraft was found in the coarsest mode (2 μm), which the model was unable to predict. It is most likely that the model used in Ross et al. (1999) did not well account for the effects of ion mediated nucleation as described by Yu & Turco (1997). However, the data from Ross et al. (1999) was some of the first in situ data to highlight the uncertainty in stratospheric aerosol coagulation models. Alumina aerosol, as well as other solid aerosols, in contrast to liquid sulfate aerosol, have since been investigated as a candidate for use in SAI (Weisenstein et al., 2015). Therefore, it is imperative that we understand the chemical, coagulation, and accumulation properties of these and other solid aerosols in a stratospheric environment.

2.3.2. Limitations of Previous Stratospheric Aircraft Wake Crossing Observations

We can look to the few times high-altitude aircraft wake plumes have been sampled in situ for another example of stratospheric plume measurements. In the early 1990s the popularity and capability of the Concorde spurred discussions of a large fleet of High Speed Civil Transport (HSCT) aircraft that would operate in the lower stratosphere between 16 and 23 km. Scientists became concerned with the effects of high-altitude aircraft and high-altitude supersonic aircraft on stratospheric ozone destruction via the creation of a large NOx source in the lower stratosphere. NASA then launched several field campaigns using the ER-2 to study the exhaust profiles of high-altitude aircraft. In 1992 NASA commissioned the Stratospheric Photochemistry Aerosols and Dynamics Expedition (SPADE) to look at the effects of HSCTs. As a part of SPADE the ER2 sampled its own plume on several occasions by making a hairpin turn and heading into its original path, therefore measuring its own wake

(Figure 2). SPADE resulted in at least 11 published studies and some of these can inform us about the mixing and aerosol dynamics that may be relevant to an SAI scenario (Stolarski & Wesoky, 1993).

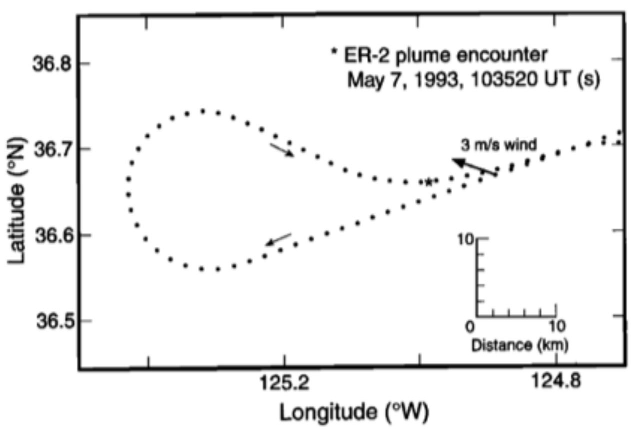


Figure 2: Shows the ER-2 flight track on a typical wake crossing trajectory (adapted from Fahey et al. 1995).

Fahey et al. (1995a) described measurements made of condensation nuclei (CN) present in the ER-2's exhaust plume from the emission of aerosol carbon and of sulfur compounds during one of its SPADE wake crossing events. Because the main focus of this study was to quantify the emission indices (EIs) of various compounds measured by the ER-2 that may have ozone depletion implications, they focused mainly on gas phase compounds. However, for the three wake crossings that the study focused on, they observed large variability in their EI measurements for CN. They noted that this is likely due to differences in mixing history of the encountered air parcels and noted that a full explanation of CN coagulation required more in-depth study and further measurements (Fahey et al, 1995b).

In another study published by Fahey et al. (1995b), they used a similar wake crossing technique to measure the exhaust of the Concorde aircraft and developed an aerosol coagulation model to predict particle formation and size as a function of the time since emission from the aircraft. The coagulation model was initialized at the observed conditions from the one-hour old Concord transect. The results from this model estimated that from 0 to 10 hr since emission from the engine, the mean particle diameter remained fairly constant at 0.06 µm before growing exponentially to a factor of 3 times its initial value over the next 1,000hr. The model predicted exponential mean particle diameter growth continuing right until the of the simulation at 1,000 hr (Fahey et al., 1995a).

Yu & Turco (1997) attempted to model the observed aerosol plume during the Concorde wake crossings with the goal of determining the driving factor for the large aerosol size distributions observed by the ER-2 in the exhaust which had not yet been explained by models. Yu proposed that aerosol formation was being aided by ion-mediated nucleation (IMN), that is, charged particles formed by chemi-ionization processes within the aircraft engines provide charged centers (H_2SO_4 [S(VI)]) around which molecular clusters rapidly coalesce. "The resulting charged micro-particles exhibit enhanced growth due to condensation and coagulation aided by electrostatic effects" (Yu & Turco, 1997). It is likely that IMN is the reason previous particle coagulation modeling of solid rocket motor plumes had overestimated the amount of aerosol in the small size ranges when compared to the in situ data, though this has not since been tested. Because of these effects, and the fact that specific size distributions of aerosol are desired to obtain the optimal radiative forcing effects for SAI (nominally smaller than observed in rocket or aircraft plumes), we must understand the aerosol nucleation and coagulation dynamics in an unperturbed stratosphere.

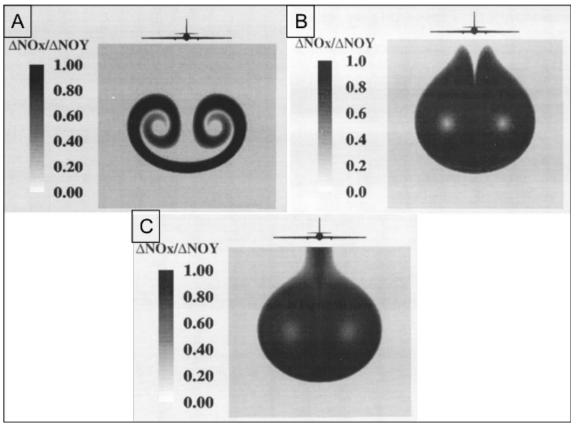


Figure 3: Shows the chemical and morphological evolution of an ER-2 plume during SPADE at 1.7 km (A), 4.8 km (B), and 7.9 km (C). (adapted from Anderson et al. (1996))

As a part of the SPADE project, Anderson et al. (1996) computed the flow field and chemical kinetics of the ER-2 aircraft exhaust using the Aerodyne Research Inc. UNIWAKE model. Their calculations address the effects of complex plume morphology on in-plume chemistry as a function of dilution time since emission from the aircraft engine. They showed that the plume morphology is highly variable out to about 5 km post emission Figure 3 and estimated that the stability of the wing vortex pair begins to break up at roughly 20 km post emission. Although this study was completed in the mid 1990s, it is still one of the only studies that attempts to compute nearfield chemistry within a dynamic stratospheric plume. However, particles were not considered as part of this study.

2.3.3. Limitations of Stratospheric Wake Crossings

Previous stratospheric plume studies of solid rocket motors and aircraft wake crossings have laid the foundation for our understanding of stratospheric plume chemical, aerosol, and mixing dynamics on transport scales of 0→100 km. These studies highlight the types of processes we must be aware of when considering the logistics of SAI. However, the violent initial conditions of engine exhaust plumes (such as temperatures of 700K, IMN) make it difficult to relate these observations to other systems. Because the engines drive the mixing and transport in the nearfield, and the ionic injection conditions of the plume create electrostatic forces that introduce complex nucleation affinities (IMN), understanding individual parameters can become analogous to finding a needle in a haystack. Moreover, because the radiative properties of any stratospheric aerosol that may be used for SRM depend on the diameter of the particle, we must understand the coagulation of that aerosol in the nearfield after the injection, which means that we must also understand the plume morphology that dictates the concentrations of that aerosol. Currently there have been no in situ data gathered that help us understand nearfield aerosol nucleation and plume dynamics in the absence of a very disruptive source. These conditions are necessary to understand as SAI may require that we mitigate the effect of IMN in order to obtain an aerosol size distribution that is small enough to provide the desired radiative properties.

2.3.4. Limitations of Naturally Occurring Analogs

Another source of useful in situ data on plume dynamics in the stratosphere can be found in literature addressing the fate and transport of convective overshooting events that often occur at the top of a Mesoscale Convective Complex (MCC) or via pyrocumulonimbus (pyro-cb) events. These events drive brief airmass exchange with the troposphere and often end up resulting in a plume-like parcel of tropospheric air being injected into the stratosphere.

Measurements of convective systems and upper troposphere-lower stratosphere exchange, as a means to interrogate stratospheric plume transport, have provided valuable in situ datasets that help us understand mid-field (10 to >1000 km) plume dynamics in the lower stratosphere. Similar to convective overshooting events, large pyro-cb events, such as the 2019-2020 Southeast Australian wildfires provide a wealth of useful information, with studies highlighting the resulting radiative forcing, ozone destruction, and persistent stratospheric warming (Heinold et al. 2021; Solomon et al. 2022; Yu et al. 2021). In addition, volcanic eruptions have provided an immense amount of in situ data that has informed us about regional and even global transport of stratospheric injections (Robock, 2000). Although these data are applicable in some sense to the transport of an SAI plume after its initial injection, the turbulent nature of a convective storm or pyro-cb event makes it difficult to measure these events at points near their injection source. Additionally, the storm/pyro-cb conditions themselves dramatically complicate the system in the lower stratosphere such that is difficult to see through the effects of the induced turbulence in the nearfield. Indeed, an important limitation of all these type of natural analogs is the spatial extent of their perturbation, which does not allow for near-field observations analogous to that of a point source. This also arises from the violent nature of these events which does not allow airborne platforms, such as the ER-2, to sample the initial conditions of the injection. We also note that volcanic eruptions are limited in their utility to evaluate dynamic response to stratospheric heating from sulfate aerosol, as they represent a perturbative pulse rather than the long-term heating one would expect from SAI.

In addition, these natural analogues provide extremely limited ability to study alternate materials, although organic and mineral dust aerosol injections into the lowermost stratosphere have been documented from convective overshoots. However, the complexity of the massive perturbations of both gas- and particle-phase preclude a study focusing on the impact on stratospheric composition and aerosol evolution that would result from SAI of a single material.

3. SCoPEx Short Overview

This section provides a brief overview of the engineering and operational aspects of SCoPEx. We first describe the platform, the instruments, and the concept of operations before describing the rationale for the overall SCoPEx design choices.

3.1. SCoPEx Platform

The SCoPEx gondola (Figure 4) is a balloon-born new research platform being developed at Harvard by the engineering and science staff within the Anderson/Keith/Keutsch laboratory group. The development builds on four decades of stratospheric research on aircraft, balloon, and rocket platforms that has focused on understanding the environmental chemistry of the ozone layer, stratospheric transport and dynamics continuing to date with the NASA EVS3 Dynamics and Chemistry of the Summer Stratosphere (DCOTSS) field campaign for which Keutsch is co-PI. The SCoPEx experiment was first described by Dykema et al. (2014). While many details of the design have changed, that paper still succinctly describes the advantages of choosing a balloon born platform over an aircraft, particularly for studying perturbations like solar geoengineering, and several of the limits of laboratory experiments that that could be addressed in a perturbative experiment like SCoPEx.

The gondola has three primary features: the frame, the ascender, and the propellers. The aluminum and carbon fiber frame contains two decks and a ballast hopper for coarse altitude control. One deck is primarily dedicated to platform support (power and flight control) and one deck is primarily dedicated to instruments. At the top of the gondola is an ascender and rope which allows the distance between the bottom of the balloon train and the gondola to vary from 0 to 150 m, which provides fine altitude control of the gondola. The ascender has been developed and tested by Atlas (Chelmsford, MA) building on their previous hardware in collaboration with the Harvard engineering team. The propellers serve two purposes: to create a well-mixed volume of air where observations of the aerosols and perturbed gas-phase can be made, and to reposition the gondola within the evolving aerosol plume. While the trajectory of the balloon and gondola system will be dictated by the balloon, the propellers allow for repositioning relative to the prevailing winds.

The ascender makes it impossible to have cables and other physical connections between the flight operations equipment and the gondola. Thus, the platform will handle its own communications and power. The SCoPEx platform will be powered using 28 V and 100 V DC power supplies which will power all operations on the platform including the propellers, ascender, and instruments. Elements of the flight platform are listed in Table 1. The gondola flight, flight safety, recovery parachute, and recovery operations will be managed by the balloon operator (in contrast to the SCoPEx team itself). Because the absolute velocity and distance capability of the gondola are so small compared to balloon drift, the trajectory will be determined by the balloon operator as if it was a passive nonpowered payload. During operations, the detailed float altitude will be jointly managed by the balloon operator via control of the balloon vents and the Harvard team via control of the ballast and ascender.

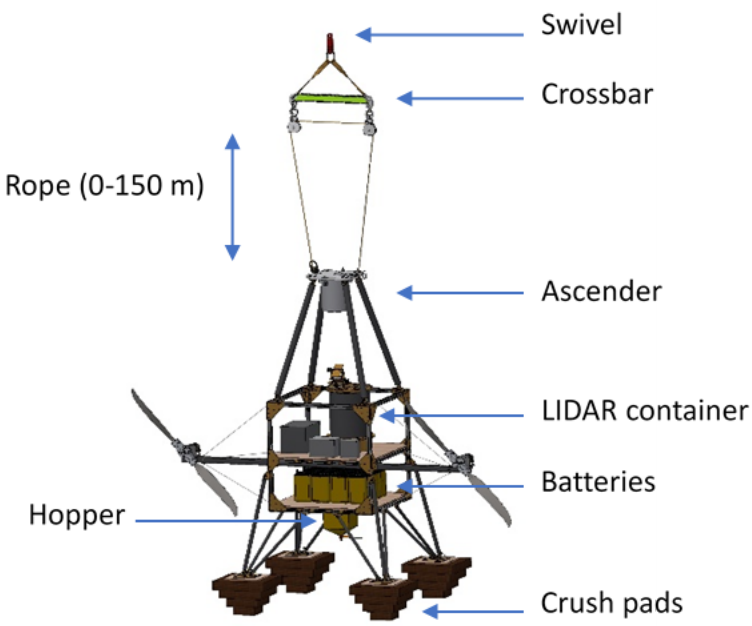


Figure 4: A representation of the SCoPEx flight platform. The final configuration may have subsystems packaged differently.

Parameter	**Description**
Total mass (Frame, all subsystems, hopper with ballast)	600 kg
Interface to balloon	Crosby 5-S-2 jaw & jaw swivel
Ascender	13 mm diameter rope Range of motion: 0-150 m Max speed: 10 m/min
Gondola propulsion	Twin propellers, 1.88 m diameter 32 N thrust each Max airspeed: 3 m/s
Power	28 V and 100 V DC power supplies with 24 MJ and 10 MJ total energy when fully charged
Communications	Satellite phone for communication between ground equipment and payload
Maximum termination shock	10 g

Table 1: Elements of the SCoPEx flight platform.

3.2. Instruments for First Science Flights (Science Goals 1 and 2)

The proposed instruments for the first science flight, addressing science Goals 1 and 2, are listed in Table 2. The corresponding science goals that motivate their inclusion are detailed in Section 4.

Measurement	Instruments	Rationale	Corresponding Science Goal
Wind speed measurement	Wind pendulum	Gondola and plume movement relative to balloon	Platform operation
Meteorology	Commercial off-the-shelf instrument	Temperature and pressure measurement throughout the flight	1, 2, 3
Wind turbulence	Constant temperature anemometer	Stratospheric mixing and modeling evolution of aerosol size distribution	1, 2
Particle dispersal	Solid Aerosolizer	Injects monodispersed particles for measurement and study	2, 3
Plume tracking	LIDAR	Tracking plume and navigation back into plume	2, 3
Particle sizer	POPS	Aerosol size distribution measurement for comparison with microphysics models of near-field evolution	2, 3
Light Scattering	Radiometer	Comparison of aerosol scattering with model prediction	2

Table 2: Instruments for first SCoPEx science flight.

Wind Pendulum: Understanding differential wind speed measurements between the balloon and payload will be important for plume evolution relative to the balloon trajectory and navigating the payload back into the plume. Commercial equipment to measure wind speed is typically not designed for the low densities found in the stratosphere. SCoPEx will therefore use a pendulum-based instrument and model to extract wind speed measurements. A camera will track a pendulum bob with high surface area and low mass, light enough to be perturbed by low winds in the stratosphere. Using the location and tilt data from the payload and a 3-dimentail kinetic model, the wind speed will be extracted from photos of the pendulum bob.

Commercial Meteorology Instrument: Commercial off-the-shelf instruments will be used for meteorological measurements on SCoPEx. They will record pressures and temperatures of the ambient stratosphere.

Constant Temperature Anemometer: A constant temperature anemometer (CTA) uses convective cooling caused by air flowing across a heated thin wire to measure flow velocity. LITOS (Leibniz-Institute Turbulence Observations in the Stratosphere) (Gerding et al., 2009; Theuerkauf et al., 2010) used such a measurement to study stratospheric turbulence up to 29 km. LITOS consisted of a 5 μm diameter and 1.25 mm long tungsten wire CTA and a 16 bit ADC with 2000 samples per second to collect measurements with a vertical resolution of 2.5 mm at 5 m/s ascent speed. The anemometer data was analyzed by performing a spectral analysis on the voltage signal to retrieve the spectral slope of the observed variation. A similar instrument will be used on SCoPEx to measure stratospheric turbulence. Air flow around the device will be simulated using CFD tools. The CFD runs will provide a means to identify key flow characteristics that drive sensor performance (sensitivity and accuracy), and to drive detailed sensor design.

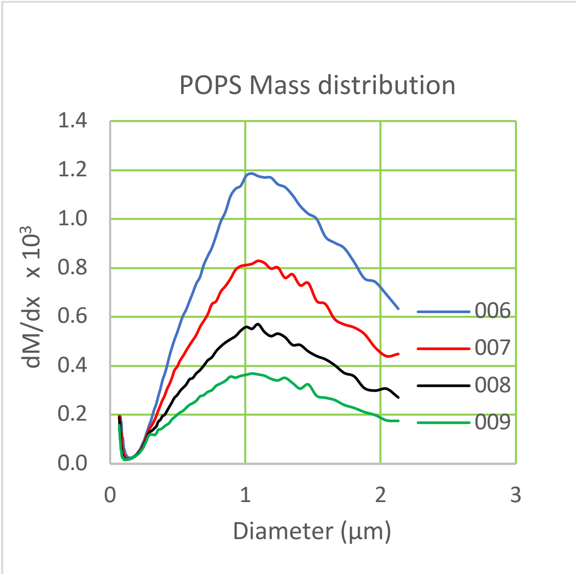


Figure 5: Successive measurements of sprayed $CaCO_3$ using an optical particle spectrometer. 006-009 indicate numbered time intervals spaced 4 minutes apart with 006 being the earliest measurement. $CaCO_3$ was sprayed using a 200 μm nozzle. In this laboratory experiment there was no significant variation in the shape of the distribution over time. (personal communication A Neukermans and team)

Solid Aerosolizer: The solid particle aerosolizer has been developed by a team lead by Armand Neukermans. For SCoPEx, the goal is to spray roughly monodisperse ~0.5 μm diameter precipitated calcium carbonate powder, the first candidate for solid SAI, through a 1-2 mm nozzle using the expansion of powder suspended in high pressure liquid CO_2. The aerosolizer would use a 1:4 weight ratio of $CaCO_3$ to CO_2. For 1 kg of $CaCO_3$ this would require a 5-7 L pressurized container. This concept has already been demonstrated in the lab. Figure 5 shows successive measurements of sprayed $CaCO_3$ with a size distribution centered at 1 μm diameter. Measurements were taken every 4 minutes using POPS (see below). In this case, total particle count decreased over time but there was no significant variation in the shape of the size distribution.

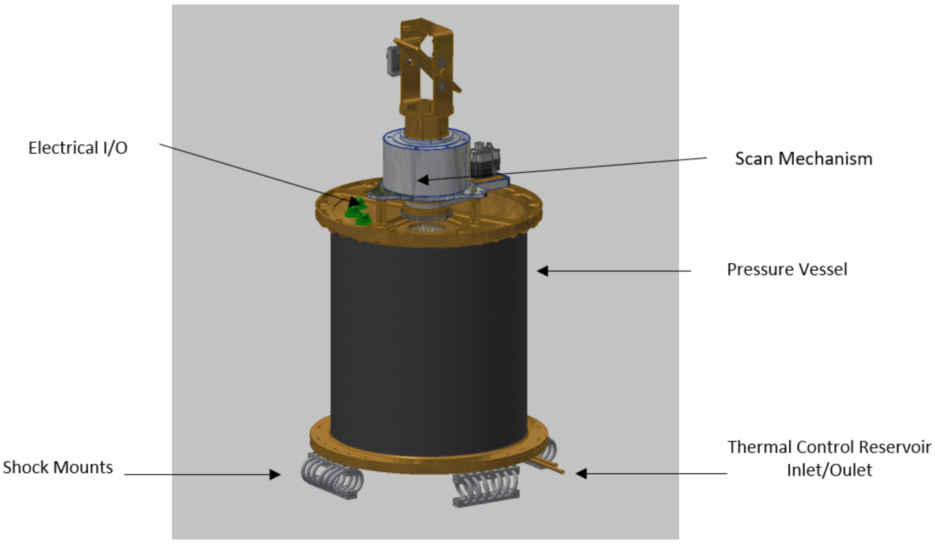


Figure 6: LIDAR pressure vessel provides safe storage and operating environment and support equipment.

LIDAR: The LIDAR is used to track the plume and allow navigation back into it. The core of the LIDAR system is an off-the-shelf eye-safe visible LIDAR, purchased from Sigma Space (now owned and operated by Droplet Measurement Technologies). This LIDAR produces 4 µJ pulses of 532 nm light at a repetition rate of 532 nm. The light that is backscattered by molecules and aerosols is collected by an 80 mm telescope and detected with a high-speed, high-sensitivity photodiode.

We have integrated this LIDAR in a pressure vessel (Figure 6) to provide a near-1 atm pressure environment with adequate temperature stability to ensure safe operation of the LIDAR at float altitude and safe storage on launch, ascent, descent, and recovery. This pressure vessel includes equipment for electrical and mechanical support, including command, data handling, and shock mounting. The LIDAR requires a scan capability to search the nearby atmosphere for the extent and geometry of the plume. The tilt and pan functions of the scan capability allows the LIDAR to be scanned over a set of angles that define the plausible location of the plume.

Portable Optical Particle Spectrometer (POPS): The POPS instrument will provide the aerosol size distribution measurements for studying aerosol formation and agglomeration. POPS is a light-weight instrument that directly samples the aerosol. It was built by and provided to SCoPEx through a collaboration with NOAA. The particles are illuminated with a 405 nm diode laser and the scattered light is collected onto a photomultiplier tube. The particle size is determined by the intensity of the scattered light. It has both the detection limit and size range (0.13 – 3 µm) to measure background stratospheric aerosol, which is more than sufficient for SCoPEx needs (Gao et al., 2016).

The Keutsch Group has already developed and extensively characterized a POPS instrument in preparation for the NASA-EVS3 Dynamics and Chemistry of the Summer Stratosphere field campaign on board the NASA-ER2, for which Keutsch is the deputy-PI. The POPS instrument tests include extensive thermal vacuum chamber characterizations to ensure operation under harsh stratospheric conditions. Compared to the ER-2, operation for SCoPEx will be simpler due to the insignificant air speed of the balloon and a much simpler operational pressure regime (on the ER-2 there is a large range of external pressures for both sampling and exhaust).

Radiometer: The aerosol plume can also be detected using a narrowband, narrow field of view radiometer with azimuthal/zenith pointing capability. The relationship between measurements of scattered solar radiation and the physical characteristics of atmospheric aerosols has been studied for more than two decades. Sky scanning measurements at multiple wavelengths between 300 nm and 1200 nm have been obtained using robotically pointed ground-based spectral radiometers deployed worldwide (Holben et al., 1998). The theory of these measurements has been refined and validated as a function of viewing geometry to provide a strong basis for inferring aerosol microphysics from radiometer data (Torres et al., 2014). The success of these approaches has motivated the development of compact sky scanning radiometers suitable for deployment on unsteady platforms like unmanned aerial vehicles (UAVs) and SCoPEx. One such design, reported by NOAA (Murphy et al., 2016), measures at 4 wavelengths (460 nm, 550 nm , 670 nm, and 860 nm) with a field of view of 0.006 sr (equivalent to 2.5° half-angle) and a circular limiting aperture of 1.1 mm diameter. A radiometer like this one deployed on SCoPEx would be capable of observing a SCoPEx plume, based on Golja et al. (2020), formed by a 0.1 g s^{-1} injection of calcite from a distance of 200 m with an approximate signal-to-noise ratio of 6000 for a 1 ms signal accumulation.

3.3. Instruments for Future Science Flights (Science Goal 3)

The additional instruments listed in Table 3 are candidates for future SCoPEx flights beyond the initial science flight, i.e., addressing science goal 3. They have not yet been adapted to fly on the SCoPEx platform. Instrument choices will be refined based on experiences in the first science flights. The corresponding science goals that motivate their inclusion are detailed in Section 4.

Measurement	Candidate Instrument	Rationale	Corresponding science goal
Aerosol composition	Drum Sampler	Collecting aerosols for offline analysis	3
Water Vapor	IR Absorption or Frost Point	H_2O outgassing of platform, Influence on coagulation and heterogenous chemistry	2, 3
Atmospheric trace gas concentrations (ex: HCl, NOx)	Spectroscopic trace gas instruments	For measuring concentrations of various atmospheric trace gases before and after addition of solid ASI material	3

Table 3: Potential instrument for future SCoPEx science flights.

Aerosol Composition: Aerosol composition can be analyzed via the collection of aerosol with a drum sampler followed by offline analysis in the laboratory using standard offline methods. Aerosol sampling has been done numerous times aboard stratospheric platforms.

Water Vapor: Gas-phase water vapor measurements are important as relative humidity likely has a large impact on the heterogeneous reactivity of solid SAI material. The balloon and gondola can outgas significant amounts of water and thus an initial experiment will characterize how long, if at all, this outgassing perturbs the SCoPEx plume. As mentioned previously, the goal of SCoPEx is to ideally minimize the perturbation to only the introduction of calcium carbonate. Water vapor measurements are common on many stratospheric platforms.

