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.