Hydrogen Chloride: HCl can be measured via infrared absorption spectroscopy. The Anderson group at Harvard, which shares a laboratory with the Keutsch group, has developed a stratospheric HCl instrument and thus has extensive experience with the design of stratospheric HCl instrumentation. In addition, the Keutsch group has designed multiple spectroscopic trace gas measurements. The much lower air speeds of the balloon compared to aircraft favor the design of an open path system, which eliminates the notorious wall effects that can make HCl measurements challenging.

NOx: For NOx there exist a number of good instrumentation options. Recently, a compact NO-LIF instrument has been designed that has spectacular detection limits in the low ppt range, more than sufficient for the needs of SCoPEx. The instrument is a close analogue of the fiber-laser based formaldehyde LIF instrument that the Keutsch Group developed, so there is a high degree of expertise available for such an instrument. There are also sensitive cavity enhanced techniques available usually in the visible range of the spectrum.

3.4. SCoPEx Concept of Operations

Flights will proceed in the following manner. The payload would be launched with the ascender retracted such that there is minimal distance between the crossbar and platform. Once the balloon reaches the float altitude, the rope will be let out through the ascender such that there is 100 m between the crossbar and platform. The platform will then be ready to perform experiments and execute maneuvers. Figure 7 illustrates a proposed flight maneuver. The platform will initially travel in a straight line laying out a plume, after which it will maneuver back through the plume to make measurements. During these maneuvers the ascender can be used to fine tune the altitude of the platform and instruments. Several series of such maneuvers can be performed within each flight. At the conclusion of the experiments the ascender retracts the rope before the descent.

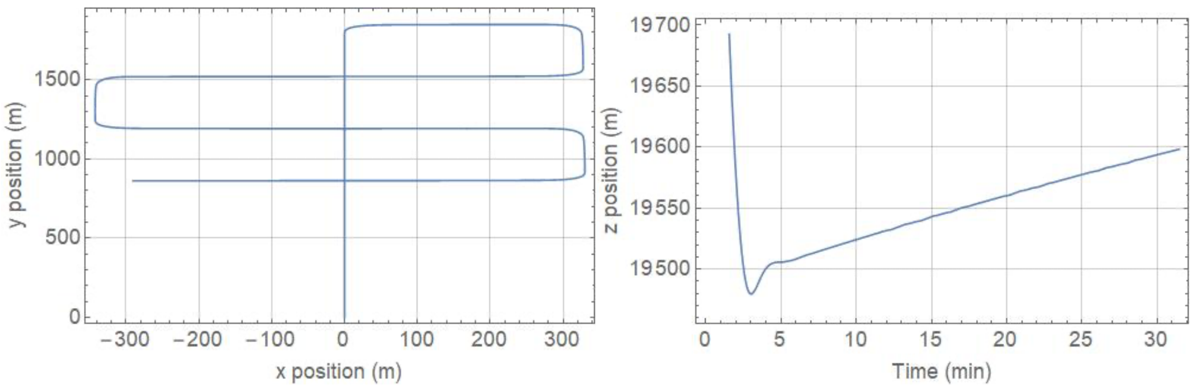


Figure 7: (left) A top down view of the proposed flight maneuvers over a 35-minute window. x and y are in the horizontal plane. The platform begins at (0,0). (right) The vertical position expected without any ascender or hopper vertical trimming over the same 35-minute platform maneuver.

4. SCoPEx Goals

In this section we describe the three long-term SCoPEx science goals. For each goal we describe the scientific problem, the need for SCoPEx, and the measurements required. The first phase of science flights targets the first two science goals. The design of the flights for the third goal will be informed by an understanding of the evolution of particle size distribution in the plume and the plume size. Thus, if later stage science flights move forward, they will be refined based on the results of the first science flights and the most up-to-date knowledge within the solar geoengineering and stratospheric science research communities.

4.1. Goal 1: Measurements of Turbulence for Small-Scale Mixing

4.1.1. The Importance of Plume-Scale Turbulence

Stratospheric turbulence influences the evolution of aerosol distribution from plume to regional to global scale. The mixing of air masses (of differing composition) in the stratosphere is a combination of two processes (Nakamura, 1996; Schoeberl & Bacmeister, 1993). The first process is strain, the distortion of streamline flow that brings air masses of differing composition adjacent to one another (Prather& Jaffe, 1990). Sometimes this is also referred to as “stirring” (Haynes, 2005). The second process occurs when air masses of differing composition are transported across the streamlines. This second process is the true “mixing” process.

In the stratosphere, mixing ultimately occurs because of molecular diffusion. This happens at the length scale of molecular viscosity. It is accelerated by turbulence, which can dramatically enhance the rate at which differing air masses are deformed to small enough spatial scales for molecular diffusion to mix them efficiently. Stratospheric turbulence is, however, highly intermittent (Vanneste, 2004). Understanding the mechanisms of stratospheric turbulence production is essential to understanding the spatial inhomogeneity and effective rate of mixing on spatial scales of 10-500 m (Schneider et al., 2017).

An understanding of this role of turbulence is of interest to stratospheric science because studies suggest that more accurate representations of mixing influence tracer distributions (Hoppe et al., 2014). Measurements of long-lived tracers are the strongest observational constraint on the stratospheric age of air, a key measure of the stratospheric large-scale circulation. Turbulence also modifies the character of kinetic energy fluxes. The magnitude and variability of these energy fluxes determine the rate of frictional dissipation in the atmosphere. This dissipation is represented in global models by a damping parameter and is the primary determinant of the mesoscale atmospheric kinetic energy spectrum. The uncertainty in kinetic spectrum is important to the understanding of the large-scale circulation of the middle atmosphere (Jablonowski & Williamson, 2011).

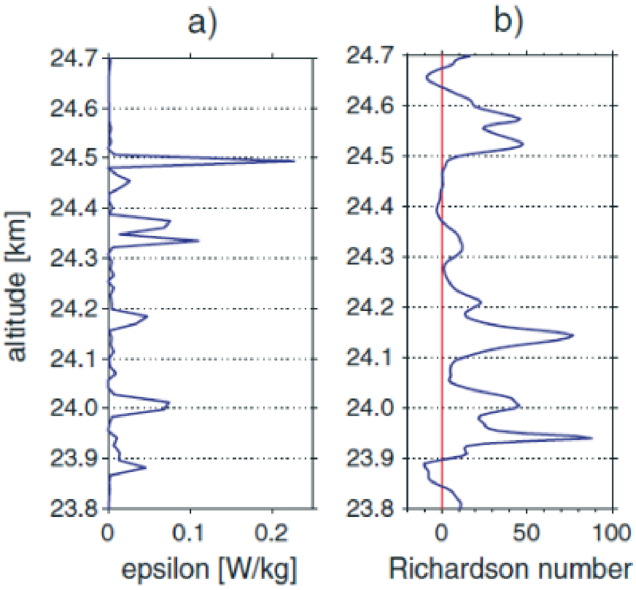


Figure 8: LITOS balloon-borne high-speed anemometer measurements reveal that models of atmospheric turbulence do not explain observed stratospheric turbulence. Physical models predict that a low Richardson number (buoyancy/shear ratio) implies turbulence, but high values of epsilon (turbulent dissipation) should be correlated with low Richardson number, which is not observed. (Haack et al., 2014)

Physical models predict that a low buoyancy/shear ratio (Richardson number) implies turbulence, and that high values of turbulent dissipation should be correlated with low Richardson number (Figure 8). However, recent balloon born measurements during the LITOS campaign did not agree with this, with numerous instances of high values of turbulent dissipation occurring at high Richardson numbers (Haack et al., 2014). As detailed above, both the impact of turbulence on mixing and the associated dissipation of energy are important for general stratospheric science. The point at which viscous fluid forces dominate atmospheric motion is the point where atmospheric motions become purely statistical and is called the dissipation scale. At this scale, models no longer require computationally expensive deterministic modeling. Furthermore, these viscous forces are also responsible for the dissipation of turbulent kinetic energy. Therefore, measurements which resolve the winds at the dissipation scale will allow numerical models to realistically close the atmospheric kinetic energy budget, an important metric of model fidelity.

4.1.2. Importance of Small-Scale Mixing for SAI and SCoPEx

From an SAI and SCoPEx perspective, plume-scale turbulence influences the frequency of collisions of monomer particles within the SCoPEx plume, which determines the rate of formation of fractal, larger aggregates. While Van der Waals forces finally determine whether particles that collide stick together and remain as a fractal aggregate (Sukhodolov et al., 2018), the collision rate is a critical quantity in determining total coagulation rate. Therefore, it is essential to know the frequency of collisions. This frequency is controlled by the wind variability at small spatial scales, i.e., the power spectrum. Intuitively, inertial forcing of particles by wind is much stronger than thermal forcing (e.g. Boltzmann distribution of velocity for ~1 µm particles at ~220 K). Fractal aggregates have a shorter lifetime in the stratosphere and are less effective at scattering light on a per mass basis (Weisenstein et al., 2015), so being able to model the formation rate of fractal aggregates is an important aspect of SAI, especially with alternate SAI materials.

Improved knowledge of collision rates from wind measurements will allow for the selection of the appropriate mathematical representation of particle coagulation, the coagulation kernel. An accurate kernel is essential for numerical models to correctly simulate aerosol microphysical processes that determine the size distribution and residence time of solid aerosol particles. Adding wind and turbulence measurements to the SCoPEx payload will therefore address the major sources of uncertainty in aerosol microphysics under real atmospheric conditions, which include small-scale fluid flow, particle composition, and humidity.

4.1.3. Experimental Methods to Measure Turbulence in the Stratosphere

Multiple technologies are possible to achieve wind measurements with the necessary spatial resolution under stratospheric conditions. Current state of the art options include pitot tubes (with high sensitivity micro-pirani pressure sensors), hot wire anemometers, and acoustic anemometers. An existing stratospheric program has utilized hot wire anemometers to make measurements that are a close analog to what is necessary for SCoPEx. The program developed LITOS (Leibniz-Institute Turbulence Observations in the Stratosphere), an instrument which made measurements of stratospheric turbulence up to 29 km (Gerding et al., 2009; Theuerkauf et al., 2011). The LITOS instrument has undergone significant calibration and has been compared against radiosondes (Schneider et al., 2015). One drawback of its deployment on a balloon has been the contamination of its wind measurements due to the influence of the balloon's wake. In contrast, SCoPEx is engineered so that the wind environment of the instrument payload is well separated from the balloon wake when SCoPEx is traveling horizontally. For this reason, SCoPEx could provide significantly more data per flight at a chosen float altitude. In this way, SCoPEx and LITOS would be very complementary. The horizontal flight path of SCoPEx, combined with measurements of the wind power spectrum, would provide an excellent complement to the LITOS observations, which are only obtained along a vertical profile. These power spectra obtained by SCoPEx would contribute to improved micrometeorology understanding relevant both to stratospheric aerosol injection and to fundamental atmospheric science.

Additionally, air flow through the turbulence instrument will be simulated using CFD tools. The CFD runs will provide a means to identify key flow characteristics that drive sensor performance (sensitivity and accuracy) and detailed sensor design. This application of the SCoPEx platform would therefore constitute a nonperturbative means to obtain necessary turbulence measurements that have, to date, eluded the scientific community. This information is important for understanding stratospheric dynamics, including the response to climate change or stratospheric heating from SAI. As no injection of particles is needed, these could be among the first scientific measurements to be conducted.

4.2. Goal 2: Evaluation of Aerosol Microphysics of AM-Sulfate and Alternative SAI Materials

One of the goals for which there are insufficient observational analogues is the near-field evolution of particles injected from a point source in the stratosphere. Specifically, observations of the temporal and spatial evolution of the aerosol size distribution (number and volume) of solid, alternate SAI materials or AM-H_2SO_4 injected form a point source can only be compared with plume model predictions via a perturbative experiment such as SCoPEx. In the following we describe a plume model by Golja et al. (2020) specifically designed for SCoPEx. We also explain the results from the model and the SCoPEx experimental approach for comparing observations with model results.

4.2.1. Plume Model

Golja et al. (2020) incorporated the SCoPEx design features in their model to study the injection of a solid aerosol and vapor-phase sulfuric acid from a balloon payload. To provide observations relevant to SAI, SCoPEx needs to produce downstream aerosols with radii within the range of roughly 0.2 to 1.0 µm. For calcium carbonate, the objective is to maintain a high fraction of the aerosol in monomer form, while for sulfate an ideal distribution would have a peak diameter of 0.6 µm (Dykema et al., 2016). The generation of largely smaller than ideal particles, while imperfect for assessing radiative efficiency relevant to SAI, does not serve to increase particle sedimentation rates within the plume. Such smaller sizes may, however, result in a larger surface to volume ratio, which can strongly influence stratospheric composition as heterogeneous chemistry is directly related to surface area. Distributions centered on small particle sizes in the near field may, however, continue to evolve beyond the domain of the study.

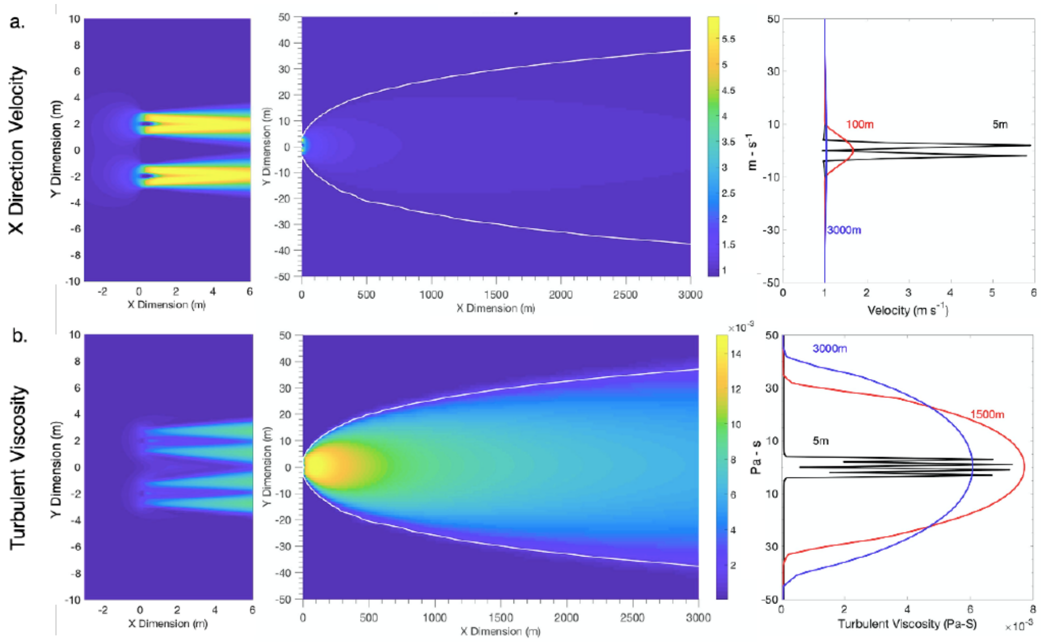


Figure 9 : ANSYS Fluent Velocity and Turbulence Fields. Shown above are the steady state x-direction velocity, u, and turbulent viscosity fields generated by ANSYS Fluent. Left panels show the genesis of disruptions to background X direction flow of 1 ms^{-1}, where propeller features are imposed at locations of 0,2) and (0,-2) meters. The center panel shows the entire domain, from 0 to 3 km, where the imposed red line contours 1 ms^{-1} in plot A, and contours 10% of the absolute maximum turbulent viscosity in plot B. Note Y direction scaling differs between the center and left panels. The right panel shows cross sections of velocity (A) and turbulent viscosity (B) through the Y plane at varying X locations. (Golja et al. 2020)

The velocity and turbulent viscosity fields from Fluent are shown in Figure 9. These fields form the basis of the simulation environment and are instructive in achieving an understanding of SCoPEx and the perturbation it achieves. Peaks in the x-direction velocity, u, are found directly downstream from the modeled propeller centers with an absolute maximum value of 6.3 ms^{-1}. By 1500 m downstream from the inlet locations, the velocity is reduced to the imposed background flow of 1 ms^{-1}. Turbulent viscosity, used as a measure of particle mixing with background air, exhibits a narrow distribution of peak values ~10 m downstream from simulated propellers. With increasing distance downstream, the turbulent velocity spatial distribution widens, attaining a full width half maximum (FWHM) of 60 m by 1500 m downstream. The wake of the balloon itself is not visible, as it is sufficiently far from the payload to avoid wake crossing/interaction. Additionally, this simulation assumes a laminar stratospheric background flow, neglecting the potential impacts of breaking gravity waves.

For SCoPEx, precipitated calcium carbonate powder with roughly monodisperse size distribution centered at ~0.5 mm diameter will be aerosolized using the expansion of powder suspended in high pressure CO_2 through a 1-2 mm nozzle (see description in Section 3). The model injects aerosol as a 3D gaussian distribution of mass flux into the model grid, where the size of that distribution represents the scale of which the high velocity jet from the nozzle mixes with ambient air. The model considered two injection scenarios: scenario 1 (S1), a single point injection between the propellers; and scenario 2 (S2), injection from the center of each propeller. The model plume diameter at 3 km is, however, insensitive to the injection scenario for injection of both AM-H_2SO_4 and calcium carbonate. This suggests that injection at or between the propellers does not significantly alter the characteristics of the particles' experienced velocity field, and scenario S1 is the one selected for testing the model of plume evolution on SCoPEx. This is also important for the SCoPEx experiment as it necessitates only one sprayer that can be more easily placed in the equipment gondola.

4.2.2. Modelled Mass Injection Rate Dependence of Aerosol Size Distribution

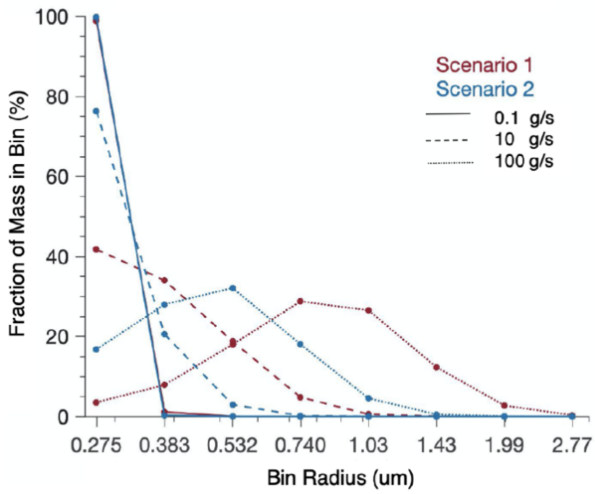


Figure 10: Calcium carbonate aerosol size distributions. Fraction of total mass in each sectional bin where the x-axis markers represent the central radius of each sectional size bin. These distributions represent the percent of total aerosol mass in the final 100 m of the plume across the full domain. Results are shown for three injection rates, 0.1 g s^{-1}, 10 gs^{-1}, and 100 gs^{-1}, for injection scenario 1 (red) and 2 (blue). (Golja et al. 2020)

Mass injection rates of 0.1, 10, and 100 g s^{-1} (0.36, 36, and 360 kg hr^{-1}) were used to test the influence of initial particle number density on the final plume aerosol size distribution. Although some of these are high, their use in the model is instructive as it can answer how different a short burst of high injection rate (much less than an hour) is from a slower but longer injection for the same total mass. Increasing calcium carbonate injection rates from 0.1 to 100 g s^{-1} reduces the share of monomer particles and increases undesired multi-monomer fractal aggregates. Figure 10 shows calcium carbonate's size distribution in the final 100 m of the modeled plume, i.e., the percent in each bin for the three different injection rates of 0.275 μm radius particles. The low calcium carbonate injection rate of 0.1 g s^{-1} is the most desirable, maintaining 99% of the total mass in the final 100 m of the plume in monomer form. Increasing mass injection rate to 10 g s^{-1} and 100 g s^{-1}, with an S1 injection, shifts peak mass loading to favor particles of radii 0.5 and 0.75 um, respectively, corresponding to fractal "dimers" and "trimers".

Golja et al. (2020) also evaluated whether, in addition to the very sensitive in-situ optical particle counting aerosol size distribution instrument which originally was designed to measure background stratospheric aerosol size distributions (Murphy et al., 2016), the plumes could also be detected optically via scattered light. It should be emphasized that this does not refer to measurements from the ground but rather from close to the plume, e.g., when the equipment gondola is in close vicinity to the plume. Measuring the scattering from one view angle gives the product of the scattering phase at that angle and the scattering efficiency. This is closely related to the radiative forcing, but it does not uniquely determine the radiative forcing. By measuring at multiple angles, we could obtain enough information to quantify the radiative forcing. For example, we could measure from the side and below to obtain the forward scatter fraction, then calculate backscatter by flux conservation.

In the model, the extinction optical depth was calculated using Mie scattering theory and vertically integrating down columns in the y-z plane. Figure 11 shows the relative optical thickness of a sulphate and calcite aerosol plume formed via scenario 1 with an injection rate of 0.1 g s^{-1}. Calcite exhibits greater optical thickness by an order of magnitude at 550 nm, with an average value of 8.6×10^{-4} and maximum of 0.014 across the domain, as compared to sulphate, with an average of 9.4×10^{-5} and maximum 0.001. From these values, Golja et al. calculated that we expect adequate SNR to confidently detect the plume with a fast-scanning radiometer via the solar radiation it scatters. This calculation assumed an altitude of 21 km, solar elevation angle of 60°, an observing instrument situated on the payload gondola, and the gondola 200 m away from the edge of the plume and 1 km downstream of the termination of a scenario 1 type injection of calcite aerosol. Details of this calculation can be found in Golja et al. (2020).

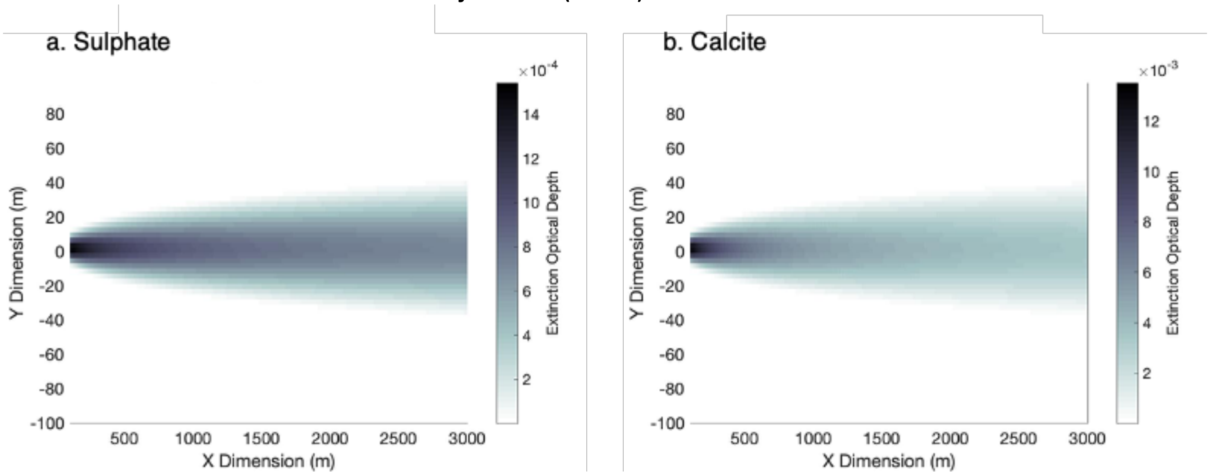


Figure 11: Extinction optical depth integrated vertically through all columns in the plume from 100-3000 m. Plots a and b show results for 0.1 gs^{-1} injections of condensable H_2SO_4 and calcite, respectively. The resulting number density of calcite aerosol is 490 cm^{-3} on the centerline at a downstream distance of 1000 m, predominantly as monomers. Aerosol optical depths were derived from Mie scattering theory at 550 nm, using refractive indices for sulphate and calcite stated in Dykema et al. (2016). (Golja et al. 2020)

4.2.3. SCoPEx Experimental Design and Analysis of Plume Evolution

For this goal, SCoPEx will follow the standard concept of operations, first spraying calcium carbonate at an injection rate suggested by the model analysis. It is desirable to maximize the contrast with the background stratosphere, both with respect to the aerosol concentration and the potential resulting chemical changes, while also maintaining calcium carbonate as monodisperse aerosol. To this end, additional models will be run at injection rates between 0.1 and 10 gs^{-1}. Based on these results, an injection rate will be chosen for the actual SCoPEx experiment. In addition to the basic components of the SCoPEx platform (gondola, ascender, propulsion, power, flight computer, communication, and wind), the calcium carbonate sprayer as well as the LIDAR and POPS instrument are critical for this science goal; without these components, there would not be a way to make and find the plume or measure the aerosol size distribution. While the turbulence measurement from goal 1 is desirable, it is, at least initially, not necessary. Similar studies of AM-H_2SO_4 injection would also be extremely useful. Our current plan is to conduct these after the calcium carbonate injection studies, as initially calcium carbonate is easier to handle than sulfuric acid and its precursors (see next section for motivation of calcium carbonate).

The aerosol size distribution measurements will be compared with the model predictions. In combination with turbulence measurements, discrepancies between the observed and modeled aerosol size distributions can be used to identify issues within the aerosol microphysical scheme or highlight misrepresentations of the velocity and turbulence field of the payload. The results of these studies will provide critical observational constraints on the aerosol microphysics and plume evolution of an injection with solid particles. It will be unique data that is ideal for testing the model of plume evolution as SCoPEx does not have to address problems resulting from the much more violent injection regime associated with injection from airplanes. Clearly, such studies are also needed, but SCoPEx represents a feasible and compelling first step in a sequence of new studies that more comprehensively investigate the aerosol microphysics of point source injections.

4.3. Goal 3: Evaluation of Process Level Chemical Models of Stratospheric Chemistry of Sulfate and Alternative SAI Materials

4.3.1. Need for Alternative SAI Materials

As previously discussed, the two largest first-order stratospheric risks of SAI with sulfate aerosol are ozone depletion and stratospheric heating. For sulfate aerosol the relative magnitude of these two risks can be adjusted if the size distribution can be controlled, e.g., via the AM-H_2SO_4 approach. It is worth noting that the impact on stratospheric ozone may be greatly reduced in the future if reactive halogen concentrations are lower. In contrast, the impact of stratospheric heating will not change. This represents a risk with a poorer understanding of its consequences, which makes it highly desirable to minimize stratospheric heating and resulting dynamic response. Therefore, it is important to investigate alternative SAI materials.

The properties of the “ideal” SAI material is (i) no absorption of radiation, i.e., purely scattering aerosol both fresh and aged, (ii) chemically inert, i.e., no direct impact of this material on stratospheric composition, and (iii) minimal down-stream effects, i.e., no impact on cirrus or other clouds, no environmental impact on deposition on the ground, etc. In reality, it is unlikely that a material with no impacts exists and rather the question is which materials can minimize these impacts. There have been a number of studies investigating

SAI materials in this context. High refractive index materials have been suggested as they reduce the mass of material that have to be lofted (Ferraro et al., 2015; Ferraro et al., 2011; Pope et al., 2012; Keith et al., 2016; Dai et al., 2020; Weisenstein et al., 2015). This largely cost-driven perspective is not a motivation for our work. In contrast, one of the goals of SCoPEx is to decrease the uncertainty in SRM models that use calcium carbonate SAI. The rationale for the choice of calcium carbonate as well as the approach to evaluate some of these risks is described in the following sections.

4.3.2. Unreactive Alternative SAI Materials

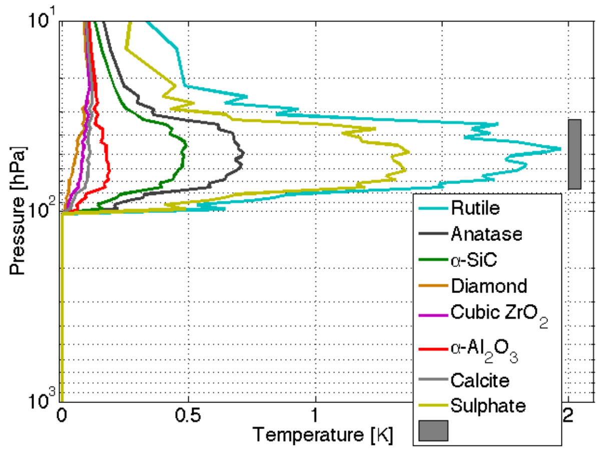


Figure 12: Comparison of stratospheric heating for different materials. Diamond has the lowest impact, although cubic zirconia and calcite are very similar. Sulfate and rutile result in much larger heating. (Dykema et al., 2016)

Diamond is probably the material with the best properties for SAI from a purely stratospheric perspective. Diamond has no absorption features in the short-wave range nor in the (terrestrial) longwave spectrum, as we also verified with commercial submicron diamond in our laboratory studies. Thus, diamond triggers the minimal possible dynamical response (see Figure 12). In addition, diamond should have ideal chemical properties. Hydrogen-terminated diamond surfaces are extremely inert and hydrophobic, precluding the ozone destroying chemistry initiated on sulfuric acid surfaces. The surface itself is also resistant to concentrated sulfuric acid. Exposure to OH radicals would probably slowly make the surface more hydrophilic. From a purely stratospheric perspective the only first-order risk of diamond would be increased ozone loss from the increased sulfuric acid surface area resulting from coagulation with background sulfate aerosol.

4.3.3. Reactive Alternative SAI Materials: The Case for Calcium Carbonate

Although the impact on cloud properties and the risk to Earth's surface from deposition of SAI diamond is likely very low, it could be preferable to have a material that dissolved easily in water, hence not persisting for long times outside of the stratosphere. It would also be preferable to have a material that is naturally abundant at Earth's surface. In addition, it would be ideal to overcome increased ozone loss due to coagulation by using a reactive aerosol. We therefore propose calcium carbonate as a prototype alternate SAI material for the following reasons: First, its optical properties are nearly equal to diamond and stratospheric heating and resulting dynamic response should be negligible compared to sulfate (Figure 12). Second, carbonates are typically quite reactive with acids, especially with concentrated sulfuric acid (Figure 13). Hence, calcium carbonate will neutralize upon coagulation with sulfate aerosol eliminating the acidic surfaces resulting from coagulation of diamond and sulfate aerosol. Of course, the reactivity of calcium carbonate also makes model predictions with calcium carbonate more complex, and the uncertainty added by this represents a substantial challenge. The evolution of chemical and optical aerosol properties has to be modeled over its stratospheric lifetimes. One of the key research questions that SCoPEx will help address is whether the reactivity of calcium carbonate and the evolution of its chemical and optical properties and those of the surrounding gas-phase correspond to the detailed hypothesis laid out below. To this end, SCoPEx will compare observations of the chemical evolution of calcium carbonate, as well as the gas-phase, with those of a model based on known properties of calcium carbonate and recent laboratory experiments (Dai et al., 2020). This will provide a real-world evaluation of kinetic parameters, such as heterogeneous uptake coefficients derived from the laboratory studies, that will enable GCMs to include reliable parameterizations of the stratospheric impacts of calcium carbonate SAI.

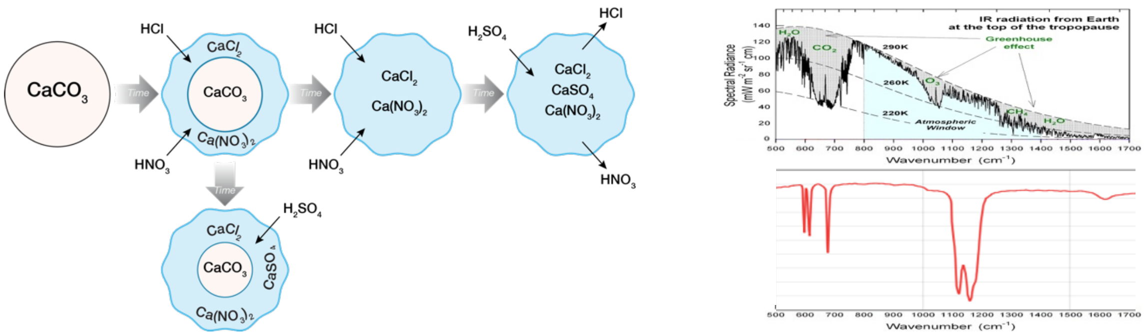


Figure 13: The left panel shows schematic of potential chemical reactivity of calcium carbonate in the stratosphere. The right panel shows the atmospheric windows in the terrestrial infrared (top) as well as the infrared absorption spectrum of calcium sulfate (bottom). The position of the 1150 cm^{-1} sulfate in part explains the stratospheric heating effect of sulfuric acid.

4.3.3.1. Optical Properties

Based on well-established chemistry, the reaction of sulfuric acid aerosol with calcium carbonate can be assumed to go to completion, i.e., be reagent limited. The optical properties of calcium sulfate in the terrestrial infrared are similar to those of sulfuric acid with only slight differences in relative band intensities and wavelengths (Figure 13 right hand inset). This is important as it implies that there will be no large first-order changes in stratospheric heating from changing background sulfuric acid to calcium sulfate. There are higher order impacts due to slight differences in the absorption of sulfuric acid, which has some liquid water compared to calcium sulfate. There are also numerous forms of calcium sulfate (anhydrite, bassanite, gypsum, etc.). However, the resulting differences are much smaller than introducing an absorbing material via SAI.

4.3.3.2. Chemical Properties

Predicting the evolution of the chemical properties of calcium carbonate under stratospheric conditions is more challenging. It is certain that calcium carbonate does not have the same heterogeneous reactions that activate ozone destroying substances as sulfuric acid. Figure 13 shows a schematic of the expected reactivity. Calcium carbonate is expected to react with acidic substances neutralizing them, forming salts and carbon dioxide. These acid neutralizing reactions can deplete gas-phase HNO_3, HCl, etc. There are a large number of ozone destroying catalytic cycles involving NOx, chlorine and other halogens, which are altitude (and latitude) dependent. NOx can be produced via HNO_3 photolysis and lost via heterogeneous reaction of N_2O_5. It participates both in ozone destroying catalytic cycles and is important for deactivation of ozone destroying halogen radicals. Thus, knowledge of the heterogeneous reaction rates of numerous substances with calcium carbonate are required to predict the impact it will have on stratospheric composition.

However, until the recent study by Dai et al. in our laboratory and Huynh et al., no heterogeneous chemistry studies of calcium carbonate under stratospheric conditions had been conducted, to our knowledge, although there exists a rich data set under tropospheric conditions (Dai et al., 2020; Huynh et al. 2021). These previous results highlight that reactive solid aerosols are indeed more complex than liquid sulfuric acid: Dai et al. observed moderate initial uptake of the gas-phase acids HCl and HNO_3 on fresh calcium carbonate, as the dry stratospheric conditions already make uptake coefficients lower than under typical tropospheric conditions. Although not important for the impact of calcium carbonate aerosol over stratospheric lifetime, the initial uptake coefficients of Huynh et al. and Dai et al. differ by 3-4 orders of magnitude difference, bringing the large uncertainty introduced by reactive aerosol into sharp focus. An additional large difference to liquid aerosol is that the surface of the solid calcium carbonate passivates, drastically reducing the uptake coefficients of HCl and HNO_3. Hence, based on the Dai et al. laboratory study, calcium carbonate rapidly becomes effectively unreactive with respect to uptake of these gas-phase acids, an important finding that confirms calcium carbonate as a good candidate as alternate SAI material. In addition, calcium carbonate particles are abundant at Earth's surface due to windblown mineral dust. And the small calcium carbonate SAI particles should dissolve rapidly in water. This does not exclude risks associated with the deposition of calcium carbonate SAI particles or impacts on clouds (Cziczo et al., 2019). However, due to its abundance at the Earth's surface, there already exists a large knowledge base for its environmental impacts in contrast to, e.g., diamond. Further laboratory work is required to study especially the $ClONO_2$ + HCl and N_2O_5 hydrolysis reactions on fresh and aged calcium carbonate. However, the existing results prepare the stage for studying them in the real stratospheric environment as outlined below. Figure 14 shows results of the AER 2-D chemistry-transport-aerosol model for annual average ozone column changes of calcium carbonate SAI compared to a control for 2040. Ignoring the passivation of calcium carbonate (thk-ind) results in increases in ozone columns from calcium carbonate SAI whereas the inclusion of passivation can either result in very little ozone column change or losses in the Southern Hemisphere, depending how the $ClONO_2$+HCl is parameterized. Either of the two, more realistic, passivation scenarios result in significantly lower ozone loss than the equivalent amount of sulfate SAI, consistent with the hypothesis.

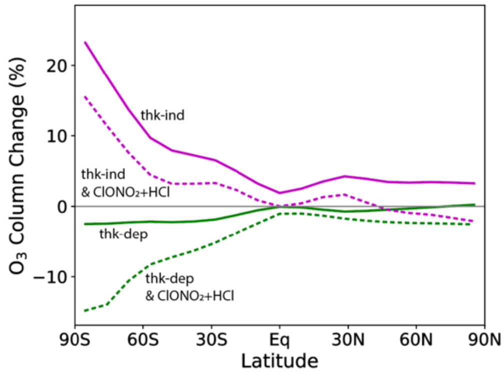


Figure 14: Shows the role of passivation and the heterogeneous $ClONO_2$+HCl reaction on ozone column change using the AER 2-D model taken from Dai et al. 2020. Inclusion of this reaction with the same rate as measured for Al_2O_3 results in a substantial reduction in ozone for scenarios including, thk-ind, or excluding passivation, thk-dep.

4.3.4. Need for SCoPEx Calcium Carbonate Plume Studies

One of the challenges for alternate SAI aerosol is the lack of materials such as calcium carbonate in the stratosphere. The only way to then study these materials in the actual stratosphere is via deliberate stratospheric injection of a small amount of these materials. In environmental studies, including stratospheric studies, it is not possible to rely purely on laboratory studies. For example, flights on the NASA ER-2 into the polar vortex over Antarctica provided the ability to test whether laboratory-derived reaction mechanisms were able to capture real-world ozone destruction chemistry. Without these flights, the level of confidence in the model predictions would have been much lower, and for good reason. It is not clear that a given experimental setup in the laboratory can faithfully capture the entire complexity of the real stratosphere; only field observations are able to provide this. For a number of natural stratospheric processes, remote observations can provide important information in addition to in situ aircraft or balloon. However, these are only possible when large-scale phenomena are at work.

Since there are no natural calcium carbonate plumes in the stratosphere that would even allow for in situ observations, intentional injection is necessary to perform these studies. Calcium carbonate injections will allow SCoPEx to provide invaluable observations as it will quantitively test the mechanisms determined in the laboratory. As stated above, there is a need for more laboratory studies, however, there is good reason to proceed with the planning of SCoPEx calcium carbonate experiments. First, by the time of the first injection experiments, additional studies should have been conducted. In addition, N_2O_5 uptake coefficients used in the model are likely a very good estimation as similar values have been found for different solid materials, e.g., Al_2O_3 and SiO_2 (Molina et al., 1997). In addition, even with these additional lab determined mechanisms, the same type of experiments as proposed here will still have to be conducted, as we expect these reactions to not make a significant difference. In other words, they will not be a deciding factor about the viability of calcium carbonate as an alternate SAI material. Only field experiments will help shed insight into these questions. In summary, there is a critical need for evaluating not just the aerosol microphysics (goal 2) but also the stratospheric chemistry of calcium carbonate due to the promise it holds as a lower risk SAI material.

4.3.5. SCoPEx Experimental Design and Analysis of Chemical Calcium Carbonate Plume Evolution

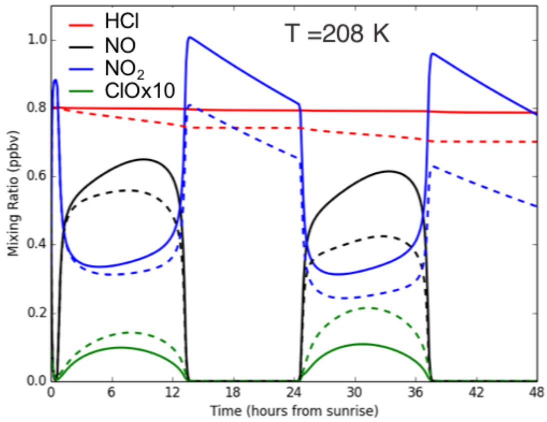


Figure 15: Solid lines: background 2µm^2 cm^{-3} sulfate 5ppm$_v$ H_2O. Dashed lines: plume 15µm^2 cm^{-3} sulfate 10 ppm$_v$ H_2O.

The experiments will again follow the standard concept of operations as under goal 2. In order to determine optimal injection rates, we will include chemical reactions in the plume model, updated with the newest mechanisms available at that time. Figure 15 shows the evolution of an air mass perturbed by a sulfate aerosol injection over multiple days, i.e., significantly longer than the initial SCoPEx experiments. Significant changes in HCl and NOx can be observed already over short time periods and these are easily detectable with existing instrumentation. For this science goal, it is desirable to measure aerosol composition and size distribution as well as key gas-phase chemical species, especially HCl, NOx and water. Therefore, this science goal requires a much larger set of instruments. In addition, the equivalent model to Figure 15 for calcium carbonate is informed by the results of science goal 2. The work of Dai et al. provides kinetic parameters needed for this model, and reactions for which there are no laboratory data to date are parameterized using close analogues and conditions, e.g., $ClONO_2$ + HCl are parameterized using the results for alumina (and silica) from Molina et al. (1997). One key question is whether the changes in HCl and NOx will indeed be smaller for calcium carbonate than those for sulfate shown in the figure above, which would confirm the hypothesis for calcium carbonate as a potential alternate SAI material.

In summary, SCoPEx experiments using calcium carbonate injections will provide a unique evaluation as to whether calcium carbonate indeed is an alternate SAI material that could substantially reduce risk from SAI compared to sulfate. Follow-up studies will be needed. For example, improved chemical and aerosol microphysics models will provide improved models of the chemical and physical evolution of calcium carbonate, which likely will motivate specific laboratory investigations. These will provide information for SCoPEx studies using “stratospherically aged” calcium carbonate as precursor for injection that can then be used to compare whether the laboratory mechanisms of this aged calcium carbonate agree with that found in the real stratospheric environment.

5. Data Management Plan and Dissemination of Results

Products of the research. The data generated during this project consists of meteorological, navigational, telemetry, and a variety of instrumentation data, in particular aerosol size distributions as well as chemical composition data during later science flights. In addition, there will be model data on plume chemical evolution.

Access to data, data sharing practices, and policies and dissemination of results. Data relevant for scientific analysis will be made public within 60 days of the end of flight. This taw data will be made public with appropriate warnings that it has not undergone QA/QC. The email address of users will be recorded so that they can be automatically notified when revised versions become available. Based on previous experiences with stratospheric airborne campaigns, this is typically 6-15 months after the flight depending on the type of data, e.g., the amount of calibration and data workup required. We have chosen to make raw data available rapidly—going far beyond what is typical for stratospheric science missions—because of the public scrutiny of SCoPEx and because of the broad commitment to Open Access data principles articulated by Harvard's Solar Geoengineering Research Program which is funding SCoPEx.

Principal Investigators (PI) and their groups have an excellent track record with presenting their work at major national and international conferences and workshops. All data that go into key analyses and figures in the group's publications will be made publicly available via the PI's group website. All publications resulting from this project will be posted on the PI's webpage (https://projects.iq.harvard.edu/keutschgroup/publications). Preprints of manuscripts submitted for publication as well as the underlying data will also be posted on Harvard's Dash manuscript repository. Publications will be made in open access formats.

Archiving of data. All data acquisition/storage computers in the PI's group are automatically backed up daily, both wirelessly to a server elsewhere on campus, and/or to a cloud server. Both of these processes ensure that data will not be lost and enable rapid access to the data. The file naming system used for all software (which includes the date of the experiment) ensures straightforward retrieval and use of archived data. Group laptops are also backed up daily, ensuring that analyzed data are archived as well.

6. SCoPEx Research Team Biographies

Frank Keutsch was born in Tübingen, Germany and received his Diplom in chemistry from the Technische Universität München, Germany, under the supervision of Vladimir E. Bondybey in 1997. He received his PhD in physical chemistry from the University of California at Berkeley in 2001. His graduate research was conducted under the direction of Richard J. Saykally and focused on vibration−rotation−tunneling spectroscopy and hydrogen-bond-breaking dynamics in water clusters. After working on stratospheric chemistry in the Department of Chemistry and Chemical Biology at Harvard University under the direction of James G. Anderson, he started his independent academic career in 2005 at the University of Wisconsin-Madison. He then moved to his current position as Stonington Professor of Engineering and Atmospheric Science at Harvard University in the Paulson School of Engineering and Applied Sciences and the Department of Chemistry and Chemical Biology and he has held numerous visiting professor positions. Keutsch Group research combines laboratory and field experiments with instrument development to investigate fundamental mechanisms of anthropogenic influence on atmospheric composition within the context of impacts on climate, humans and the environment. Keutsch's main focus has been on understanding how *unintentional* emissions of pollutants such as nitrogen oxides, sulfur dioxide, and hydrocarbons have changed key chemical pathways controlling ozone and particulate matter, two key pollutants affecting human health and climate. Keutsch has been the PI of numerous research grants for this research and currently is the deputy-PI for the NASA-EVS3 DCOTSS campaign. Keutsch has also been focusing on improving the understanding of how *intentional* emissions within the context of stratospheric solar radiation modification could impact the protective stratospheric ozone layer and stratospheric dynamics and climate, and how known risks can be better quantified or reduced. He is currently the PI of SCoPEx. Keutsch has received awards for his teaching, which spans a wide range of courses including introductory chemistry, engineering design and atmospheric chemistry.

David Keith has worked near the interface between climate science, energy technology, and public policy since 1991. He received his B.Sc. in physics from the University of Toronto in 1986 and received his PhD in experimental physics from the Massachusetts Institute of Technology in 1991 under the supervision of David Prichard. He took first prize in Canada's national physics prize exam, won MIT's prize for excellence in experimental physics, and was one of TIME Magazine's Heroes of the Environment. David is Professor of Applied Physics at the Harvard School of Engineering and Applied Sciences and Professor of Public Policy at the Harvard Kennedy School, and founder of Carbon Engineering, a Canadian company developing technology to capture CO_2 from ambient air to make carbon-neutral hydrocarbon fuels. Best known for his work on the science, technology, and public policy of solar geoengineering, David led the development of Harvard's Solar Geoengineering Research Program, a Harvard-wide interfaculty research initiative. His work has ranged from the climatic impacts of large-scale wind power to an early critique of the prospects for hydrogen fuel. David's hardware engineering work includes the first interferometer for atoms, a high-accuracy infrared spectrometer for NASA's ER-2, and the development of Carbon Engineering's air contactor and overall process design. On SCoPEx, he is the faculty lead for platform design and engineering. David teaches science and technology policy, climate science, and solar geoengineering. He has reached students worldwide with an edX energy course. David is author of >200 academic publications with total citation count of >15,000. He has written for the public in op-eds and *A Case for Climate Engineering*. David splits his time between Cambridge, Massachusetts and Canmore, Alberta.

Norton Allen is Head Software Engineer for the Anderson, Keith, and Keutsch groups in the Harvard John A. Paulson School of Engineering and Applied Sciences. Working closely with electrical and mechanical engineering, he is responsible for the design and deployment of software for data acquisition and control on all flight instruments. He has successfully deployed over two dozen instruments and supported field deployments in locations around the world. He received an AB cum laude from Harvard College, studying math, applied math, computer science, and physics.

John Dykema is a Project Scientist at the Harvard John A. Paulson School of Engineering and Applied Sciences and the LIDAR principal investigator on SCoPEx. His main interests are atmospheric radiation and remote sensing instrumentation, with an emphasis on development of novel, compact LIDARS for trace gas and aerosol measurement. John earned his AB in physics from UC Berkeley and his PhD in applied physics from Harvard University, where his dissertation focused on developing a new airborne infrared sounder that was a prototype for a climate-focused atmospheric radiation mission. He is participating in the NASA DCOTSS mission as the principal investigator for the POPS optical particle counter and as a member of the DCOTSS aerosol science subgroup. He also collaborates with several external organizations in designing and simulating new LIDAR prototypes, incorporating emerging laser and optical technology. John leads the engineering development and data analysis for the SCoPEx LIDAR and works on the radiative and micrometeorological science aspects of the SCoPEx mission.

Mike Greenberg is the Lead Optical-Mechanical engineer for the Anderson, Keith, and Keutsch groups in the Harvard John A. Paulson School of Engineering and Applied Sciences. He is responsible for the mechanical development and implementation of flight and laboratory based instrumentation, equipment packaging, documentation, and platform integration. Working closely with the electrical, software, and science team members, he has over 20 years of experience developing, delivering, and supporting designs and has been on more than a dozen airborne campaigns with the ER-2, WB-57, and DA-42 aircraft platforms and with stratospheric balloons. Mike received a BSME from Tufts University and a MSME from Stanford University. His additional work experiences include time spent Argonne National Laboratory and The Raytheon Company.

Michael Litchfield is the Senior Engineering Lead for Climate Research in the Anderson, Keith, Keutsch groups at the Harvard John A. Paulson School of Engineering and Applied Sciences and the engineering lead on the SCoPEx Flight Platform development program. He and the rest of the engineering team are focused on taking high level SCoPEx flight platform requirements through the design, fabrication, assembly, test, and validation processes. Michael earned his BS and MS degrees in Electrical Engineering specializing in controls and communications systems at Worcester Polytechnic Institute. Prior to joining the lab to assume this role, Michael worked for over 30 years in industry across 5 start-ups leading their various engineering teams in bringing first products to market where those markets included; X-ray Semiconductor Lithography, 3D Ultrasound

Medical imaging , X-ray 2D Projection / 3D CT Airport Baggage Security Imaging, and 4D (3D movies) mmWave Personnel Security imaging.

Craig Mascarenhas is a mechanical engineer for the Anderson, Keith, and Keutsch groups in the Harvard John A. Paulson School of Engineering and Applied Sciences. He is responsible for the mechanical design and integration of instrumentation, equipment packaging, and aerodynamic analysis of flight systems. He has previously been involved in instrument design for airborne campaigns with the ER-2 and stratospheric balloons. Craig received a BASc from the University of Toronto and an SM from MIT. His additional work experiences include engineering roles in the nuclear, biotech, and hydro-power industries.

Terry Martin is an electronics technician with the Anderson, Keith, and Keutsch research groups. She has worked on electrical build up and documentation of numerous scientific experiments over the course of the 42 years she has been with the group and is presently helping with the electronic assembly and wiring of the SCoPEx instrument.

Marco Rivero is a senior Electrical Engineer in the Anderson, Keith, and Keutsch groups in the Harvard John A. Paulson School of Engineering and Applied Sciences. As such, he has been primarily involved in the electrical engineering design, fabrication, and testing of the SCoPEx platform and payload instrumentation since inception. Marco holds a BS in Microelectronic Engineering from Rochester Institute of Technology and a MS in Electrical Engineering from Tufts University. During his 25 years with the group, Marco has been involved in the electronics and systems design of 14 airborne instruments and supported their deployment in over 20 NASA national and international field campaigns; most recently, a HCl instrument deployment out of NASA's Columbia Scientific Balloon Facility in Fort Sumner NM in August of 2018.

Yomay Shyur is a Postdoctoral Fellow at the Harvard John A. Paulson School of Engineering and Applied Sciences and a project manager and project scientist on SCoPEx. She leads technical project coordination, works on science instrument design and analysis, and assists with platform engineering tasks. Yomay earned her BA in physics from Wellesley College and her PhD in physics from the University of Colorado Boulder, where her dissertation focused on developing new experimental methods of manipulating cold molecules using high-voltage electrodes and laser detection techniques.

References

Anderson, M. R., Miake-Lye, R. C., Brown, R. C., & Kolb, C. E. (1996). Calculation of exhaust plume structure and emissions of the ER 2 aircraft in the stratosphere. *Journal of Geophysical Research: Atmospheres*, *101*(D2), 4025–4032. https://doi.org/10.1029/95JD02366

Bala, G., Duffy, P. B., & Taylor, K. E. (2008). Impact of geoengineering schemes on the global hydrological cycle. *Proceedings of the National Academy of Sciences*, *105*(22), 7664–7669. https://doi.org/10.1073/pnas.0711648105

Balluch, M. G., & Haynes, P. H. (1997). Quantification of lower stratospheric mixing processes using aircraft data. *Journal of Geophysical Research: Atmospheres*, *102*(D19), 23487–23504. https://doi.org/10.1029/97JD00607

Benduhn, F., Schallock, J., & Lawrence, M. G. (2016). Early growth dynamical implications for the steerability of stratospheric solar radiation management via sulfur aerosol particles. *Geophysical Research Letters*, *43*(18), 9956–9963. https://doi.org/10.1002/2016GL070701

Butchart, N., Anstey, J. A., Hamilton, K., Osprey, S., McLandress, C., Bushell, A. C., Kawatani, Y., Kim, Y.-H., Lott, F., Scinocca, J., Stockdale, T. N., Andrews, M., Bellprat, O., Braesicke, P., Cagnazzo, C., Chen, C.-C., Chun, H.-Y., Dobrynin, M., Garcia, R. R., … Yukimoto, S. (2018). Overview of experiment design and comparison of models participating in phase 1 of the SPARC Quasi-Biennial Oscillation initiative (QBOi). *Geoscientific Model Development*, *11*(3), 1009–1032. https://doi.org/10.5194/gmd-11-1009-2018

Conway, J. P., Bodeker, G. E., Waugh, D. W., Murphy, D. J., Cameron, C., & Lewis, J. (2019). Using Project Loon Superpressure Balloon Observations to Investigate the Inertial Peak in the Intrinsic Wind Spectrum in the Midlatitude Stratosphere. *Journal of Geophysical Research: Atmospheres*, *124*(15), 8594–8604. https://doi.org/10.1029/2018JD030195

Crutzen, P. J. (2006). Albedo Enhancement by Stratospheric Sulfur Injections: A Contribution to Resolve a Policy Dilemma? *Climatic Change*, *77*(3), 211. https://doi.org/10.1007/s10584-006-9101-y

Cziczo, D. J., Wolf, M. J., Gasparini, B., Münch, S., & Lohmann, U. (2019). Unanticipated Side Effects of Stratospheric Albedo Modification Proposals Due to Aerosol Composition and Phase. *Scientific Reports*, *9*(1), 18825. https://doi.org/10.1038/s41598-019-53595-3

Dai, Z., F. Keutsch, D. Weisenstein, and D. W. Keith. (2020). Experimental reaction rates constrain estimates of ozone response to calcium carbonate geoengineering, *Manuscript submitted for publication.*

Dykema, J., Keith, D., Anderson, J. G., & Weisenstein, D. (2014). Stratospheric controlled perturbation experiment (SCoPEx): A small-scale experiment to improve understanding of the risks of solar geoengineering. *Philosophical Transactions of the Royal Society A*, *372*. https://doi.org/10.1098/rsta.2014.0059

Dykema, J., Keith, D., & Keutsch, F. (2016). Improved aerosol radiative properties as a foundation for solar geoengineering risk assessment. *Geophysical Research Letters*. http://onlinelibrary.wiley.com/doi/10.1002/2016GL069258/full

Fahey, D. W., Keim, E. R., Boering, K. A., Brock, C. A., Wilson, J. C., Jonsson, H. H., Anthony, S., Hanisco, T. F., Wennberg, P. O., Miake-Lye, R. C., Salawitch, R. J., Louisnard, N., Woodbridge, E. L., Gao, R. S., Donnelly, S. G., Wamsley, R. C., Del Negro, L. A., Solomon, S., Daube, B. C., … Chan, K. R. (1995a). Emission Measurements of the Concorde Supersonic Aircraft in the Lower Stratosphere. *Science*, *270*(5233), 70–74.

Fahey, D. W., Keim, E. R., Woodbridge, E. L., Gao, R. S., Boering, K. A., Daube, B. C., Wofsy, S. C., Lohmann, R. P., Hintsa, E. J., Dessler, A. E., Webster, C. R., May, R. D., Brock, C. A., Wilson, J. C., Miake-Lye, R. C., Brown, R. C., Rodriguez, J. M., Loewenstein, M., Proffitt, M. H., … Chan, K. R. (1995b). In situ observations in aircraft exhaust plumes in the lower stratosphere at midlatitudes. *Journal of Geophysical Research: Atmospheres*, *100*(D2), 3065–3074. https://doi.org/10.1029/94JD02298

Farquhar, G. D., & Roderick, M. L. (2003). Pinatubo, Diffuse Light, and the Carbon Cycle. *Science*, *299*(5615), 1997–1998. https://doi.org/10.1126/science.1080681

Ferraro, A. J., Charlton-Perez, A. J., & Highwood, E. J. (2015). Stratospheric dynamics and midlatitude jets under geoengineering with space mirrors and sulfate and titania aerosols. *Journal of Geophysical Research: Atmospheres*, *120*(2), 414–429. https://doi.org/10.1002/2014JD022734

Ferraro, A. J., Highwood, E. J., & Charlton-Perez, A. J. (2011). Stratospheric heating by potential geoengineering aerosols. *Geophysical Research Letters*, *38*(24). https://doi.org/10.1029/2011GL049761

Gao, R. S., Telg, H., McLaughlin, R. J., Ciciora, S. J., Watts, L. A., Richardson, M. S., Schwarz, J. P., Perring, A. E., Thornberry, T. D., Rollins, A. W., Markovic, M. Z., Bates, T. S., Johnson, J. E., & Fahey, D. W. (2016). A light-weight, high-sensitivity particle spectrometer for PM2.5 aerosol measurements. *Aerosol Science and Technology*, *50*(1), 88–99. https://doi.org/10.1080/02786826.2015.1131809

Gerding, M., Theuerkauf, A., & Suminska, O. (2009). Balloon-borne hot wire anemometer for stratospheric turbulence soundings. *Proceedings of the 19th ESA Symposium on European Rocket and Balloon Programmes and Related Research, SP-671*.

Golja, C. M., L. W. Chew, J. A. Dykema, and D. W. Keith. (2020). Aerosol Dynamics in the Near Field of the SCoPEx Stratospheric Balloon Experiment, *Manuscript submitted for publication.*

Gu, L., Baldocchi, D. D., Wofsy, S. C., Munger, J. W., Michalsky, J. J., Urbanski, S. P., & Boden, T. A. (2003). Response of a Deciduous Forest to the Mount Pinatubo Eruption: Enhanced Photosynthesis. *Science*, *299*(5615), 2035–2038. https://doi.org/10.1126/science.1078366

Gu, L., Baldocchi, D., Verma, S. B., Black, T. A., Vesala, T., Falge, E. M., & Dowty, P. R. (2002). Advantages of diffuse radiation for terrestrial ecosystem productivity. *Journal of Geophysical Research: Atmospheres*, *107*(D6), ACL 2-1-ACL 2-23.

https://doi.org/10.1029/2001JD001242
Haack, A., Gerding, M., & Lübken, F.-J. (2014). Characteristics of stratospheric turbulent layers measured by LITOS and their relation to the Richardson number. *Journal of Geophysical Research: Atmospheres*, *119*(18), 10,605-10,618. https://doi.org/10.1002/2013JD021008
Haynes, P. H. (2005). Transport and Mixing in the Atmosphere. In W. Gutkowski & T. A. Kowalewski (Eds.), *Mechanics of the 21st Century* (pp. 139–152). Springer Netherlands. https://doi.org/10.1007/1-4020-3559-4_8
Heinold, B., Baars, H., Barja, B. Christensen, W., Kubin, A., Ohneiser, K., Schepanski, K., Schutgens, N., Senf, F., Schrödner, R., Villanueva, D., & Tegen, I. (2021). Important role of stratospheric injection higher for the distribution and radiative forcing of smoke aerosol from the 2019/2020 Australian wildfires. *Atmospheric Chemistry and Physics Discussions*, https://doi.org/10.5194/acp-2021-862
Holben, B. N., Eck, T. F., Slutsker, I., Tanré, D., Buis, J. P., Setzer, A., Vermote, E., Reagan, J. A., Kaufman, Y. J., Nakajima, T., Lavenu, F., Jankowiak, I., & Smirnov, A. (1998). AERONET—A Federated Instrument Network and Data Archive for Aerosol Characterization. *Remote Sensing of Environment*, *66*(1), 1–16. https://doi.org/10.1016/S0034-4257(98)00031-5
Homeyer, C. R., Bowman, K. P., Pan, L. L., Atlas, E. L., Gao, R.-S., & Campos, T. L. (2011). Dynamical and chemical characteristics of tropospheric intrusions observed during START08. *Journal of Geophysical Research: Atmospheres*, *116*(D6). https://doi.org/10.1029/2010JD015098
Hoppe, C. M., Hoffmann, L., Konopka, P., GrooÃ, J.-U., Ploeger, F., Günther, G., Jöckel, P., & Müller, R. (2014). The implementation of the CLaMS Lagrangian transport core into the chemistry climate model EMAC 2.40.1: Application on age of air and transport of long-lived trace species. *Geoscientific Model Development*, *7*(6), 2639–2651. https://doi.org/10.5194/gmd-7-2639-2014
Huynh, H. N., & McNeill, V. F. (2021). Heterogeneous Reactivity of HCl on $CaCO_3$ Aerosols at Stratospheric Temperature. *ACS Earth Space Chem.*, *5*, 1896-1901. https://doi.org/10.1021/acsearthspacechem.1c00151
Jablonowski, C., & Williamson, D. L. (2011). The Pros and Cons of Diffusion, Filters and Fixers in Atmospheric General Circulation Models. In P. Lauritzen, C. Jablonowski, M. Taylor, & R. Nair (Eds.), *Numerical Techniques for Global Atmospheric Models* (pp. 381–493). Springer. https://doi.org/10.1007/978-3-642-11640-7_13
Ji, D., Fang, S., Curry, C. L., Kashimura, H., Watanabe, S., Cole, J. N. S., Lenton, A., Muri, H., Kravitz, B., & Moore, J. C. (2018). Extreme temperature and precipitation response to solar dimming and stratospheric aerosol geoengineering. *Atmospheric Chemistry and Physics*, *18*(14), 10133–10156. https://doi.org/10.5194/acp-18-10133-2018
Keith, D., Weisenstein, D., Dykema, J., & Keutsch, F. (2016). Stratospheric Solar Geoengineering without Ozone Loss. *Proceedings of the National Academy of Sciences*. http://www.pnas.org/content/113/52/14910.full

Kravitz, B., Robock, A., Boucher, O., Schmidt, H., Taylor, K. E., Stenchikov, G., & Schulz, M. (2011). The Geoengineering Model Intercomparison Project (GeoMIP). *Atmospheric Science Letters*, *12*(2), 162–167. https://doi.org/10.1002/asl.316

Legras, B., Pisso, I., Berthet, G., & Lefèvre, F. (2005). Variability of the Lagrangian turbulent diffusion in the lower stratosphere. *Atmospheric Chemistry and Physics*, *5*(6), 1605–1622. https://doi.org/10.5194/acp-5-1605-2005

Määttänen, A., Merikanto, J., Henschel, H., Duplissy, J., Makkonen, R., Ortega, I. K., & Vehkamäki, H. (2018). New Parameterizations for Neutral and Ion-Induced Sulfuric Acid-Water Particle Formation in Nucleation and Kinetic Regimes. *Journal of Geophysical Research: Atmospheres*, *123*(2), 1269–1296. https://doi.org/10.1002/2017JD027429

Maruca, B. A., Marino, R., Sundkvist, D., Godbole, N. H., Constantin, S., Carbone, V., & Zimmerman, H. (2017). Overview of and first observations from the TILDAE High-Altitude Balloon Mission. *Atmospheric Measurement Techniques*, *10*(4), 1595–1607. https://doi.org/10.5194/amt-10-1595-2017

McCormick, M. P., Thomason, L. W., & Trepte, C. R. (1995). Atmospheric effects of the Mt Pinatubo eruption. *Nature*, *373*(6513), 399–404. https://doi.org/10.1038/373399a0

Molina, M. J., Molina, L. T., Zhang, R., Meads, R. F., & Spencer, D. D. (1997). The reaction of ClONO2 with HCl on aluminum oxide. *Geophysical Research Letters*, *24*(13), 1619–1622. https://doi.org/10.1029/97GL01560

Murphy, D. M., Telg, H., Eck, T. F., Rodriguez, J., Stalin, S. E., & Bates, T. S. (2016). A miniature scanning sun photometer for vertical profiles and mobile platforms. *Aerosol Science and Technology*, *50*(1), 11–16. https://doi.org/10.1080/02786826.2015.1121200

Nakamura, N. (1996). Two-Dimensional Mixing, Edge Formation, and Permeability Diagnosed in an Area Coordinate. *Journal of the Atmospheric Sciences*, *53*(11), 1524–1537. https://doi.org/10.1175/1520-0469(1996)053<1524:TDMEFA>2.0.CO;2

National Academy of Sciences, National Academy of Engineering, & Institute of Medicine. (1992). *Policy Implications of Greenhouse Warming: Mitigation, Adaptation, and the Science Base*. The National Academies Press. https://doi.org/10.17226/1605

National Research Council. (2015). *Climate Intervention: Reflecting Sunlight to Cool Earth*. The National Academies Press. https://doi.org/10.17226/18988

National Academies of Sciences, Engineering, and Medicine (2021). Reflecting Sunlight: Recommendations for Solar Geoengineering Research and Research Governance. Washington, DC: The National Academies Press. https://doi.org/10.17226/25762

Niemeier, U., & Timmreck, C. (2015). What is the limit of climate engineering by stratospheric injection of SO2? *Atmospheric Chemistry and Physics; Katlenburg-Lindau*, *15*(16), 9129. http://dx.doi.org.ezp-prod1.hul.harvard.edu/10.5194/acp-15-9129-2015

Orsolini, Y., Simon, P., & Cariolle, D. (1995). Filamentation and layering of an idealized tracer by observed winds in the lower stratosphere. *Geophysical Research Letters*, *22*(7),

839–842. https://doi.org/10.1029/95GL00389
Phoenix, D. B., Homeyer, C. R., & Barth, M. C. (2017). Sensitivity of simulated convection-driven stratosphere-troposphere exchange in WRF-Chem to the choice of physical and chemical parameterization. *Earth and Space Science*, *4*(8), 454–471. https://doi.org/10.1002/2017EA000287
Pierce, J., Weisenstein, D., Heckendorn, P., Peter, T., & Keith, D. (2010). Efficient formation of stratospheric aerosol for climate engineering by emission of condensible vapor from aircraft. *Geophysical Research Letters*, *37*. https://doi.org/10.1029/2010GL043975
Pope, F. D., Braesicke, P., Grainger, R. G., Kalberer, M., Watson, I. M., Davidson, P. J., & Cox, R. A. (2012). Stratospheric aerosol particles and solar-radiation management. *Nature Climate Change*, *2*(10), 713–719. https://doi.org/10.1038/nclimate1528
Prather, M., & Jaffe, A. H. (1990). Global impact of the Antarctic ozone hole: Chemical propagation. *Journal of Geophysical Research: Atmospheres*, *95*(D4), 3473–3492. https://doi.org/10.1029/JD095iD04p03473
Rauthe, M., Gerding, M., & Lübken, F.-J. (2008). Seasonal changes in gravity wave activity measured by lidars at mid-latitudes. *Atmospheric Chemistry and Physics*, *8*(22), 6775–6787. https://doi.org/10.5194/acp-8-6775-2008
Richter, J., H., Tilmes, S., Glanville, A., Kravitz, B., MacMartin, D. G., Mills, M. J., Simpson, I. R., Vitt, F., Tribbia, J. J., & Lamarque, J.-F. (2018). Stratospheric Response in the First Geoengineering Simulation Meeting Multiple Surface Climate Objectives. *Journal of Geophysical Research: Atmospheres*, *123*(11), 5762–5782. https://doi.org/10.1029/2018JD028285
Robock, A. (2000). Volcanic eruptions and climate. *Reviews of Geophysics*, *38*(2), 191–219. https://doi.org/10.1029/1998RG000054
Ross, M. N., Ballenthin, J. O., Gosselin, R. B., Meads, R. F., Zittel, P. F., Benbrook, J. R., & Sheldon, W. R. (1997). In-situ measurement of Cl2 and O3 in a stratospheric solid rocket motor exhaust plume. *Geophysical Research Letters*, *24*(14), 1755–1758. https://doi.org/10.1029/97GL01592
Ross, M. N., Whitefield, P. D., Hagen, D. E., & Hopkins, A. R. (1999). In situ measurement of the aerosol size distribution in stratospheric solid rocket motor exhaust plumes. *Geophysical Research Letters*, *26*(7), 819–822. https://doi.org/10.1029/1999GL900085
Russell, L. M., Rasch, P. J., Mace, G. M., Jackson, R. B., Shepherd, J., Liss, P., Leinen, M., Schimel, D., Vaughan, N. E., Janetos, A. C., Boyd, P. W., Norby, R. J., Caldeira, K., Merikanto, J., Artaxo, P., Melillo, J., & Morgan, M. G. (2012). Ecosystem Impacts of Geoengineering: A Review for Developing a Science Plan. *AMBIO*, *41*(4), 350–369. https://doi.org/10.1007/s13280-012-0258-5
Schneider, A., Gerding, M., & Lübken, F.-J. (2015). Comparing turbulent parameters obtained from LITOS and radiosonde measurements. *Atmospheric Chemistry and Physics*, *15*(4), 2159–2166. https://doi.org/10.5194/acp-15-2159-2015

Schneider, Andreas, Wagner, J., Söder, J., Gerding, M., & Lübken, F.-J. (2017). Case study of wave breaking with high-resolution turbulence measurements with LITOS and WRF simulations. *Atmospheric Chemistry and Physics; Katlenburg-Lindau*, *17*(12), 7941–7954. https://doi.org/10.5194/acp-17-7941-2017

Schoeberl, M. R., & Bacmeister, J. T. (1993). Mixing Processes in the Extra Tropical Stratosphere. In M.-L. Chanin (Ed.), *The Role of the Stratosphere in Global Change* (pp. 135–152). Springer. https://doi.org/10.1007/978-3-642-78306-7_5

Simpson, I. R., Tilmes, S., Richter, J. H., Kravitz, B., MacMartin, D. G., Mills, M. J., Fasullo, J. T., & Pendergrass, A. G. (2019). The Regional Hydroclimate Response to Stratospheric Sulfate Geoengineering and the Role of Stratospheric Heating. *Journal of Geophysical Research: Atmospheres*, *124*(23), 12587–12616. https://doi.org/10.1029/2019JD031093

Söder, J., Gerding, M., Schneider, A., Dörnbrack, A., Wilms, H., Wagner, J., & Lübken, F.-J. (2019). Evaluation of wake influence on high-resolution balloon-sonde measurements. *Atmospheric Measurement Techniques*, *12*(8), 4191–4210. https://doi.org/10.5194/amt-12-4191-2019

Solomon, S., Dubee, K., Stone, K., & Degenstein, D. (2022). On the stratospheric chemistry of midlatitude wildfire smoke. *PNAS*, *119*(10), e2117325119. https://doi.org/10.1073/pnas.2117325119

Sparling, L. C., & Bacmeister, J. T. (2001). Scale dependence of tracer microstructure: PDFs, intermittency and the dissipation scale. *Geophysical Research Letters*, *28*(14), 2823–2826. https://doi.org/10.1029/2000GL012781

Stolarski, R. S., & Wesoky, H. L. (1993). *The atmospheric effects of stratospheric aircraft*. NASA. http://ntrs.nasa.gov/search.jsp?R=19930013868

Sukhodolov, T., Sheng, J.-X., Feinberg, A., Luo, B.-P., Peter, T., Revell, L., Stenke, A., Weisenstein, D. K., & Rozanov, E. (2018). Stratospheric aerosol evolution after Pinatubo simulated with a coupled size-resolved aerosol–chemistry–climate model, SOCOL-AERv1.0. *Geoscientific Model Development*, *11*(7), 2633–2647. https://doi.org/10.5194/gmd-11-2633-2018

Teller, E., Wood, L., & Hyde, R. (1996). *Global warming and ice ages: I. prospects for physics based modulation of global change* (UCRL-JC-128715; CONF-9708117-). Lawrence Livermore National Lab., CA (United States). https://www.osti.gov/biblio/611779-global-warming-ice-ages-prospects-physics-based-modulation-global-change

Theuerkauf, A., Gerding, M., & Lübken, F.-J. (2011). LITOS – a new balloon-borne instrument for fine-scale turbulence soundings in the stratosphere. *Atmospheric Measurement Techniques*, *4*(1), 55–66. https://doi.org/10.5194/amt-4-55-2011

Tilmes, S., Fasullo, J., Lamarque, J.-F., Marsh, D. R., Mills, M., Alterskjær, K., Muri, H., Kristjánsson, J. E., Boucher, O., Schulz, M., Cole, J. N. S., Curry, C. L., Jones, A., Haywood, J., Irvine, P. J., Ji, D., Moore, J. C., Karam, D. B., Kravitz, B., … Watanabe, S. (2013). The hydrological impact of geoengineering in the Geoengineering Model Intercomparison Project (GeoMIP). *Journal of Geophysical Research: Atmospheres*, *118*(19), 11,036-11,058. https://doi.org/10.1002/jgrd.50868

Tilmes, S., Richter, J. H., Kravitz, B., MacMartin, D. G., Mills, M. J., Simpson, I. R., Glanville, A. S., Fasullo, J. T., Phillips, A. S., Lamarque, J.-F., Tribbia, J., Edwards, J., Mickelson, S., & Ghosh, S. (2018). CESM1(WACCM) Stratospheric Aerosol Geoengineering Large Ensemble Project. *Bulletin of the American Meteorological Society*, *99*(11), 2361–2371. https://doi.org/10.1175/BAMS-D-17-0267.1

Torres, B., Dubovik, O., Toledano, C., Berjon, A., Cachorro, V. E., Lapyonok, T., Litvinov, P., & Goloub, P. (2014). Sensitivity of aerosol retrieval to geometrical configuration of ground-based sun/sky radiometer observations. *Atmospheric Chemistry and Physics*, *14*(2), 847–875. https://doi.org/10.5194/acp-14-847-2014

Vanneste, J. (2004). Small-Scale Mixing, Large-Scale Advection, and Stratospheric Tracer Distributions. *Journal of the Atmospheric Sciences; Boston*, *61*(22), 2749–2761.

Vattioni, S., Weisenstein, D., Keith, D., Feinberg, A., Peter, T., & Stenke, A. (2019). Exploring accumulation-mode H2SO4 versus SO2 stratospheric sulfate geoengineering in a sectional aerosol–chemistry–climate model. *Atmospheric Chemistry and Physics*, *19*.

Vehkamäki, H., Kulmala, M., Napari, I., Lehtinen, K. E. J., Timmreck, C., Noppel, M., & Laaksonen, A. (2002). An improved parameterization for sulfuric acid–water nucleation rates for tropospheric and stratospheric conditions. *Journal of Geophysical Research: Atmospheres*, *107*(D22), AAC 3-1-AAC 3-10. https://doi.org/10.1029/2002JD002184

Voigt, C., Schumann, U., Graf, K., & Gottschaldt, K.-D. (2013). Impact of rocket exhaust plumes on atmospheric composition and climate — an overview. *Progress in Propulsion Physics*, *4*, 657–670. https://doi.org/10.1051/eucass/201304657

Weisenstein, D., Keith, D., & Dykema, J. (2015). Solar geoengineering using solid aerosol in the stratosphere. *Atmospheric Chemistry and Physics*, *15*, 11835–11859. https://doi.org/10.5194/acp-15-11835-2015

World Meteorological Organization, United States, National Oceanic and Atmospheric Administration, United States, National Aeronautics and Space Administration, United Nations Environment Programme, & European Commission. (2019). *Scientific assessment of ozone depletion: 2018.*

Yu, F., & Turco, R. P. (1997). The role of ions in the formation and evolution of particles in aircraft plumes. *Geophysical Research Letters*, *24*(15), 1927–1930. https://doi.org/10.1029/97GL01822

Yu, P., Davis, S. M., Toon, O. B., Portmann, R. W., Bardeen, C. G., Barnes, J. E., Telg, H., Maloney, C., & Rosenlof, K. H. (2021). Persistent Stratospheric Warming Due to 2019-2020 Australian Wildfire Smoke. *Geophysical Research Letters*, *48*, e2021GL092609. https://doi.org/10.1029/2021GL092609

Appendix D-2

Terms of Reference for Panel of Experts

Terms of Reference

The SCoPEX Advisory Committee has established an expert Panel to support its scientific review of the proposed SCoPEx experiment. The Panel's responsibilities would include:

- supporting the selection of peer reviewers;
- evaluating the reviews;
- providing a summary report to the Advisory Committee about the scientific merit of the experiment based on that evaluation; and
- meeting with the Advisory Committee to communicate the findings of the summary report.

The Panel's summary report to be submitted to the Advisory Committee should include (but is not limited to) the answers to the following questions:

1. Will the proposed study make an important scientific contribution? If so, what is that expected contribution?
    a. How likely is it that the experiment will yield new relevant knowledge that has not already been gained from numerical modeling, laboratory studies, or other approaches?
    b. Can the questions outlined in the proposal be answered in another way? If so, what are the benefits and limitations of this approach versus others?
2. Can the experiment as designed, achieve its objectives by the methodology proposed in the experiment plan?
    a. Is the methodology described sufficiently?
    b. Is there a substantial/reasonable chance/probability that the methodology will enable achieve the stated goal?

The Panel should ensure the quality of reviews (depth and breadth), clearance of potential biases (and the potential need for an additional review(s)), and prepare the summary report.

Please note that the Advisory Committee is conducting other types of reviews, including a societal review, and that the task of the Panel is the review of scientific merits only. You are, however, welcome to include any additional thoughts on the proposed experiment.

To ensure the integrity and impartiality of the entire review process, the Panel members should not have any conflict of interests with the proposed experiment or any members of the core research team (Frank Keutsch, David Keith, and/or John Dykema) as described below:

- Any professional benefit from the project proceeding or not proceeding.
- Current or previous employment or association at Harvard University as a professor, adjunct professor, visiting professor, consulting or advisory arrangement in the past 5 years.
- Previous employment or association within Harvard University in the last 5 years.
- Received an award or grant from Harvard University in the past 5 years.
- Past or present association with any members of the research team as a thesis or dissertation advisor/mentee in the last 5 years.
- Collaboration on a project or on a book, article, report, or paper with any members of the research team in the last 5 years
- Co-editing of a journal, compendium, or conference proceedings with any member of the research team listed in the foundational document within the last 5 years.
- Have past or present grant proposals with any members of the research team in the past 5 years.

Appendix D-3

Round 1 Panel Summary Report

July 15, 2022

The SCoPEX Advisory Committee invited a panel of experts to support a scientific review of the proposed SCoPEx experiment. The Panel (Jim Hurrell, Long Cao and Karen Rosenlof) was responsible for: (1) suggesting external reviewers to the advisory committee; (2) evaluating and summarizing the reviews, with a focus on the scientific merit of the experiment; and (3) communicating the findings to the Advisory Committee via a written summary.

The questions external reviewers were asked to address included:

1. Will the proposed study make an important scientific contribution? If so, what is that expected contribution?
2. Can the experiment, as designed, achieve its objectives by the methodology proposed in the experiment plan?
3. Is there anything else relevant to the scientific merit of this experiment plan that raises concern that has not been covered in the previous questions?

External reviews were obtained from five preeminent atmospheric scientists who specialize in aerosol and chemical processes. Some were instrumentalists and others were modelers. The five experts have also worked on issues related to stratospheric aerosol injection as a possible solar geoengineering technique.

The reviewers had overall agreement on many aspects of the proposed SCoPEx experiment. Overall, the reviewers noted that the proposal has some compelling and important goals that cannot be addressed in laboratory studies or from numerical modeling, and that the Harvard team has in-depth knowledge of key aspects and uncertainties of stratospheric aerosol injection approaches. However, all of the reviewers expressed some significant concerns about the proposed methodology and casted doubt on the successful achievement of the proposed goals. The reviewers also expressed concerns that the proposal falls short in delineating the benefits of SCoPEx. The reviewers made comments beyond the specific questions posed in the terms of reference, and some of those thoughts are captured in the summary below.

Overarching issues:

On common issue noted in the external reviews was that it was very difficult to assess from the proposal if SCoPEx can meet it scientific goals. Much more detailed information on methodology and implementation is needed. One reviewer, for example, desired information on the number of balloon flights that were being proposed, as well as the time period and how long will each flight be. This information is relevant to assessing the risk of complete failure. Reviewers asked whether multiple flights are planned to account for the possibility of failure on an initial attempt and whether adequate time is planned for the case when problems are encountered on the first attempt. Finding the plume is key to success, but there was skepticism whether that was possible, and not enough details were given to assess the likelihood of success. One comment was that to use a lidar to find the plume, the lidar can't be in the plume.

Specifically, to assess whether the experiment is viable, reviewers wanted to know: 1) What is the sequence of detection and sampling? 2) How does the communication work between balloon operations and gondola operations? And 3) Will there be adequate data collected and over a long enough period of time to measure turbulent dissipation and aerosol evolution? It was suggested that a plume model be used to sample a simulated balloon flight in order to determine if useful results would be obtainable.

It was noted by one reviewer that an overriding issue in the proposal is the substantial risk of not achieving the objectives due to the complexity of developing and operating both a new platform and a new payload. Risk and risk mitigation are not discussed in the foundational document at all, and this was noted as a significant shortcoming.

Some reviewers raised important questions about the instrumentation. The optical particle counter was noted to not be well suited for the size of aerosol particles being studied. Two reviewers questioned whether the sun photometer would work to measure scattered light from the injected aerosol plume, and one questioned the detection limits cited for the sun photometer.

Another concern was that there needs to be attention paid to the issue of societal opposition, and the proposal needs to be clear, exact, and explicit about uncertainty. One of the scientific reviewers recommended an educational campaign be conducted in conjunction with SCoPEX.

Specific Goals:

The proposed work is structured as having three scientific goals: (1) Measurements of Turbulence for Small Scale Mixing; (2) Evaluation of Aerosol Microphysics of AM-Sulfate and Alternative SAI Materials; and (3) Evaluation of Process Level Chemical Models of Stratospheric Chemistry of Sulfate and Alternative SAI Materials.

Reviewers noted that the equipment and instruments are described in great detail in the SCoPEx proposal, but that the science questions are not. This is contrary to most proposals that begin with scientific questions, then follow with the proposed methodology to answer them.

The first goal, **measurements of turbulence**, was recognized as important. There is little experimental data related to this problem and some of the reviewers felt that this is where SCoPEX could make the most significant contribution. However, one reviewer stated that, because the balloon has propellers, that it would be unsuitable for making turbulence measurements, and that the injector would also make a gondola platform unsuitable. Another reviewer interpreted this portion of the experiment as useful mainly for understanding and testing coagulation theory for solid $CaCO_3$ particles in the wake of injection behind the balloon package. Although it is suggested that the "measurements which resolve the winds at the dissipation scale will allow numerical models to realistically close the atmospheric kinetic energy budget", details on how this analysis will be performed are missing. That is, this is not a standalone goal to inform SAI in general, but to understand coagulation for the injection specifically proposed in SCoPEx. If this aspect of the experiment could be modified to also address how turbulence would act to disperse gaseous SAI material that forms into particulates, it would be more useful. As currently designed, SCoPEx will likely only sample regions of low turbulence because the balloon will not be launched into regions of high winds. Also, the experiment will not be able to globally characterize turbulence, because of the limited number of measurements that can be obtained from a few balloon flights over a limited region. Understanding turbulent mixing in regards to SAI deployment is critical; however, as described in the SCoPEX document, it is not clear that this portion of the experiment will be successful unless, perhaps, the objective is simply to understand the turbulence that is specific to SCoPEx. Even then, as already noted, other reviewer concerns regard the suitability of the proposed balloon gondola for measuring turbulence. Another reviewer noted that an actual SAI deployment is unlikely to use balloons; thus, the proposal should explain the relevance of the experiment to the more likely aircraft-based approach if SAI was ever implemented.

How the second goal, **evaluation of aerosol microphysics**, will be addressed was not viewed favorably by most of the reviewers. One reviewer noted that this is the highest risk objective of the proposal due, in part, to the lack of details of the approach. Another comment was that we already have a decent understanding of the size, chemistry, and radiative impact of stratospheric aerosol, both in background and volcanically enriched conditions, and that SCoPEx is unlikely to add new information to that understanding. Evaluating how aerosols in a plume evolve is important for SAI, but it wasn't clear from the proposal whether an evolving plume could actually be followed, especially for the time period relevant for SAI injections. This reviewer further noted that even if the gondola could be maneuvered into the plume, it would be difficult (if not impossible) to know where it was within the plume, noting that the lidar cannot measure close to itself. Another reviewer noted that it is very likely that the team can add a known mass of an alternative SAI material; however, exactly how AM-Sulfate will be added was not clear, nor was how an appropriate size distribution would be generated. The results of this portion of the experiment should allow improved understanding of how the size distribution will evolve immediately after injection. However, whether this is useful to inform any realistic future injection effort was questioned.

One reviewer noted that the proposed aerosol particle counter is not well suited for the size of the aerosol particles being studied. Another stated that the primary issue for SAI is the relationship between the injected material and the subsequent evolution of the aerosol size distribution. In situ measurements are the way to measure the evolution, and at present it is not well understood. A much better description of the methodology for this part of the experiment is needed. Another comment was that SCoPEx will not be able to look at the conversion of SO_2 to H_2SO_4, because the time scale for the flights is too short.

The third goal, **evaluation of process level chemical models**, was not viewed favorably. One reviewer noted that the study of alternate materials would be of little interest to the broader atmospheric community. Another had a different opinion, stating that understanding how the addition of SAI materials might alter the distribution of photochemically active constituents that impact ozone chemistry is important, as it will inform whether alternative SAI agents would provide benefits compared to SO_2 or sulfate addition. However, that reviewer noted that it was not clear what new environmental information SCoPEx might reveal that would not be available from appropriate laboratory studies. The availability in the laboratory of much broader analytical capabilities to study both the gas and condensed phase chemistry, moreover, would likely provide a much more robust and less expensive evaluation of these chemical interactions. Another reviewer expressed the opinion that this was a very high-risk aspect of the proposed experiment, largely because it was not clear that the instruments could sample over the length of time needed to sample heterogeneous chemical processes. Another reviewer felt that this was a topic for the future, and sufficient details were not provided to assess how SCoPEX will address this topic.

To summarize comments regarding the primary two questions noted:
Will the proposed study make an important scientific contribution? If so, what is that expected contribution?
Overall the reviewers were not convinced that SCoPEx, as described, will make an important scientific contribution. Exploring turbulent mixing was deemed to be important, although adjustments to the experimental design are likely needed in order to reduce risk and achieve success.

Can the experiment as designed, achieve its objectives by the methodology proposed in the experiment plan?
There were many comments from the reviewers noting that the methodology was not adequately presented to be able to actually answer this question.

Whether the experiment will achieve its goals is questionable. Overall, the reviewers noted that there is compelling science to be addressed by SCoPEx. Specific goals include improving understanding of near-field properties and the surface chemistry of injected aerosols, and turbulent mixing in ambient stratospheric conditions. These are important goals that, if achieved, would represent a contribution to SAI research in the form of improved process understanding, useful SAI modelling studies and could provide information for decisions related to SAI implementation. The foundational document provides justification for in situ measurements to better understand the relevant microphysical and chemical processes.

One reviewer noted that the most likely result will be a well-documented model for the near-field coagulation physics for solid and liquid particles in the wake of the balloon under a subset of well characterized small scale turbulence conditions. Another reviewer had difficulty stating that the experiment is likely to achieve its goals because the methodology was not adequately described. Even considering that, however, that reviewer noted that the experiment has merit: society needs information to make informed decisions about SAI, and this cannot be done in the absence of observed data.

One reviewer noted that the best outcome is that the experiment will yield useful, but flawed, information that will require subsequent experiments to obtain better data. There should also be more attention paid to the societal challenges, in that this experiment may be objected to by some people and organizations. It would make sense to preemptively point out why these injections are not an environmental risk.

In summary, the experiment itself has negligible potential to alter the background stratosphere or harm the atmosphere or the Earth's climate. There may be valuable information gained from this experiment, but the reviewers did not indicate that this will make an important scientific contribution. Also, the reviewers noted that much more information is needed on the details of implementation. However, SCoPEx could be a first step that may provide guidance as to how future related experiments should be conducted.

Appendix D-4

Research Team Response to Round 1 Panel Report

Response to reviewer comments on the scientific contribution of SCoPEx

We thank the reviewers for their thoughtful comments that have helped us assemble a more in-depth and quantitative document. We also appreciate the reviewers' comments that conducting this review was challenging due to the convolution of the question whether the engineering of the SCoPEx platform will enable conducting the required maneuvers with the question of whether the proposed science would be valuable. Our response focuses on addressing reviewer questions about the latter, which also contain aspects that can be viewed as engineering but these are not concerning the engineering platform and its ability to maneuver as required. In addition, platform safety is an important aspect, but this is part of a separate safety review.

The rational for the separation of the evaluation of the platform performance from the science merit is as follows. The review for the platform engineering is quite distinct from that of the scientific merit of the proposed work. For the science merit review at the center of our initial document and this review response document we assume that the platform has been shown to operate successfully, as defined here:

- The platform can float at a desired altitude of ca. 20km.
- The platform can sustain flight operations of 6-12 hours, consistent with the capabilities of a zero-pressure balloon
- The propulsion and navigation functions (horizontal control) are capable of executing the maneuvers described in the science document with reasonable fidelity
- The vertical control system (ascender) performs consistent with manufacturer specifications and ground testing to date, and is reasonably decoupled from the horizontal movement and control of the platform
- The lidar performs with beam steering and sensitivity consistent with manufacturer specifications and optical analysis of the pan/tilt mechanism

A crucial benchmark is that the platform be able to successfully fly through its own wake detecting the wake with the LITOS turbulence instrument. This benchmark allows an end-to-end test of winch, electronics, communication, and navigation. Achieving this benchmark is a goal for the initial flights.

The research team is acutely aware that the SCoPEx platform may not perform successfully. Most of our effort is focused on engineering tests including a set of payload hang tests from a tall crane and thermal-vacuum tests to reduce these risks.

A review of the engineering risks of the SCoPEx platform would necessarily require a very different document describing the system design, component specifications, and test results to date. These materials stand in contrast to the materials (of the science proposal) essential to presenting the science questions, testable hypotheses, and quantitative analysis of the instrumentation based on atmospheric modeling of the stratosphere and solid particles.

We therefore request that the AC and reviewers agree that the science review focus on the contingent questions: can the team achieve useful science assuming the payload engineering works. Contingency is appropriate because the experiment team will not proceed to science flights until the engineering aspects are validated.

In this document we respond to reviewer comments on the scientific merit of SCoPEx. We focus on the specific scientific contributions of SCoPEx and how our methodology supports these with improved quantitative descriptions. We focus on the summary of the panel as well as specific referee criticisms we found most salient. We are not addressing comments on science goal 3 in the original proposal, i.e., the chemical evolution, as this is a more distant goal and we believe that given the likely momentum in geoengineering research different approaches could exist by the time these goals would be pursued with the SCoPEx platform. If this is not the case, we will revisit the comments on these science goals.

Research into the physical basis of stratospheric aerosol injection as a form of climate intervention is steadily increasing. Despite this increase, experimental research is lagging, arguably with deleterious consequences for the reliability of scientific knowledge available to policymakers(Keith et al., 2020). There is a broad class of small-scale processes that mediate the efficacy and risks of SAI that are poorly constrained by observations. The SCoPEx gondola equipped with the proposed instrument suite is intended to refine the current state of knowledge for the following set of science questions that pertain to a subset of these small-scales processes. Observational constraints on these processes would further the objective of improving models of aerosol dynamics in a stratospheric plume for SAI

Question 1 – *What is the horizontal variability of stratospheric turbulence?*

The mechanisms of production and dissipation for stratospheric turbulence and its amplitude and spatiotemporal distribution exert a fundamental control on stratospheric aerosol mixing and microphysics. Measurements of tracer species have been used to infer turbulent parameters for decades, however, comprehensive observations to directly measure stratospheric turbulence remain uncommon. Because stratospheric turbulence is a transient phenomenon, with a very inhomogeneous spatial distribution, the spatial and temporal variability of stratospheric turbulence is not well quantified. Direct measurement of stratospheric turbulence, co-located with a well-characterized aerosol plume, therefore provides a unique opportunity to quantify coupling between aerosol microphysics and fluid motions at small scales. For these reasons, SCoPEx wishes to observe the spatial variability of stratospheric turbulence, while at the same time measuring the size distribution of a coagulating solid aerosol at high space and time resolution.

Question 2- *How well does an advection-coagulation model driven by winds from a Large Eddy Simulation (LES) predict the temporal evolution and spatial distribution of a plume of sub-micron solid aerosol?*

Computationally intensive CFD models are required to explicitly resolve the atmospheric kinetic energy spectrum at the scale of the plume produced by the SCoPEx experimental approach. Finding a compromise between detail and computational affordability leads us to a class of CFD models called Large Eddy Simulations (LES). LES is a form of CFD that operates under the assumption that one is interested in the larger scales of one's flow, that these structures contain the bulk of the energy in the flow and that the scales much smaller than these have a smaller impact and are more amendable to modeling with a lower fidelity. In the case of LES, "larger" structures refer to scales on the order of meter to sub-meter, but not down to centimeter scales (the domain of costly Direct Numerical Simulation, or DNS).

Question 3 – *Does the coagulation of solid aerosol particles vary systematically as a function of stratospheric turbulence?*

Our ability to accurately model an injected aerosol size distribution and its evolution in time and space depends on the representation of sub-grid scale (SGS) processes (Keith et al., 2020; Sun et al., 2022). These SGS processes occur on timescales of seconds to minutes, and on length scales of sub-meter to tens of kilometers. Constraining SGS processes is essential for reducing the uncertainty in aerosol properties and plume evolution at this scale. The high-resolution measurement datasets that will be obtained by SCoPEx coupled with the fine scale computational fluid dynamics (CFD) modeling proposed here are ultimately intended to provide critical process level information that will be integrated into and/or improve existing parameterizations used in GCMs. The relevant processes are as follows. The interaction of aircraft wakes, into which aerosol or aerosol precursors are injected in SAI scenario simulations, with the unperturbed atmosphere is controlled by SGS processes. The most uncertain SGS processes that influence aerosol microphysics (via coagulation) and plume dilution are those of turbulent energy production and dissipation.

Detailed Experimental Operations of SCoPEx

Consistent with the restatement and refinement of the SCoPEx science objectives and scientific questions in the previous section, the experimental operations of SCoPEx will focus on calcite solid aerosol. Because calcite is found in vanishing small quantities in the stratosphere, it is an effective passive tracer for plume evolution and mixing with ambient stratospheric air. Additionally, because there is no significant evaporation of the aerosol material, and because it is initially introduced in a monodisperse form, it is a simpler system for the study of coagulation. The SCoPEx team has experience in nebulizing 0.5 µm calcite aerosols in the laboratory, and has studied the chemistry of these particles in the laboratory. While these laboratory activities provide a calcite experience base, the primary drivers of selection of calcite as the aerosol system to study are the simpler microphysics and its extreme scarcity in the unperturbed stratosphere.

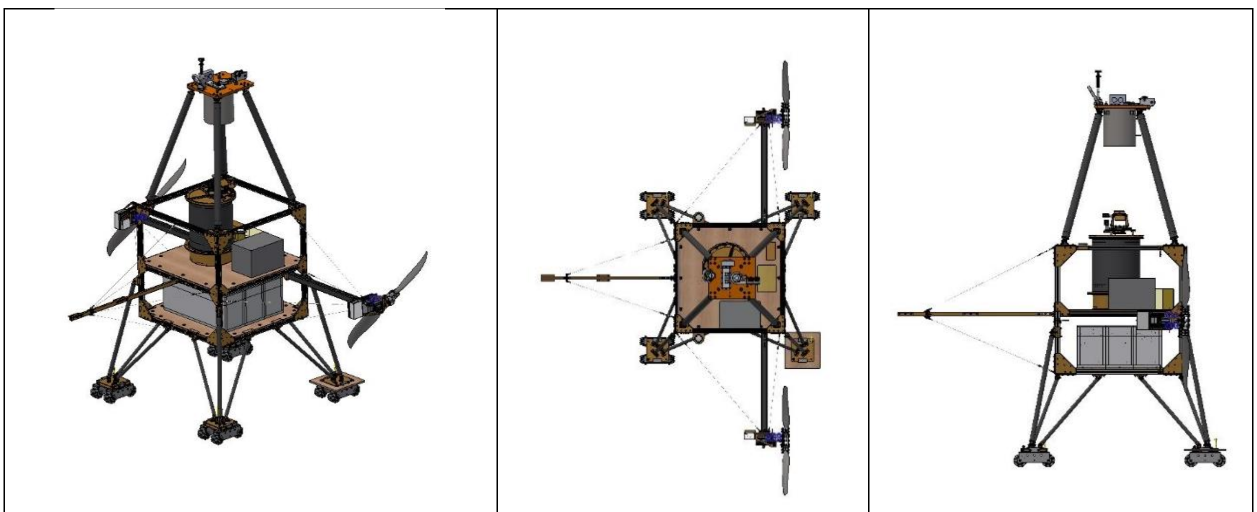


Figure 1: SCoPEx gondola includes boom to place the LITOS turbulence measurement external to the influence of the gondola and propeller aerodynamic wakes. Views are near isometric (left), top (middle), side (right). This model represents the as-built equipment for the crane test described in the appendix.

Experimental time-scale and altitude:

The SCoPEx payload is designed to provide instrument mechanical, thermal, and power accommodations, to implement high-bandwidth real-time communications with the ground support team, and to locate instruments to facilitate their scientific objectives. The SCoPEx equipment gondola will be suspended from a zero-pressure balloon, which will allow experiment operations of at least 6-8 hours. This 6-8 hour duration excludes the approximately 1.5 hours to ascend to the 65,000 ft nominally float altitude. The predominant horizontal motion of SCoPEx is to drift with the same prevailing winds that advect the balloon downstream. This means that SCoPEx has the potential to continue measuring the evolution of the same plume for 8 hours, if the operational conditions (winds, local thermal environment, aviation/landing considerations) permit.

Vertical and horizontal scale and control:

The length of the fixed tether that connects the mechanical interface at top of the SCoPEx gondola to the balloon is over 100 m in length with an ascender beyond that to adjust the vertical position. This length was selected using CFD simulations of the balloon wake to determine a conservative distance that created a clear separation between the balloon aerodynamic wake and the aerodynamic wake due to the propellers. The control of the movement of the SCoPEx payload relative to the prevailing local winds is facilitated in 3 dimensions by an ascender and the two propellers. The ascender allows vertical control of the gondola position relative to the balloon by winching the gondola up or down relative to the fixed interface at the end of the balloon tether. Differential thrust can be applied to the propellers to move the gondola left or right relative to the prevailing wind, providing horizontal control. Additional coarse vertical control can be achieved by dropping ballast (rising) or actuating the balloon's valve to allow escape of fill gas (sinking).

Turbulence measurement placement:

Because of the importance of accurate turbulence measurement to the scientific objectives, the LITOS anemometer instrument is located on a boom upstream on the gondola (on the opposite side of the propellers). This upstream position isolates LITOS from the propeller wakes that form downstream, with an underlying physical principle analogous to the upstream location of turbulence probes fixed wing aircraft by using boom mounts. During the experimental phase at the balloon's float altitude, LITOS is also separated from the balloon wake by the 100 m tether and the additional distance provided by the position of ascender.

Locating plume and sampling maneuvers:

SCoPEx is capable of two maneuvers that allow sampling of its aerodynamic wake (created by the propellers). Recent tests of the payload from a long-reach construction crane indicate that SCoPEx should be able to rotate itself in place when it is static relative to prevailing winds (eg when it is “stopped”). This capability allows one sampling option, after a plume injection operation (Fig. 2), to turn off the propellers, drift to a stop, and then turn 180° and propel itself back into the plume along the plume's axis. The second sampling maneuver is to perform a turn while moving forward, which takes it away from the plume. This kind of maneuver can be used to move to a vantage point of at least 100 m from the plume so that the lidar can be used to scan the field of regard and identify the plume endpoints. This maneuver also facilitates perpendicular transects from the plume spaced along the plume's axis (Fig. 3).

The lidar includes a mirror with pan and tilt capability to allow it to scan a field of regard that is ±115° of pan motion and ±15° of tilt motion. The mirror can also be flipped 180° to permit viewing ±115° in the direction of the propellers. Although there are obscurations at 45° by structural pillars of the payload, when the lidar is facing the plume (eg at one of the starred locations in Fig. 3) from a distance of 150 m or greater, it is able to scan for the location and extent of the plume.

The combination of vertical and horizontal motion of the gondola allows it to be placed in locations to obtain optical extinction measurements of the plume. In a publication detailing an advection-coagulation model built to simulate SCoPEx (Golja et al., 2021), we analyzed the brightness of scattered light from the plume as viewed by specific multi-band photometer (Murphy et al., 2016) in an almucantar scan. The vertical control capability of SCoPEx should allow a maneuver to conduct almucantar scans located such that the plume is both within and excluded from the scanning photometer's field of regard. However, the power spectrum of gondola oscillations during this kind of vertical maneuvering is not yet known, and could prevent measurement of the plume's optical extinction with low uncertainty. After an engineering flight to quantify the gondola stability, appropriate data will be available to determine whether a scanning photometer or a nephelometer with a fixed field of view is a better choice. Ultimately, the optical extinction of the plume is of interest for understanding the radiative impacts of the particles studied. However, this measurement is not necessary to achieve the primary goals of testing hypotheses about turbulence in the propeller aerodynamic wake and about coagulation of solid calcite monomer particles in a wake with measured turbulence.

Project timeline:

The project schedule depends on the performance of the SCoPEx gondola to perform the maneuvers described in this document, and the homogeneity and repeatability of stratospheric conditions encountered when the experiment is performed. Because of the uncertainty about these engineering and environmental issues, any project schedule and timeline is notional. The current project schedule is to perform an engineering flight to validate the platform systems and quantify the horizontal and vertical control performance. Additional flight time and engineering effort may be needed to optimize these platform systems. Once the platform has been validated, flights can begin that focus on achieving the scientific objectives. The program will plan for the possibility that multiple flights are required to achieve the science objectives. Preliminary plume data, including turbulence measurement and particle size distributions, will allow for much more quantitative assessment of flight time required to answer the SCoPEx **science questions with high confidence.**

SCoPEx flight operational phases:

The first two SCoPEx experimental operations will consist of 3 phases, which may be repeated multiple times per flight. The first phase is the aerosol injection phase, in which the gondola travels at constant potential temperature in a consistent horizontal direction at a fixed speed. The second phase is the sampling phase. During the sampling phase, the gondola maintains the same local altitude as during the injection phase. The gondola will make repeated transects of the propeller wake connected by roughly U-shaped turns with an along-plume spacing of approximately 100 m. These maneuvers will be within a horizontal plane that is parallel to the Earth's surface. The final phase will be a vertical maneuver designed to allow for optical measurements made from above, at the level of, and below the propeller aerodynamic wake.

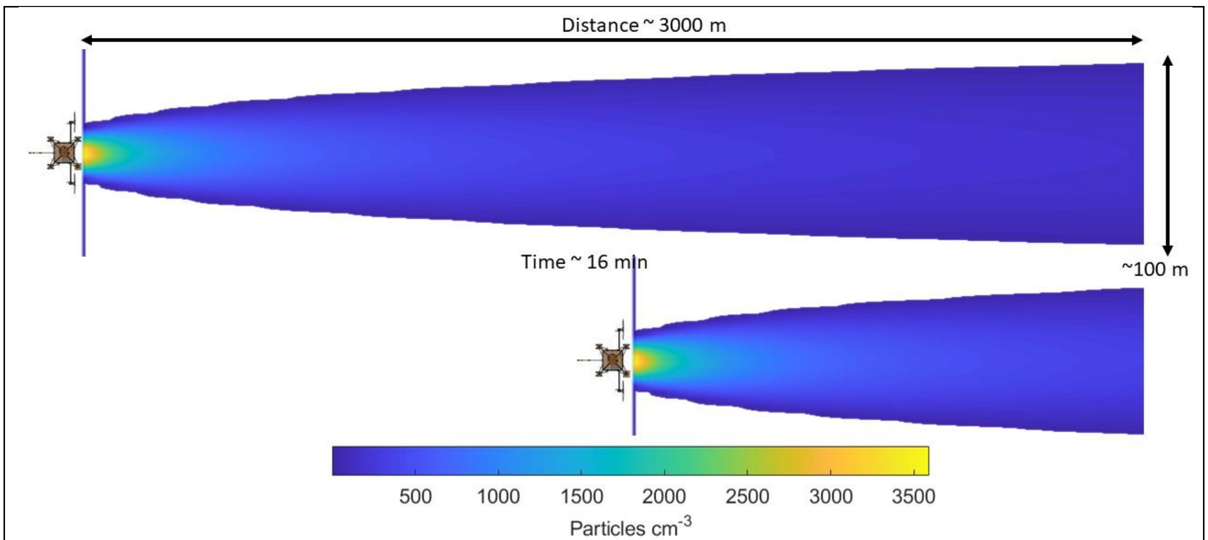


Figure 2: Plume injection (Phase 1) as viewed from above. calcite plume after approximately 8 m of travel and injection (lower plot) and after 16 min of travel and injection (upper plot). The contour colorbar is proportional the number density of monomer calcite particles and is produced by the advection-coagulation model of Golja et al. (2021).

Phase 1 (Fig. 2) will occur with the gondola being propelled at 3 m s^{-1}. This speed is chosen to minimize the flight time that is allocated to creating a plume. This approach leaves more time to allow the plume to evolve, for calcite particles to coagulate, and to make contrasting measurements of turbulence inside and outside the plume. During this phase, measurements will be made of the atmospheric kinetic energy spectrum (LITOS), the unperturbed stratospheric aerosol size distribution (POPS), and, by using the pan/tilt mechanism, the normalized relative backscatter (micropulse lidar).

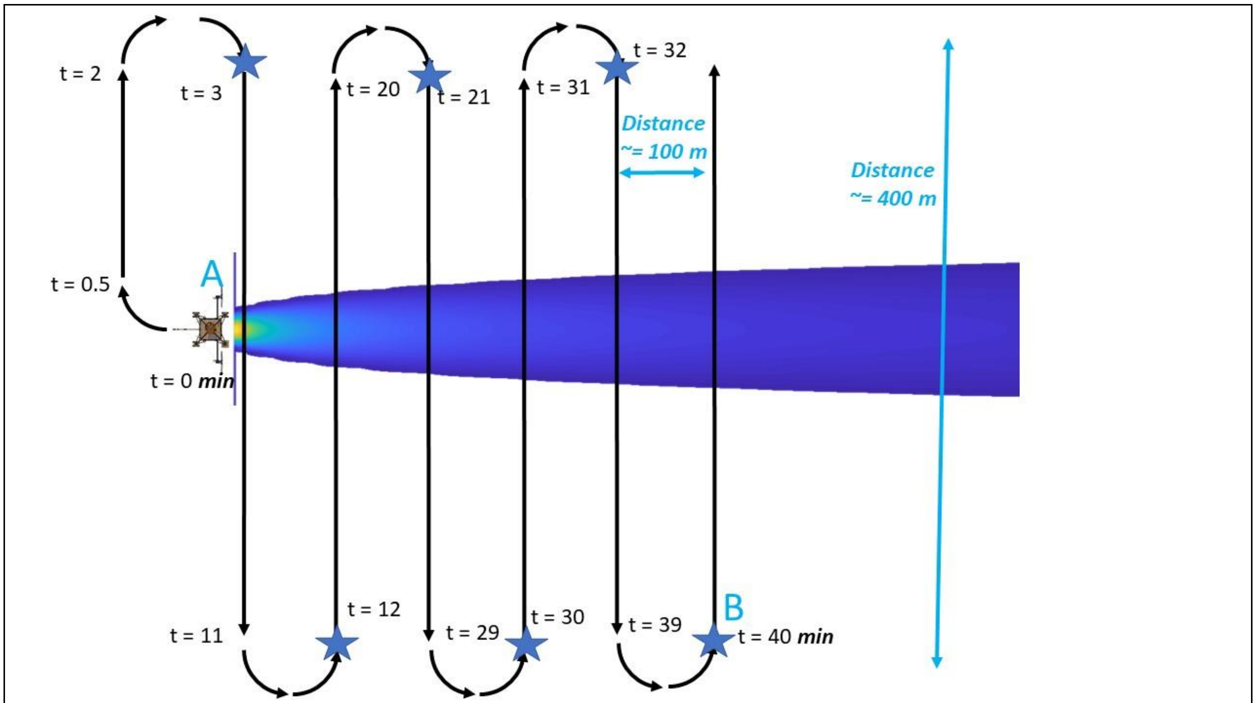


Figure 3: Plume sampling maneuvers (Phase 2), shown from an overhead view (gondola only). The repositioning capability of the gondola is used to turn the gondola and allow it to move relative to the plume, while changing its orientation. The approximate estimated time to perform each maneuver are shown in minutes, beginning from t=0 minute, which is when plume injection is terminated. Note these times will vary significantly in a real flight (during which we anticipate the plume will also be missed on some maneuevers). The plume transects will be spaced at about 100 m along the plume length (axial direction). The transects will begin and end at least 150 m from the edge of the plume to allow valid lidar measurements of plume backscatter. Lidar vantage points A and B illustrate different viewing geometries (see next section). For this notional flight, 6 transects will be obtained within 48 minutes after the plume is injected, and will sample 500-600 m of plume axial distance.

Phase 2 will occur with the balloon being propelled at speeds selected to optimize in-plume measurement time traded against plume integrity. The rationale for utilizing the platform's maximum speed is to carry out the desired number of plume transects in the shortest time possible to maintain the spatial coherence of the plume against distortion by wind shear and dissipation due to mixing with ambient air. However reducing the speed while in the plume increases measurement duration, allowing additional integration time to improve SNR. The transect maneuver will include travel away from the plume for a distance of 150 m, followed by a turn to reorient the gondola to permit travel towards the plume. At the end of the turn, before initiation of the travel towards the plume (to be followed by the transect), propulsion will be slowed to zero to permit a coarse scan of the lidar field of regard. This scan will identify the end points of the plume, which will be used to locate the gondola's current location relative to the plume. A fine scan of the lidar through a reduced field of regard will be conducted to identify the balloon centerline. The gondola will then begin moving towards the plume, and continue for a distance of approximately 400 m, consisting of 150 m to reach the plume, 100 m to transect the plume, and 150 m to travel past the plume before initiating the next turn/transect maneuver (Fig. 3). During this phase, measurements will be made of the atmospheric kinetic energy spectrum inside and outside the plume (LITOS), of the ambient aerosol size distribution (outside the plume) and number density for calcite fractal aggregates (inside the plume), and of the NRB (lidar).

Lidar operation and performance for plume location:

One major question about SCoPEx's experimental operations is how SCoPEx will be able to locate the plume. The micropulse lidar, integrated with a pan/tilt scan mirror, will be a major capability for achieving this task. Here we consider two lidar viewing cases, one for the lidar viewing the plume behind it (location A in Fig. 3) and one for the lidar viewing the plume from a distance of 1000 m (similar to location B in Fig. 3, but further displaced down the plume axis and more distant from the plume).

In both cases, an important feature of the lidar return signal will be the invalid signal corresponding to the first 150 m of the backscatter profile. The nominal blind zone for an off-the-shelf MiniMPL lidar is 100 m, due to a combination of after pulse from the laser source and scatter from the optical surfaces. As the SCoPEx lidar has two additional optical elements (the pan/tilt mirror and the window of its pressure vessel), we increase the length of this blind zone to 150 m. The exact length of the blind zone of the lidar will be measured by ground testing and the distances corresponding to the different legs in phase 2 will be adjusted accordingly.

Species	**Backscatter (m^{-1} sr^{-1})**
Calcite (plume axial view, location A)	3.9×10^{-5}
Calcite (plume tangent view, location B)	1.2×10^{-5}
Molecular species	1.2×10^{-7}
Background aerosol	6.3×10^{-8}

Table 1: lidar backscatter coefficients for species contributing to horizontal lidar measurements

The key values of the lidar backscatter coefficients, in units of m^{-1} sr^{-1}, for these two viewing scenarios are shown in Table 1. The main sources of unwanted backscatter in the lidar profiles are molecular backscatter (from gas phase species) and backscatter from ambient aerosol. Both of these sources of backscatter cause an offset of the profile due to the plume backscatter. The dominant noise mechanism for the lidar is photon shot noise (detector noise is suppressed by the photon counting detector), so that the SNR for measurement of the plume backscatter is degraded by the molecular and ambient aerosol shot noise. The relative magnitudes of these shot noise terms can be calculated from Table 1, since the photoelectron return for each backscatter species as a function of range (distance between the lidar and the backscattering species) will be proportional the product of beta times the inverse of the range squared.

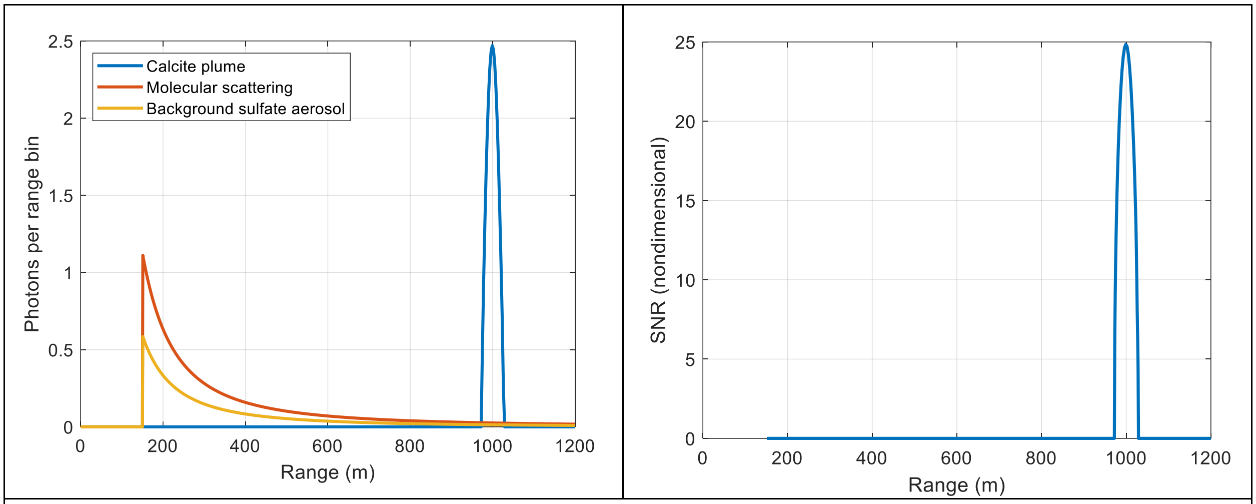


Figure 4: Photons per range bin for plume viewed from location B, 1000 m from plume center for a single pulse (left) and SNR for an integration of 250 pulses (right).

The backscatter for the background aerosols is computed assuming pure sulfuric acid/water aerosol with size distributions at 20 km from the balloon-borne Wyoming Optical Particle Counter dataset averaged over the period of moderate volcanic activity from 2004-2014. The molecular backscatter assumes a pressure of 55 hPa and temperature of 210 K with cross-section from a first-principles approach (Bodhaine et al., 1999). Calcite is based on the Golja *et al*. 2021 model runs with tabulated values for calcite refractive index and Mie theory.

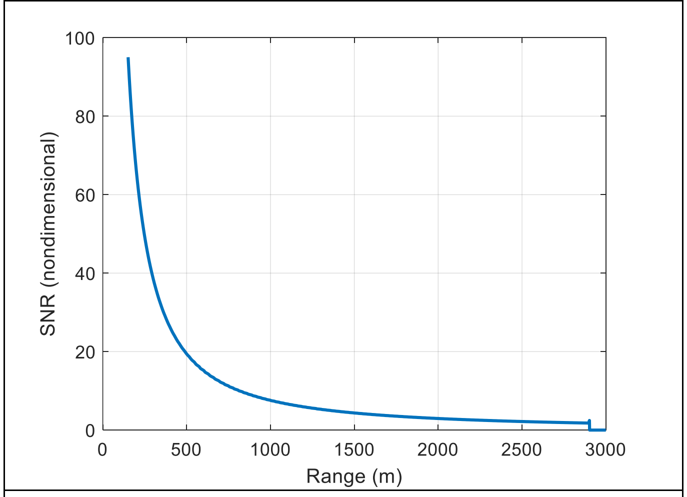


Figure 5: SNR of lidar measurement along axis of plume from location A, during injection, for an integration of 25 pulses.

The SNR for the two viewing geometries is calculated from the standard lidar equation. The key parameters for the lidar are a pulse energy of 4 μJ, a pulse repetition rate of 2500 Hz, an aperture of 8 cm diameter, a wavelength of 532 nm, a field of view of 532 nm, and a total optical efficiency of 80%. The SNR for the tangential geometry (location B) for a single pulse is shown in Fig. 4 (left panel). This single pulse acquisition corresponds to an integration time of 0.4 ms. An SNR in excess of 1 is achieved in the case, where the gondola is displaced 1000 m from the plume, and the plume has expanded for a time of about 17 minutes from since its injection. The right panel of Fig. 4 shows the SNR when the acquisition time is increased to 100 ms. In this case the peak SNR is 25. This SNR with a 100 ms acquisition time provides a good compromise between maximizing SNR and minimizing the time to scan the lidar field of regard, quantifying the capability of SCoPEx to scan and locate the plume with high confidence from a significant distance.

The SNR for the axial viewing geometry is shown in Fig. 5. The proximity to the relatively fresh plume means that high particle concentrations are encountered, even when factoring in the blind zone of the first 150 m of the lidar profile. These along-axis lidar measurements during the injection of the plume will allow continuous monitoring of the plume with respect to shear and unanticipated events that disrupt the plume morphology. Detection of distortions in the plume can be used to modify the experimental plan, cutting short the plume injection phase to sample more quickly and provide opportunities to understand causes of complications to the experimental operations.

Data Analysis

<u>LITOS turbulence measurement:</u>

The LITOS team estimates that the uncertainty in the determination of the turbulent dissipation rate ε is ±30-50%. This uncertainty is driven primarily by fitting errors encountered when fitting the kinetic energy spectrum to estimate the dissipation length l_0. The observation time required for LITOS to obtain a full kinetic energy spectrum is about 4 s, so when SCoPEx is traveling at 0.25 m s^{-1}, an estimate of ε will be obtained at 1 m horizontal resolution. The LITOS probe faces forward and thus is not obstructed by the instrument gondola and based on communication with the LITOS team for the short duration of the measurement the propellers are turned off. The LITOS turbulence measurements alone are a valuable contribution to the existing datasets on stratospheric turbulence; previous data of this nature almost exclusively consists of vertical profiles that can be impacted by balloon wakes; the SCoPEx measurements will provide extended data at a given pressure level far from the balloon wake and outside of the influence of the gondola.

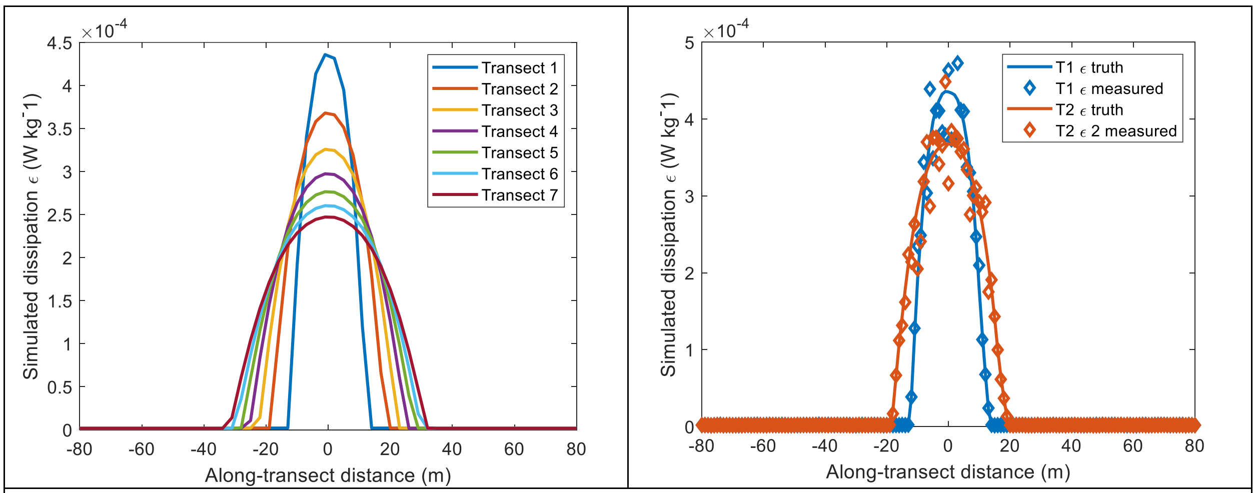


Figure 6: Expected profile of turbulent kinetic dissipation based on CFD inputs to Golja *et al*. 2021 for 7 transects spaced at 100 m (left). A Monte Carlo simulation using a conservative estimate of LITOS measurement noise for the first two transects is also shown (right).

Given the expected timing and duration of the plume transects based on the operational plan in the previous section, the plume width (eg the horizontal spatial extent of the plume along the direction of the transect) should range from about 40 m to 100 m. We therefore anticipate approximately 40-100 measurements of ε per transect, with fewer measurements for earlier transects where the plume is less expanded (Fig. 46 left). Given an uncertainty of 30-50% and assuming uncorrelated measurements, the mean value of the dissipation will be measured with an uncertainty of 3-8%. This is less to the decrease in the peak dissipation between subsequent transects, and given the consistent profile shape from CFD simulations, the measurement uncertainty is sufficiently low to provide an observational test of the CFD (Fig. 6, right).

<u>POPS particle size distribution measurement:</u>

The operating principle of POPS is that particles are injected across a transverse laser beam, and a collection mirror focuses a significant fraction of scattered photons onto an avalanche photodiode, creating an electronic pulse. The pulse height is proportional to the number of photons scattered to the APD. This photon number is set by the particle scattering cross-section integrated over the angles spanned by the collection mirror. For the polarized laser used by POPS, it is necessary to account for the proportion of photons scattered parallel and perpendicular to the scattering plane (defined by the direction of the laser beam and the vector along the direction of the scattered photon towards the collection mirror). When this optical geometry-specific scattering cross section is plotted versus radius, there is non-monotonic behavior due to Mie optical resonances. For the specific case of SCoPEx, however, where the objective is to count fractal aggregates consisting of integral numbers of monomers, the geometry-specific scattering cross sections differ significantly (Fig. 7). Discriminating trimers from tetramers may however require a longer wavelength laser to replace the stock 405 nm laser. The differences in the cross-sections mean that the number density can be counted independently for the calcite fractal aggregates, allowing a test of whether the Brownian coagulation kernel is consistent with the SCoPEx observations of coagulating calcite monomers in the propeller aerodynamic wake.

The counting precision of the POPS instrument can be evaluated by noting that for well-controlled flow conditions the particle arrival time will follow Poisson statistics. The number of particles anticipated during a sampling period may be determined from the POPS linear flow rate in L min^{-1}, the linear speed of the gondola along the transect, and the number density of the particles as a function of aggregate number (Fig.8 left panel). The particle number density is calculated from the advection-coagulation model of Golja *et al*. 2021, driven by the CFD simulation with turbulent dissipation shown in Fig. 6. Note that the microphysics scheme used measure aggregates composed of numbers of aggregates that are powers of 2, up to 256 (eg 1,2,4,8,16,32,64,128,256). For a gondola speed and integration time consistent with the 4 m sampling shown in Fig. 6, the counting precision exceeds 1 in the core of the plume for monomers, dimers, and tetramers (Fig. 8, right panel).

Scientific Analysis

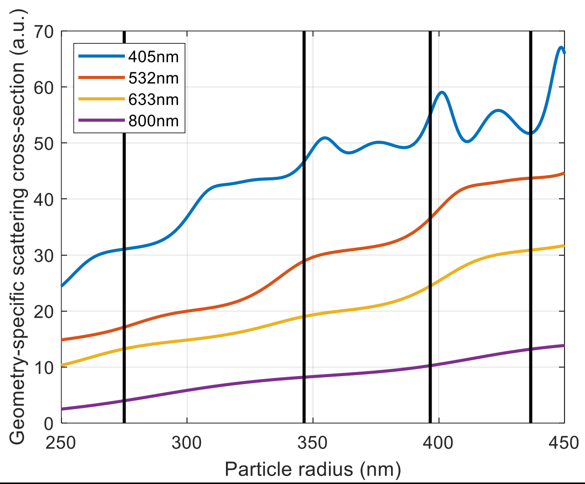


Figure 7: Calibration curve for POPS for calcite spheres as a function of laser wavelength. The radii of the equivalent spheres for fractal aggregates composed of N=1…4 aggregates are shown by the vertical black lines.

Provided the SCoPEx platform can perform the experimental procedures described above, and that the turbulence and size distribution measurements meet their specifications, we expect to have scientifically useful results on turbulence and coagulation. These measurements stand on their own as contributions to ongoing research in these atmospheric science topics. However, these results need to be incorporated with numerical models in order to effectively advance the current state of knowledge about stratospheric aerosol injection as a climate intervention.

Turbulence science analysis:

The first task we have is utilizing our measurements of background stratospheric turbulence and in-plume turbulence to validate and improve simulations of stratospheric winds on spatial scales of 10 km down to the turbulence scale. We will begin by repeating the Reynold-Averaged Navier-Stokes (RANS) simulation by ANSYS Fluent of Golja et al. (2021) with updated background flow conditions (turbulent viscosity) and a velocity flow field (linear speed of the gondola during plume injection) that is true the experimental conditions achieved with SCoPEx. The RANS simulation does compute turbulent viscosity using a k-epsilon model, which can be converted into turbulent dissipation for direct comparison with the LITOS measurements. This constitutes a hypothesis test: can a RANS simulation with realistic background conditions predict the measured turbulence in an aerodynamic wake?

In improving on this RANS simulation, finding a compromise between detail and computational affordability leads us to a class of CFD models called Large Eddy Simulations (LES). LES have been used to study single aircraft contrails, bridging the scales between jet expansion, which can be described analytically, and evolution in the atmosphere after disturbances from aircraft have dissipated (Lewellen & Lewellen, 2001; Paoli et al., 2017; Paugam et al., 2010; Unterstrasser et al., 2014). This existing body of research provides an ideal foundation for analysis of SCoPEx data, and aerosol plume evolution, because contrails are controlled by the same physical processes: turbulence, radiation, and microphysics. Furthermore, LES results are recognized as being well-suited to the development of parameterizations for global models.

After validation of the LES has been performed, we will be ready to perform a high-fidelity simulation of the SCoPEx experiment that is suitable for comparison with LITOS wind measurements. The results of this simulation will be analyzed to determine whether the range over which the turbulence structures generated by the SCoPEx platform are isotropic at small scales (as in the contrail results of Paoli et al. 2017) or anisotropic (as in the contrail results of Unterstrasser et al. 2017). Confirming the anisotropy or isotropy of the turbulence will provide an important insight on the interpretation of the LITOS data, which is inherently insensitive to wind direction. Spectral models of turbulence are an area of active research (Strelnikov et al., 2022), and the LITOS measurements will provide an important case study for this topic.

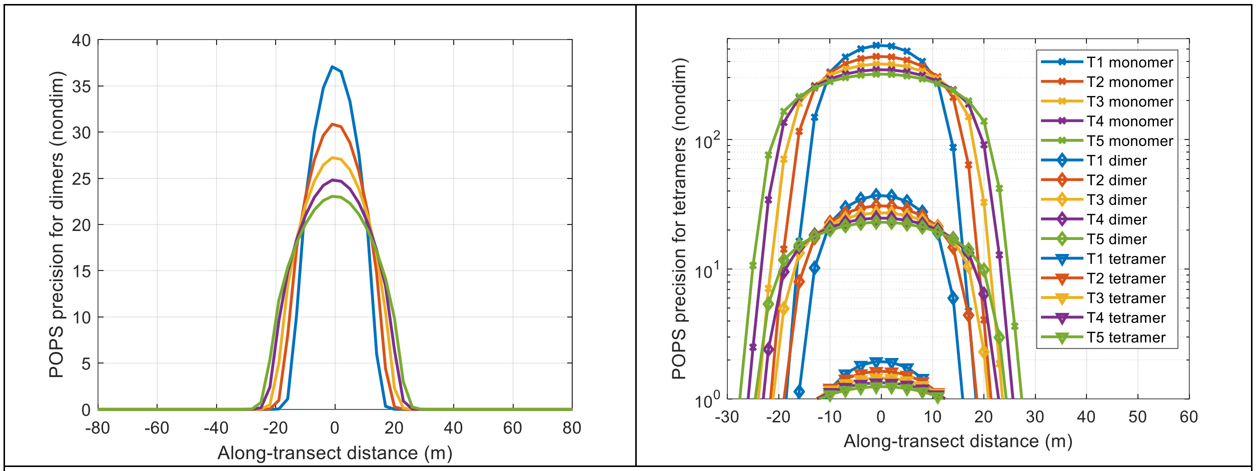


Figure 8: Precision of POPS measurement of number of particles N per 4 m transect ($\Delta N/N$) for a flow rate of 0.1 L min^{-1}. The data for dimers are shown on left panel, the right panel superimposes monomers, dimers, and tetramers on a log scale. The SNR exceeds 1 in the core of the plume for all transects and aggregates plotted.

Understanding the mechanisms of stratospheric turbulence production is essential to understanding the spatial inhomogeneity and effective rate of mixing on spatial scales of 10-500 m (Schneider et al., 2017). One of the most important and yet uncertain sources of stratospheric turbulence is gravity wave breaking. The horizontal trajectory of SCoPEx leads to a novel measurement approach that provides a unique opportunity to test hypotheses about the spatial distribution of stratospheric turbulence, and its relationship to breaking gravity waves. If SCoPEx does observe an unexpected patch of turbulence during its operation, it would provide a unique case study for to test for potential gravity wave breaking or filtering. LITOS PI Michael Gerding has experience in this type of analysis. High-resolution atmospheric models such as the Weather Research and Forecasting (WRF) resolve individual gravity waves when run at high enough resolution. Gravity waves simulated by WRF can be compared with ray tracing models to increase confidence in the interpretation of model-simulated gravity waves.

Science analysis of plume evolution and aerosol microphysics:

Moving on from turbulence considered in isolation, we wish to consider the coagulation of 275 nm radius calcite monomers in the presence of known turbulence. The advection-coagulation model used in Golja et al. 2021 provides a simulation of solid and gas phase aerosol in flow field simulated by CFD. The solid phase microphysics includes coagulation but excludes sedimentation. The exclusion of sedimentation is relatively less consequential for this experimental phase of SCoPEx, since only the monomers, dimers, trimers, and tetramers are relevant to testing the coagulation kernel for the science questions under consideration. The tetramers have four times the mass of the monomers, which is significantly less than the 37x larger mass of a 1 µm particle relative to a 0.3 µm particle. Moreover, given the fractal nature of calcite aggregates, the aerodynamic diameter of the tetramer is larger than the aerodynamic diameter of the equivalent compact sphere. We will revisit the inclusion of the sedimentation in the microphysical scheme for future experimental phases of SCoPEx. The coagulation kernel in this microphysics scheme considers only Brownian motion without gravitational, convective, or van der Waals corrections. The first improvement of this advection-coagulation model will be obtained by driving the advection by output of the LES simulation described above, which has been tuned to best represent the observed turbulence during SCoPEx experimental operations.

The next improvement that may be necessary to improve agreement between the measured size distributions (that is, the measured populations of fractal aggregates by core number as a function of time and position) is to include the effects of turbulence on the coagulation kernel. Turbulence modifies coagulation relative to the diffusive Brownian case because the effects of turbulent friction on particle motions need to be included. There are size-dependent effects for turbulent friction, meaning that it is possible that the monomers, dimers, trimers, and tetramers (etc.) may experience different frictional forces, ultimately modifying their rates of coagulation. Turbulent coagulations kernels due to shear and inertial effects have been described (Saffman & Turner, 1956), updated for larger particles (Kruis & Kusters, 1997), and evaluated in numerical (Reade and Collins 2000) and experimental (Okuyama et al., 1978; Reade & Collins, 2000) studies. Ultimately there are multiple factors about numerical implementation and choice of parameterizations that are inherent both in LES and coagulation simulations. Depending on the disparity between the SCoPEx measurements and model results, different modeling approaches and collaboration will be appropriate to improve the state of knowledge of turbulence and coagulation relevant to SAI.

Summary

The simulation of a hypothetical SCoPEx plume composed of aggregating 275 nm radius calcite monomers has provided a basis to quantitatively assess the instruments intended to investigate specific science questions about in-plume turbulence and coagulation. This analysis has provided insights on the suitability of the POPS optical particle counter and the scanning sun photometer for performing the necessary measurements for the SCoPEx objectives. For POPS, the stock 405 nm laser is judged to be sufficient to distinguish calcite monomers from dimers, and dimers from trimers and tetramers. Distinguishing the trimers and tetramers may require a longer wavelength laser (for example 532 nm), which can be accommodated in the size, weight, and power envelope available for POPS on SCoPEx. Given the capability of the lidar for plume detection, the optical extinction measurement from the scanning sun photometer is not necessary to answer the primary coagulation and turbulence science questions. More detailed knowledge about the platform motion will allow a detailed investigation of the potential of different instruments for plume optical extinction measurements. For example, a simple fixed view nephelometer that is translated relative to the plume by the platform's 3-dimentional translation capabilities could be a better choice.

The combination of turbulence measurements from LITOS and size distributions from POPS can be analyzed with existing data tools; that is, an advection-coagulation model driven with wind fields from a RANS CFD simulation, and incorporating a classic Brownian coagulation kernel. The analytical results from this existing model will inform development of higher-fidelity numerical simulations. This next level of complexity in simulations will likely begin with a simulation of the plume with LES CFD, implemented to maximize agreement with LITOS measurements. This LES CFD output can be coupled to the Brownian kernel, and the resulting size distributions compared with POPS measurements. Disagreement between this numerical simulation and the POPS measurements can be studied by modifying the coagulation kernel to include turbulent effects on coagulation. Taken together, these findings will provide constraints for developing improved in-plume coagulation parameterizations for larger scale models.

References

Bodhaine, B. A., Wood, N. B., Dutton, E. G., & Slusser, J. R. (1999). On Rayleigh optical depth calculations. *Journal of Atmospheric and Oceanic Technology, 16*(11), 1854-1861.

Golja, C., Chew, L., Dykema, J., & Keith, D. (2021). Aerosol dynamics in the near field of the SCoPEx stratospheric balloon experiment. *Journal of Geophysical Research: Atmospheres, 126*(4), e2020JD033438.

Keith, D., Keutsch, F., & Floerchinger, C. (2020). Empirical methods to reduce uncertainty about solar geoengineering. Retrieved from https://keith.seas.harvard.edu/files/tkg/files/keith_keutsch_floerchinger_emperical_srm_for_nas.pdf

Kruis, F., & Kusters, K. (1997). The collision rate of particles in turbulent flow. *Chemical Engineering Communications, 158*(1), 201-230.

Lewellen, D., & Lewellen, W. (2001). The effects of aircraft wake dynamics on contrail development. *Journal of the atmospheric sciences, 58*(4), 390-406.

Murphy, D., Telg, H., Eck, T., Rodriguez, J., Stalin, S., & Bates, T. (2016). A miniature scanning sun photometer for vertical profiles and mobile platforms. *Aerosol science and technology, 50*(1), 11-16.

Okuyama, K., Kousaka, Y., & Yoshida, T. (1978). Turbulent coagulation of aerosols in a pipe flow. *Journal of Aerosol Science, 9*(5), 399-410.

Paoli, R., Thouron, O., Cariolle, D., García, M., & Escobar, J. (2017). Three-dimensional large-eddy simulations of the early phase of contrail-to-cirrus transition: effects of atmospheric turbulence and radiative transfer. *Meteorologische Zeitschrift (Berlin), 26*(6).

Paugam, R., Paoli, R., & Cariolle, D. (2010). Influence of vortex dynamics and atmospheric turbulence on the early evolution of a contrail. *Atmospheric Chemistry and Physics, 10*(8), 3933-3952.

Reade, W. C., & Collins, L. R. (2000). Effect of preferential concentration on turbulent collision rates. *Physics of Fluids, 12*(10), 2530-2540.

Saffman, P., & Turner, J. (1956). On the collision of drops in turbulent clouds. *Journal of Fluid Mechanics, 1*(1), 16-30.

Strelnikov, B., Rapp, M., Fritts, D., & Wang, L. (2022). Assessment of the precision of spectral model turbulence analysis techniques using direct numerical simulation data. *Journal of Geophysical Research: Atmospheres, 127*(4), e2021JD035516.

Sun, H., Eastham, S., & Keith, D. (2022). Developing a Plume-in-Grid Model for Plume Evolution in the Stratosphere. *Journal of Advances in Modeling Earth Systems, 14*(4), e2021MS002816.

Unterstrasser, S., Paoli, R., Sölch, I., Kühnlein, C., & Gerz, T. (2014). Dimension of aircraft exhaust plumes at cruise conditions: effect of wake vortices. *Atmospheric Chemistry and Physics, 14*(5), 2713-2733.

Weisenstein, D. K., Keith, D. W., & Dykema, J. (2015). Solar geoengineering using solid aerosol in the stratosphere. *Atmospheric Chemistry and Physics, 15*(20), 11835-11859.

Appendix D-5

Advisory Committee Summary of Round 2 Reviews

Advisory Committee Summary of the Second Round of Reviews

Note: The AC summarized the second round of reviews because the Panel of Experts was not available.

After the first round of review, the 3-member Panel concluded that, though the reviewers were in agreement about potential scientific contributions for some of the research questions, they were not convinced about the feasibility of SCoPEx achieving its scientific goals. The main issues raised related to the inclusion of the last of three main scientific questions (evaluation of process-level chemical models) and the inadequate description of the engineering design elements that would ensure the balloon, gondola, and associated elements would perform as expected and successfully deliver the desired measurements.

The Research Team had the opportunity to respond to the first round of reviews, which included individual responses to each reviewer comment and summarized in their Response to the 1st Round of Review. They provided substantially more detail about their scientific plan. They also proposed to separate the detailed engineering evaluation from the scientific merit review. In addition, they elected not to focus on the third scientific question about chemical evolution as it is likely to be a long-term research topic.

The Research Team's response was redistributed to the original 5 reviewers for a second round of review. In the second round of the reviews, most reviewers were more positive in their assessments, primarily because the Research Team provided substantially more detailed information about the flight experiments, including details of locational identification of plumes, measurements of turbulence and aerosol size distributions. Reviewers also welcomed the discussion of the plume sampling, the improved explanation of the use of the lidar, and the decision to separate detailed engineering considerations from the scientific merit review document, while still providing some essential, though limited, information on the engineering design. However several reviewers still had questions about whether the engineering design would deliver the desired scientific results. While most reviewers found the proposed experiment has scientific merit, two reviewers did not agree and argued it did not have sufficient scientific merit to move forward. The Advisory Committee judged that this result is sufficient to conduct public engagement and that any public engagement should be apprised of this range of reviewers' judgements.

Outstanding issues

Key outstanding issues raised by the reviewers are summarized below. The Advisory Committee has requested responses to these items as well as a revised research plan from the Research Team. The Research Team has been advised to thoroughly update their research plan by incorporating their responses to both rounds of comments in the scientific merit review. Completing these tasks and addressing these issues is necessary before proceeding with any public engagement.

1. Rationale needed for using calcite, as opposed to sulfate aerosol in the experiment

2. Clarification needed of methodology proposed for evaluating the influence of turbulence on particle coagulation
3. The Research Team must demonstrate that they can maneuver the balloon (gondola) as detailed in the scientific plan
4. A description of funding and resources for accomplishing the experiment's scientific goals
5. An explanation is needed of specifications for the injector as the initial condition for aerosol evolution
6. A detailed possible timeline for the proposed SCoPEx test flights and decision points is needed

As noted above, the Advisory Committee has asked that the Research Team respond to these outstanding questions and update the experiment plan to reflect all the updates made throughout the review process. In this updated plan, the Advisory Committee has requested that the Research Team acknowledge the important linkage between the engineering design and scientific process and identify milestones, decision points, and potential off ramps if equipment does not perform as expected or other experiment performance issues arise.

Appendix D-6

Research Team Response to Second Round Reviews

Compiled Scientific Merit Review 2nd Round for Publication

SCoPEx Research Team response in bold-italic

As per the reviewers' terms of the reference, comments are anonymous and confidential. Instead, we summarize them [in brackets] below.

Reviewer #1

General Comments:

[It would be nice to see an updated experimental plan. I have only seen responses to the previous comments.]

The team will consult with the Advisory Committee on recommendations for a new draft of the SCoPEx Experimental Plan. When aggregating across all reviewers, a request for more in-depth studies of several physical phenomena inherent in the experiment plan is perceptible.

[Separation or integration of science plan, technical design, and engineering remains an issue.]

The team agrees that there is a diversity of responses within our community to the integration of science investigation, technical solutions, and engineering approaches. We expect this will be a point of discussion with the Advisory Committee.

[What is the scientific merit of this kind of project, in light of a possible future federal research program on experimental atmospheric research? There will be a competition for funding and the scientific merit should be clearly articulated.]

The team thanks the reviewer for raising these critical questions for the development of a federal SAI [(stratospheric aerosol injection)] program. SCoPEx was conceived in a very different environment with respect to familiarity with, and interest in, SAI research. For this reason, the SCoPEx objectives attempted to strike a balance between what is necessary to support SAI decision-making (most importantly, whether to consider SAI at all, ever), and what was useful for improving the fidelity of global chemistry-climate models (particularly the stratospheric component). The motivations for research are worth re-examining under the current circumstances for SAI understanding and interest. However, small-scale turbulence and the evolution of plumes in the stratosphere are of relevance also beyond SAI.

Question 1: Does the response from the research team adequately address the concerns raised in your review?

[The response mostly addresses the concerns raised in the previous review, and the discussion on plume sampling (e.g., orthogonal crossing paths) was useful.]

The team is gratified to learn that our improvements to the technical narrative were helpful. Moreover, we acknowledge that there is scope for further analysis of turbulence and other technical aspects of the experiment plan.

[The discussion on the lidar part has improved, though there is an uncertainty about the plume detectability by the MPL [(micro pulse LIDAR)].]

The team appreciates the reviewer caution here. We acknowledge that engineering data from the MiniMPL as

packaged in its pressure vessel with integrated steering optics would be a useful empirical input to understanding the minimum standoff range required for plume detection, and to verifying the degree of quantitative aerosol information that can be obtained from the lidar echo. We agree that the plume should be detectable from a standoff distance of 150-200 m.

[The Research Team assumes calcite particles are spherical, which is unlikely. More analysis will be necessary to deal with non-spherical, aggregate particles.]

The team agrees that the calcite monomers will not be spherical, and will likely have a faceted geometry, such as the cubic one suggested by the reviewer. Inspection by scanning electron microscope of calcite particles from one vendor in fact confirms that the particles are prismatic, irregular, but roughly spherical. We agree with the reviewer that scattering calculations with more realistic geometry will be necessary to support interpretation of the light scattering data (eg lidar and optical particle counter). The team is aware of recent work, such as Sorensen, Christopher M. "Light scattering and absorption by particles of any shape." Light Scattering and Absorption by Particles: The Q-space approach. IOP Publishing, 2022, that can assist with this task. From our non-exhaustive search of the literature, small-N aggregates of prismatic particles are a relatively unusual topic for scattering calculations.

Question 2: The proposed experiment will happen in a series of flights. What, if any, should be conditional triggers to either move forward to stop the experiment from proceeding?

[The team must demonstrate: (1) the ability to maneuver the balloon and gondola, (2) the ability to measure turbulence, and (3) the ability to qualify / quantify particle density and coagulation in the plume. And the team must be ready to spend extra time to analyze data in the actual flight (compared to simulations).]

The team thanks the reviewer for this examination of experimental goals and flight objectives. We generally agree that these are appropriate objectives and expectations about what can be accomplished within a given flight. We take note of the recommendation to leave sufficient time for analysis and to not underestimate the challenges posed by real data (as compared to models). The team really appreciates this final point because SCoPEx is motivated by the expectation that real SAI will differ in maddening ways from modeled SAI.

Reviewer #2

Question 1: Does the response from the research team adequately address the concerns raised in your review?

[The reviewer believes that the previous response did not adequately address the issues raised, and that the SCoPEx is not really a scientific project.]

The team would like further clarification of why the reviewer characterizes SCoPEx as "not a scientific project." We did learn from this second round of reviews that from this reviewer's perspective, the utilization of calcite as an experimental aerosol serves to set a precedent for release of climate-modifying material. We believe this is an important criticism and will discuss this further within this review response.

In the previous response about Science Question 3, the Research Team used the term "stratospheric turbulence" while in reality it would be "propeller-induced turbulence." Given the experimental design, the reviewer believes it is almost impossible to measure stratospheric turbulence.]

The reviewer makes an excellent point and we regret our mistake and lack of clarity in referring to "stratospheric turbulence" and "propeller turbulence" interchangeably. They are most certainly different, and this difference is

essential to answer the scientific questions we wish to address. We have made a new figure (Fig. 1) to clarify the experimental flight plan for SCoPEx. This figure is meant to clearly distinguish between stratospheric turbulence and propeller turbulence, and to illustrate that during the planned flight maneuvers, the anemometer boom locates the anemometer in propeller turbulence that has not been further disturbed by the SCoPEx gondola. That is not to say that the anemometer is measuring the propeller turbulence instantaneously, or nearly instantaneously. The propeller turbulence will have had approximately 10 minutes to dissipate before the first measurement, and an additional 10 minutes to dissipate before each subsequent measurement (via transects by the balloon platform). Our steady-state Reynolds Averaged Navier Stokes (RANS) CFD simulations suggest that the propeller turbulence exceeds the background turbulence for at least 1hour. We do acknowledge the shortcomings in RANS modeling for this application, as well as the nuances arising from the differences between realistic spatiotemporal distributions of ambient stratospheric turbulence compared with the spatiotemporally homogeneous stratospheric turbulence we imposed in our RANS simulation. The value and priority of improved CFD modeling and better representations of stratospheric turbulence will be a subject of discussion with the Advisory Committee.

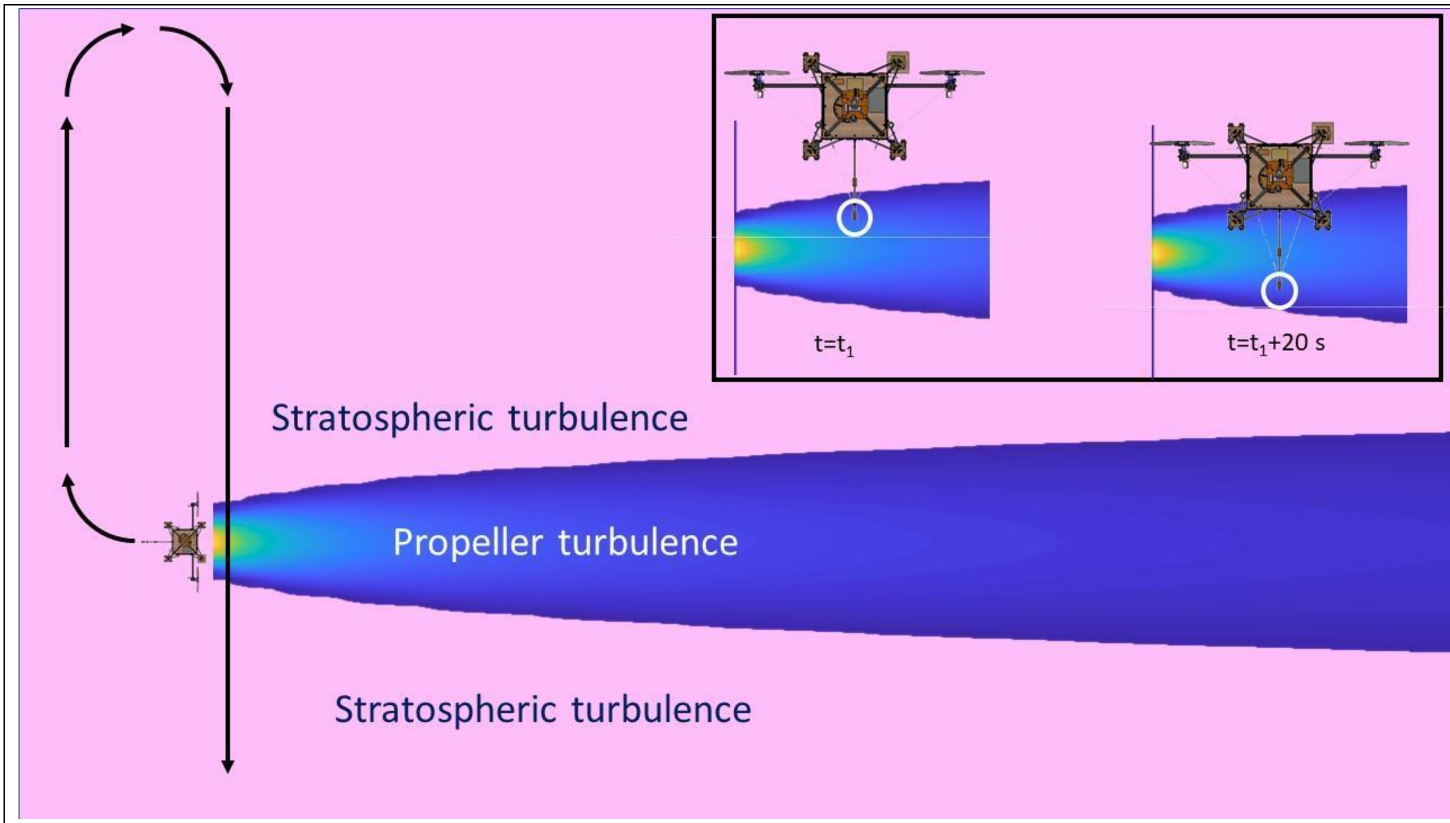


Figure 1: Details of anemometer boom and anemometer position during transects of plume. The extension of the boom in front of the gondola allows the anemometer to intercept the propeller turbulence, eg the aerodynamic wake of the propeller-gondola combination, seconds before the gondola encounters the air mass during the same transect. Note that the propeller turbulence will have had time to dissipate from its initial value at its creation by the gondola movement (a right to left movement across the page as shown here).

[Why the research team chose calcite is not clear since there are alternatives that are widely used for research (e.g., polystyrene spheres). Along with the point about stratospheric vs. propeller turbulence, It is as if the research team had been trying to spin this as a "scientific project" to have a wider effect.]

We apologize for the sloppy use of "stratospheric turbulence" indiscriminately and regret its effect on the impression of our intentions. Similarly, we appreciate a clear articulation of the perspective that our preference for utilizing calcite is consistent with a desire to create a precedent for climate-modifying materials. After conversations with our

colleagues that study the health impacts of nanomaterials that find their way into the environment, we perceived an advantage of calcite was that it would be absorbed into hydrometeors and therefore would not contribute to the environmental burden of 100 nm-range particles. As the review process has narrowed our scientific objectives for SCoPEx to quantifying fundamental turbulent and microphysical processes (and their interactions), and measuring scattered light from the particles, well-studied materials like polystyrene latex (PSL) spheres would be a very attractive option. We therefore intend to discuss these issues with Advisory Committee as we deliberate the future of SCoPEx.

[The reviewer suspects that the proposed SCoPEx is motivated for an undescribed reason, and that it is "greenwashing."]

Question 2: The proposed experiment will happen in a series of flights. What, if any, should be conditional triggers to either move forward to stop the experiment from proceeding?

[The reviewer believes that SCoPEx should be rejected purely on scientific grounds, and that this question is misleading.]

Whatever the merits or demerits, SCoPEx, is <u>not</u> a practical pathway to deployment. All SCoPEx can do is generate knowledge. It is designed to fill a knowledge gap by providing observational constraints on small-scale aerosol atmospheric processes that control relevant aspects of the temporal evolution of material injected to the stratosphere. A major goal of that knowledge is improving the scientific understanding of the atmospheric response to potential climate modifying agents. This intent is not hidden. It is impossible to know in advance whether any particular research activity, whether laboratory experiment, numerical simulation, observational campaign, or small-scale perturbative experiment, will have the effect of normalizing the injection of climate-modifying material into the climate system.

Reviewer #3

Question 1: Does the response from the research team adequately address the concerns raised in your review?

[To some extent.]

We appreciate that the reviewer believes we have made some progress in addressing the reviewer's concerns, and intend to continue to do so here.

[The reviewer appreciates that the Research Team is finally separating engineering aspects from scientific aspects, in response to many review comments.]

We acknowledge that we have really benefited from this anonymous review process and regret that we did not seek other anonymous critical reviews earlier.

[The reviewer also appreciates the dropping of the third scientific goal in the original proposal, atmospheric chemical evolution, which requires long-term efforts.]

The review process has helped us understand the expectations of our peers in atmospheric chemistry much more vividly. We take the point that we were significantly underestimating these expectations with regard to the detailed chemistry measurements that would be necessary to adequately quantify the chemistry of stratospheric aerosols and

their interactions with the ambient stratosphere.

[Exclusion of the discussion on risks and resources makes it difficult to make an evaluation.]

We appreciate this point and look forward to having exactly this discussion—about the adequacy of resources to the objectives of the investigation and its credibility among peer scientists – with the Advisory Committee.

[Many years have already passed since the beginning of the project. The management of schedule and resources for this SCoPEx project would not meet the standard of a publicly funded project.]

We agree that the approach to risk management and resourcing that has been employed by SCoPEx to date would not pass muster as a government program. We feel that we have recognized the high-risk character of the proposed investigation, and the efficacy of our risk management approach is certainly up for debate. The Advisory Committee's external review process has been very beneficial and has led us to institute significant changes to the investigation scope and plan. Furthermore, we believe we have significantly improved the science questions, and our probability of success, by re-scoping and focusing them. These changes will support a critical review of resources available and their adequacy to the objectives with the Advisory Committee.

[The lack of specifications of injectors makes it to difficult to evaluate.]

These are valid criticisms of the approach to the aerosol injector to date. In fact, we have de-prioritized the injector, and a clear enumeration of its requirements, to focus on an engineering flight to demonstrate the control and instrument support capabilities of the platform. We acknowledge the reviewer's point that the quantitative performance of the injector must be known to provide an adequate foundation for quantitative interpretation of the data.

Question 2: The proposed experiment will happen in a series of flights. What, if any, should be conditional triggers to either move forward to stop the experiment from proceeding?

[The conditions are (i) adequate funding resources, (ii) no conceivable environmental risks, and (iii) competence in launch and recovery operation of the balloon and gondola. Because SCoPEx is in a high-risk, high-reward category, the Research Team should be given as much autonomy as possible.]

We appreciate the stark summary of the risks and rewards of this investigation. We agree that the conditions enumerated by the reviewer: 1) adequate funding, 2) no conceivable climate risk posed by the experiment, and 3) the balloon operations need to be provided and overseen by qualified balloon operators.

Reviewer #4

[The Research Team now has a focused research plan by dropping long-term research areas. This has improved the proposal.]

We are grateful to the reviewer for taking the time to participate in this process.

[The Research Team should proceed with the project, though the material injection should be allowed only after the demonstration of gondola performance.]

We acknowledge and appreciate the caution about the controversy associated with the injection of active materials. From the other reviewers, we are also attuned to the necessity for the balloon and gondola to perform to its design

specifications for the investigation to be successful. We will discuss the sequencing of platform demonstration and active material injection systemically with the Advisory Committee.

Reviewer #5

Question 1: Does the response from the research team adequately address the concerns raised in your review?

[No, the Research Team does not give sufficient support for scientific merits. Specifically, the justification for using calcite in the SCoPEx project, not sulfur (a leading candidate material for solar geoengineering), is lacking. How an experiment with calcite solid particles yield insights into coagulation and condensation of sulfate liquid particles is not described.]

We thank the reviewer for the comment and will explain the contradiction between our focus on calcite with our acknowledgement that sulfur is the most likely candidate for a hypothetical Stratospheric Aerosol Injection deployment. We agree with this reviewer that sulfur-based SAI, whether deployed via gas precursors or accumulation-mode particles, will be controlled by the interactions of gas- and particle-phase microphysical processes. As was pointed out by other reviewers, testing interaction of these multi-phase interactions credibly will require additional instruments beyond those we have been analyzing and engineering for the current supported phase of SCoPEx flights. As another reviewer pointed out in the 2nd round of reviews, it is likely that SCoPEx experimental data will be more complex and more difficult to interpret than we anticipate. And as yet another reviewer points out, SCoPEx is a high-risk investigation that would provide an unprecedented dataset of observations about aerosol injections in the stratosphere. To manage the risk while building a necessary but not sufficient process-level understanding of aerosol injection, we are therefore focusing on solid aerosol materials. This eliminates the need to understand the interactions of the gas and solid phase. And following on a different reviewer's critique, we will strongly consider a solid aerosol that is well-studied for calibration purposes, but not under consideration for SAI. Using such a well-studied calibration aerosol will reduce risks associated with data interpretation. We agree that we have not provided a clear narrative linking how to build a better understanding of sulfur-based SAI on an improved understanding of solid aerosol turbulence-microphysics interaction. However, as mentioned by the reviewer an improved understanding of small-scale turbulence and plume evolution is a small, first step toward understanding the larger scale plume evolution that is relevant for sulfur SAI.

Question 2: The proposed experiment will happen in a series of flights. What, if any, should be conditional triggers to either move forward to stop the experiment from proceeding?

[The reviewer does not have a good idea. Safety and political issues need to be considered.]

Appendix E

Societal Review Documents

Appendix E-1

Proposed Societal Engagement Plan from Advisory Committee

Proposed Engagement Process for SCoPEx

Prepared by the Independent SCoPEx Advisory Committee

Final Version, January 8, 2021

Background.

The Stratospheric Controlled Perturbation Experiment (SCoPEx) is a proposed research experiment to release small quantities of calcium carbonate powder, an inert chemical, from a balloon in the stratosphere and see how these particles interact with one another, with the background stratospheric air, and with solar and infrared radiation. The experiment could help assess the impacts or feasibility of the large-scale release of such particles in the atmosphere to reflect sunlight and offset some of the heating caused by the release of heat-trapping (or greenhouse) gases. Because such an experiment raises important ethical issues, Harvard University created the independent SCoPEx Advisory Committee to provide advice on the research and governance of SCoPEx. The Committee is reviewing the legal frameworks that apply to the experiment, scrutinizing the financial support for this work, and overseeing a peer review of the scientific and technical merits of the research. The Advisory Committee has also prepared a process for public engagement. This document is the final draft of that engagement process, and it incorporates responses to several external comments and suggestions provided on the first draft version.

Toward a roadmap for public participation in solar geoengineering experiments.

Solar geoengineering is the intentional effort to modify the global climate system through changing the Earth's reflectivity (albedo). SCoPEx aims to inform the science related to stratospheric aerosol injection (SAI), one type of solar geoengineering where particles are released in the stratosphere. While the intentional modification of local and even regional environments is not new, and we are already in the midst of human influence on global climate from the global energy system and large-scale land-use changes, the intentional modification of global climate to address climate change is unprecedented.

Such intentional efforts are, at present, without any agreed national or international governance. The Advisory Committee and the experimental team agree that any decision to utilize solar geoengineering should be based on an intentional, deliberative process that is inclusive (especially of the Global South and of those people who are likely to be most impacted by climate change or solar geoengineering), iterative (as decisions will be influenced by local context and changing circumstance), and informed by a continually improving body of evidence. However, it is not evident what the best process is for how to make decisions about experiments and technological developments that may or may not lead to larger scale solar geoengineering. That is the central issue that the Committee is grappling with in thinking about public engagement: What are appropriate and feasible ways to conduct public participation deliberation around an outdoor experiment that may or may not lead to larger scale solar geoengineering research? Outdoor experiments are tangible touchstones for the prospect of solar geoengineering, raising important questions about governance and the future of research.[1] This is clear from the fact that this Committee was assembled specifically to provide governance over SCoPEx.

It is useful to think about research governance over outdoor experiments in two extremes. In one extreme, anyone with the technological capability to do solar geoengineering research would be able to pursue that research without regard to outside governance, as some researchers have done. A danger of this is the blurry line between developing a technology and deploying it: at what scale does solar geoengineering move from an experiment to deployment? Another danger is that the development of capabilities without public oversight increases the potential that those capabilities could be misused or evolve in directions detrimental to many of those people potentially affected. An extremely important consideration is that currently the people with the capability to do the research don't currently represent, and might not take into the account, the interests of the people who are most likely to be impacted by climate change and solar geoengineering.

On the other extreme, we could suppose that all research into solar geoengineering should halt until there is a decision - or at least a process for making a decision - about deploying geoengineering.[2] A key danger of this approach is that delaying field research in solar geoengineering, potentially for many years, could delay or prevent action if emerging climate conditions make geoengineering deployment desirable or necessary, such that any delay could

---

[1] Talati, Shuchi, and Peter C. Frumhoff. 2020. *Strengthening Public Input on Solar Geoengineering Research: What's Needed for Decisionmaking on Atmospheric Experiments.* Cambridge, MA: Union of Concerned Scientists.

[2] The Committee is aware that some organizations view the 2010 decision by the Convention on Biological Diversity as a moratorium on solar geoengineering outdoor experiments. However, this decision states "that no climate-related geo-engineering activities that may affect biodiversity take place, until there is an adequate scientific basis on which to justify such activities and appropriate consideration of the associated risks for the environment and biodiversity and associated social, economic and cultural impacts." As SCoPEx will not affect biological diversity and is meeting the criteria laid out, it is consistent with the 2010 decision.

have significant consequences. In addition, a research delay could also make any deployment decision less informed and potentially more difficult. This approach may also limit discovery and the advance of knowledge, including knowledge that might be useful for other approaches to mitigate or adapt to climate change. Finally, this approach may increase the possibility that a small number of people may operate outside global norms to advance and even deploy solar geoengineering without international agreement.

The approach of the Advisory Committee regarding SCoPEx is to steer between these two extremes. We believe that public engagement in SCoPEx is an opportunity to engage multiple, diverse publics in these and related issues so that we can learn more about how to do public engagement in solar geoengineering research. This will contribute knowledge toward a process that is commensurate with the unprecedented potential impact and opportunity of solar geoengineering.

SCoPEx is a small-scale, early experiment with sufficient funds to implement the research and to support a governance process. It has the opportunity to "pay it forward" by asking SCoPEx engagement participants about what they think ideal research governance should look like for future field experiments and publicly sharing the findings. This will increase knowledge relevant to global governance and help build norms consistent with the development of global governance. We believe that if every small experiment conducted a public engagement process similar to or better than what is described here, then both science and global governance of that science would be advanced.

Scope and Outcome of Public Engagement.

The focus of the public engagement in SCoPEx has dimensions that are well-informed by existing experiences as well as dimensions that are unprecedented. The former consists of a fairly narrow and focused discussion on the particulars of the experiment: whether it is acceptable to local residents for Harvard researchers to launch a balloon and release a small quantity of inert chemicals in the atmosphere over their geographic region. Our process embodies the principles of meaningful public engagement that provide for community input into decision making, and there are existing protocols for this that we can build from. From this engagement process, the Committee hopes to gain a sense of the overall perspectives of the local community. Based on this information, along with other aspects of our review (legal, financial, and scientific), the SCoPEx advisory group will recommend whether or not the experiment can proceed.
The unique and unprecedented part of the engagement is focused on the issues associated with solar geoengineering research governance. As outlined above, and after considerable feedback

on the draft engagement plan, the advisory group has decided to focus this aspect of the public engagement plan on the following question: *What would an ideal form of research governance (oversight, transparency, & public engagement) for solar geoengineering experiments look like?* While the feedback on this question will not inform the decision making for SCoPEx, it will deeply affect future research governance for outdoor experiments. We will make this feedback public and share it with academics, policymakers, and research teams that can build formal governance processes as early as possible into future experiments. Learning what ideal engagement looks like to those that have an understanding of solar geoengineering and SCoPEx will provide invaluable feedback on what legitimacy means to external stakeholders.

To deal with the inherent, large, and systemic ethical issues around solar geoengineering, the world needs a large-scale, multinational governance system for solar geoengineering research. It is beyond the scope of SCoPEx, or this Committee, to set up such a global governance process, though we point out that there are emergent efforts underway.[3] Nevertheless, Harvard and SCoPEx have the responsibility to contribute to such a process and have the opportunity and influence to advance these processes. Given Harvard's early work in solar geoengineering, their prestige, and access, we strongly urge them to take a catalytic and cooperative role.

A Process for Engaging the Public.

The engagement process will focus on the particle release portion of the SCoPEx experiment, not the engineering test of the balloon and platform. The Committee will contract an independent and experienced engagement group to recruit citizens in and around the region where SCoPEx research experiment might occur to participate in deliberative dialogue about the experiment itself as well as governance of solar geoengineering research. Our intent with this process is not to engage all local stakeholders in the larger issues of solar geoengineering research or deployment, but to investigate a process for engagement around this research that can be used in multiple places to engage a larger, more globally representative, set of publics. The Committee will additionally contract an external group to conduct and oversee a larger-scale, global engagement process. These processes would include the following elements:

1) A briefing book
2) Framing the dialogue
3) Local deliberative dialogue
4) Global engagement and dialogue

---

[3] Examples of such efforts include the Carnegie Climate Governance Initiative, the Solar Radiation Management Governance Initiative, and additional work of environmental non-government organizations.

5) Developing recommendations
6) Sharing the lessons learned.

1) Briefing Book

Working with the Advisory Committee the independent engagement experts will develop a briefing book designed to help members of the public consider a) local scale impacts of the small scale SCoPEx experiment; b) the larger set of impacts associated with deploying, or not deploying, solar geoengineering; and c) the moral and ethical issues associated with the large scale deployment of solar geoengineering, and with the lack of such deployment.

For the local scale impacts, the briefing book will focus on the specifics of the SCoPEx research and any risks and potentials for harm from that research. As mentioned earlier, this will be situated around the idea that communities deserve the right to review and contribute to the decision of whether open-air experiments should occur in the places they live.

For the larger set of deployment impacts, the briefing book will present a summary of the impacts (both benefits and risks) of both solar geoengineering as well as global warming according to the best available science. This will include consideration of potential risks to local communities and ecosystems of both global warming and solar geoengineering, including information about which regions may experience disproportionate impacts. This will include descriptions of the regional outcomes and impacts that could result from global warming and from large-scale solar geoengineering based on research thus far, and where key uncertainties still remain.

For the moral and ethical issues, the briefing book will also provide information on the multiple dimensions of the ethical issues and uncertainties around solar geoengineering research.

The Committee will review the information in the briefing book and invite external reviewers as well. This includes scholars who have studied these issues as well as passionate and informed thought leaders with diverse perspectives. We will also review this book for accessibility and test our findings with focus groups. The goal is accessible, neutral information that invites consideration of SCoPEx and its governance (and, by extension, future solar geoengineering experiments and governance).

2) Framing the Dialogue

The Committee and external engagement group will design a set of questions to first prompt consideration of the multiple dimensions of SCoPEx, including consideration of any known and potential risks to local communities and ecosystems. A second set of questions will focus on what ideal research governance for outdoor experiments might look like, including what measures should be in place for oversight, transparency, and engagement, when these processes should occur, and who should lead them.

3) Local Deliberative Dialogue

The external engagement team will lead the local deliberative dialogue. This team must include trusted local partners who help find and select stakeholder groups and encourage people to participate. Using information on where the experiment will occur, they will identify and recruit groups of local stakeholders and publics to participate in deliberative dialogues. The stakeholder groups will reflect the diversity of the region in which the experiment takes place (including the launch and landing sites). The Committee will direct the team to strive for inclusivity and representation of communities. Importantly, the team will be required to make extra effort to include people who are from communities that are historically underserved or climate-vulnerable, or currently and historically hold less power. Using the briefing book as the reference source, the team will lead and facilitate deliberative dialogues. In these dialogues, members of the stakeholder groups will offer their perspectives about the SCoPEx experiment. As stated previously, these dialogues will also consult the participants on ideal research governance processes for future outdoor experiments.

The external team will subsequently prepare an analysis and summary of the dialogue, and a synthesis of the main points raised.

4) Global Engagement and Dialogue

The Committee will supplement this local engagement with engaging and gathering input from members of the global public who reside outside of the region of the experiment. As noted earlier, the Committee will engage a separate external team to oversee this process, which will proactively invite input from people from the research, advocacy, social equity, and other communities with interest in the research. The Committee will also offer open comments on their website so that any member of the public can participate in a discussion related to the briefing materials.

5) Developing Recommendations from Deliberation

In addition to the analysis and synthesis provided by the team that leads the local engagement, the Committee will analyze and synthesize the outcomes from the dialogues and the global engagement. This synthesis will include reflections on stakeholder perspectives and the Committee's analysis of the processes and outcomes. Based on this and completion of other elements of the review process, the Committee will make a recommendation to the SCoPEx team and Harvard on whether the experiment should proceed. This recommendation, and the materials on which it was based, will be made public and all work will occur prior to a potential particle release flight.

6) Sharing Lessons Learned

Based on our experience and outputs with this engagement process, the Committee will make revisions to the process and the guides and then share them for others to use. Our hope is that the process we develop and feedback we receive will be adapted to engage various and distributed publics for future experiments and help shape future research governance. We hope this will build awareness of solar geoengineering, engage the broad set of publics that are commensurate with the global nature of solar geoengineering, and engage publics and regions that stand to be disproportionately impacted by solar geoengineering and by climate change.

Appendix E-2

Local Engagement Guidelines

October 26, 2022

SCoPEx ADVISORY COMMITTEE
GUIDANCE ON LOCAL ENGAGEMENT

Deliberative engagement at the site of a potential balloon launch is a critical element for the governance of the SCoPEx project. Deliberative engagement is a structured, two-way process, where participants consider evidence and diverse perspectives, 'deliberate' options, ask questions, and provide feedback on the proposed experiment and associated activities that can inform next steps and future work.

Societal engagement is one of the five elements of the framework that inform the Advisory Committee's recommendation on whether and under what conditions the experiment can proceed. This document serves as high level guidance for a local engagement process for the SCoPEx project and will serve as the framework by which the Advisory Committee will assess the research team's local engagement process.

Purpose of Local Engagement

The literature firmly establishes the need for broad public engagement in solar geoengineering research.[1] A responsible, well built local public engagement process can:

- Implement and demonstrate good governance of solar geoengineering research
- Gauge local community views, concerns, and interest in the proposed experiment among participants who have the opportunity to review briefing information through discussions and deliberative activities
- Encourage and enable local participants to disseminate engagement results through their own community networks
- Learn of local knowledge, conditions, or concerns that might not otherwise be known to the research team, and provide opportunities for the research team to respond
- Enhance legitimacy and quality of the research process and outcomes by giving community members opportunities to provide meaningful input on the conduct and directions of the proposed research and reporting back to the local community on the outcomes of engagement

To achieve these goals, the Advisory Committee developed these Guidelines for Local Engagement for the SCoPEx project.

---

[1] Fiorino, D. J. (1990). Citizen participation and environmental risk: A survey of institutional mechanisms. Science, Technology, & Human Values, 15(2), 226-243.
Stirling, A. (2008). "Opening up" and "closing down" power, participation, and pluralism in the social appraisal of technology. Science, Technology, & Human Values, 33(2), 262-294.
Shepherd, J. G. (2009). Geoengineering the climate: science, governance and uncertainty. Royal Society.
NASEM (2021). *Reflecting sunlight: recommendations for solar geoengineering research and research governance*. Washington DC: The National Academies Press. doi:https://doi.org/10.17226/25762
Patt et al. (2022). Chapter 14: International Cooperation. In Skea et al. *Climate Change 2022: Mitigation of Climate Change. Working Group III contribution to the Sixth Assessment Report of the Intergovernmental Panel on Climate Change*.

Recommended Elements of the Local Engagement Process

The Advisory Committee recommends that the Research Team conduct a local engagement process that includes the elements below. The Advisory Committee will provide guidance and support to the research team and engagement facilitator to ensure the goals, process, and expected outputs of the proposed engagement meet the expectations of the Advisory Committee. Outputs from the local engagement will be reviewed by the Advisory Committee and will inform their recommendation(s) to the Vice Provost, including whether to proceed with the experiment and if so, under what conditions.

Independent Engagement Facilitator and Local Partner(s)

An independent **engagement facilitator** will work with the research team to organize and conduct local engagement activities, advise on briefing materials, produce an analysis of local input for the SCoPEx Advisory Committee and research team, and prepare an accessible summary to share with the public. The engagement process will also include a trusted, **local partner** to serve as a convener and host for the above work**.**

- The research team should hire an independent engagement facilitator (organization or individual) for the local engagement. The research team should provide their rationale for selecting a facilitator
- The engagement facilitator should have expertise and experience in designing and facilitating deliberative engagement processes[2] and will not have a real or perceived interest in the outcome of the engagement process, including in whether or not the project ultimately moves forward.
- The engagement facilitator will:
    - Design the engagement, set the parameters for a set of participants who are representative of the community, facilitate the engagement, and report on the results.
    - Recruit a local partner or partner(s) who will work with the facilitator to arrange meeting space, recruit participants, and ensure accessibility to the local community.
    - Determine the timeline of workshops and conduct roughly two to four deliberative workshops (depending on geographical scope of proposed experiment- launch and landing sites), held in places that facilitate easy access for community members / participants. Conduct one workshop online for local participants who are unable to travel to an in-person workshop.
    - Discuss with the research team and Advisory Committee any other such activities that they deem necessary or strongly recommend to conduct a meaningful local engagement process.
    - Within one month of the workshops, the engagement facilitator should provide a publicly-available written summary and analysis of the workshops to the research team and Advisory Committee.

[2] There are many examples of deliberative engagement conducted by multiple organizations. We believe that the types of engagement conducted by the following groups are useful starting points.
Consortium for Science, Policy & Outcomes, Arizona State University (https://cspo.org/)
Understanding Risk Research Group, Cardiff University

Publicly-Available Briefing Materials

The engagement facilitator and local partner, in consultation with the research team and Advisory Committee, will develop briefing materials (which should include information about how the output of deliberations will be used in the work of the research team and the Advisory Committee). The Briefing Materials will provide background information on:

- Climate change,
- Solar geoengineering,
- The proposed research and its expected benefits and risks,
- The proposed launch, and
- The principal arguments for and against (and even neutral ones). For instance, participants might be presented with both the arguments and then counter arguments (e.g., opposed to deployment but research launch is not deployment, or research has more uncertainties than that presented, etc.).

Workshop Participants

The engagement facilitator and local partner(s) will recruits participants who:

- Reside in the geographic region of the proposed launch, defined by jurisdictions such as cities, counties, and tribal governments.
- Represent a diverse sample of the local communities, with weighting to historically marginalized voices
- Are willing to participate constructively in deliberative exercises and can help disseminate engagement results to other members of their community

Participants will be guaranteed anonymity in all written materials resulting from engagement activities. No participant names or any identifying information will be made public, though participants will be free to disclose their participation if they so desire.

Workshop Structure and Format

- Workshop participants deliberate with one another about the proposed launch on topics including:
    - General impressions on the idea of solar geoengineering and research needs
    - Identify any concerns and/or benefits to the local community with research taking place in this location. If there are concerns, might they be allayed and if so, how?
    - Should the launch take place and if so, under what conditions?
    - How should research governance proceed in the future, e.g. if and how future engagements of this type ought to take place
    - Other topics as recommended by engagement facilitator and local partner
- In making their recommendations, participants are encouraged to consider both their local views and concerns, and also the implications of their recommendations for people around the world, now and into the future. Relevant information on the latter should be included in the briefing materials.
- At least one member of the Advisory Committee will attend each workshop.

Accessibility

The engagement facilitator will ensure that all elements of the local engagement process are accessible to local residents. This includes, but is not limited to:

- Language accessibility in all written and oral formats
- Mobility accessibility for all participants, in recruitment, conduct, and output of the workshops

Engagement Summary

At a minimum, the engagement facilitator will produce the following documents:

- A summary report from the engagement facilitator that will be made public and written in accessible, non-technical language
- The SCoPEx Advisory Committee will share the final recommendation(s) that it makes to Harvard University with the communities that participated in the engagement process.

Appendix E-3

Letter to SCoPEx Advisory Committee from Saami Council

SÁMIRÁÐÐI
SAAMELAISNEUVOSTO
SAMERÅDET
СОЮЗ СААМОВ
SAAMI COUNCIL

24.02.2021

To: Members of the SCoPEx Advisory Committee

cc:
Swedish Space Corporation
Government of Sweden

Please find attached an open letter from Swedish environmental organisations and the Saami Council, representing Saami indigenous peoples' organisations in Sweden, Norway, Finland and Russia, to the SCoPEx Advisory Committee regarding SCoPEx plans for test flights at the Swedish Space Corporation in Kiruna, Sweden.

We ask you to forward the letter to Ms Shuchi Talati for whom we can not locate an e-mail address after her transition to the US government.

We also ask the Swedish Space Corporation as well as Ministers Bolund, Baylan and Ernkans in the Swedish government (copied) to take note of our concerns as elaborated in the letter.

Sincerely, and on behalf of the signatories

Christina Henriksen

Christina Henriksen
President
Saami Council

Čállingoddi/Sekretariat/Sihteeristö/Secretariat/Секретариат
Postboks 162
N-9735 Kárášjohka/Karasjok, Norway

saamicouncil@saamicouncil.net
saamicouncil.net
Tel. +47 480 217 34

24 February 2021

To:
The SCoPEx Advisory Committee

Cc:
The Swedish Space Centre
Government of Sweden

Regarding SCoPEx plans for test flights at the Swedish Space Corporation in Kiruna

We are writing this letter to SCoPEx's Advisory Committee as representatives of Swedish environmental and Saami indigenous peoples' organizations to express our rejection of the Harvard project's plans for test flights related to research and development of Stratospheric Aerosol Injection (SAI) technology at the Swedish Space Corporation.

We note that SAI is a technology that entails risks of catastrophic consequences, including the impact of uncontrolled termination, and irreversible sociopolitical effects that could compromise the world's necessary efforts to achieve zero-carbon societies. There are therefore no acceptable reasons for allowing the SCoPEx project to be conducted either in Sweden or elsewhere.

We furthermore note serious problems in terms of governance and decision-making in relation to SCoPEx.

- We find it remarkable that the project has gone so far as to establish an agreement with SSC on test flying without, as we understand, having applied for any permits or entered into any dialogue with either the Swedish government, its authorities, the Swedish research community, Swedish civil society, or the Saami people, despite the controversial nature of SCoPEx. The first flight's direct purpose to enable release of particles in a later test can not be treated in isolation to SCoPEx overall intentions. We request the Advisory Committee to ensure that SCoPEx does not continue pursuing such hollow claims, but instead treat the test flight as integral to the overall goal of SCoPEX.
- It is noteworthy that Harvard University considers it reasonable for a committee whose role it is to decide whether this controversial project should go ahead, to not have any representation from the intended host country, Sweden. Instead, the committee is composed of almost exclusively US citizens and/or residents.
- We note that SCoPEx "independent" Advisory Committee appears to be extremely homogeneous, is far from representative and appointed through Harvard itself, without any inclusion of affected groups and without directly critical and non-US voices.
- The SCopEx project's comment on its Advisory Committee's draft "Engagement Process for SCoPEx" highlights core issues and shows the project's problematic approach to ethics, responsibility and decision making. The SCoPEx project states that no one research project should have to answer questions such as "Does solar geoengineering research or deployment pose a moral hazard? Is it ethical to deploy solar geoengineering, and who should decide? Can solar geoengineering deployment be governed, and can we trust that governance? Is research a slippery slope to deployment?". The SCoPEx project states that under such requirements research would have to halt, and complains that this has not been the case for other areas of research, and therefore "should not be the burden for solar geoengineering research".

> We state that precisely because of the extraordinary and particular risks associated with SAI[1], this technology and SCoPEx cannot be treated like other research. The type of key issues cited above must be considered first, and in forums that are significantly more representative and inclusive than the SCopEx Advisory Committee. Experimentation and technology development through projects such as SCoPEx must therefore be halted.

We call on the SCoPEx Advisory Committee as well as SSC to recognise these shortcomings, and to cancel the planned test flight in Kiruna.

The SCoPEx plans for Kiruna constitute a real moral hazard, and threaten the reputation and credibility of the climate leadership Sweden wants and must pursue as the only way to deal effectively with the climate crisis: powerful measures for a rapid and just transition to zero emission societies, 100% renewable energy and shutdown of the fossil fuel industry.

Stratospheric Aerosol Injection research and technology development have implications for the whole world, and must not be advanced in the absence of full, global consensus on its acceptability.

Yours sincerely,

Christina Henriksen,
President, Saami Council

Johanna Sandahl
President, Swedish Society for Nature Conservation

Mikael Sundström
Chairperson, Friends of the Earth Sweden

Isadora Wronski
Programme Manager, Greenpeace Sweden

---

[1] For example, risks for devastating, unintended impacts on global weather patterns and ecosystems, including food and water supplies for billions of people; irreversible technology lock-in over millennia to avoid the danger of termination shock; impossible requirements for continuously functioning and centralized governance regimes over hundreds and thousands of years; risks of weaponization and new, unmanageable geopolitical tensions, and not least, unavoidable moral hazard effects that could cause the world to miss its chance to incur rapid, transformative changes in time.

Appendix E-4

Letter from SCoPEx Advisory Committee to Saami Council

March 2, 2021

Christina Henriksen
President, Saami Council

Dear Ms. Henriksen:

On behalf of the SCoPEx Advisory Committee, I want to thank you for your letter of February 25, 2021.

The issues that your letter raises are of great concern to the Advisory Committee. The issues of moral hazard, the nature and degree of local involvement in the decision-making process, and the degree to which broader issues should be considered in connection with any initial test flight are all matters that the Advisory Committee has been discussing in detail.

The Advisory Committee has not yet decided what recommendation to make to Harvard University concerning the test flight. Your letter provided very useful insights. The Committee has already held one meeting devoted to discussing your letter, and we will have more.

As our deliberations continue, we expect to be able to get back to you soon with further thoughts and possible next steps.

Yours truly,

Sally Klimp

Sally Klimp
Executive Coordinator to the SCoPEx Advisory Committee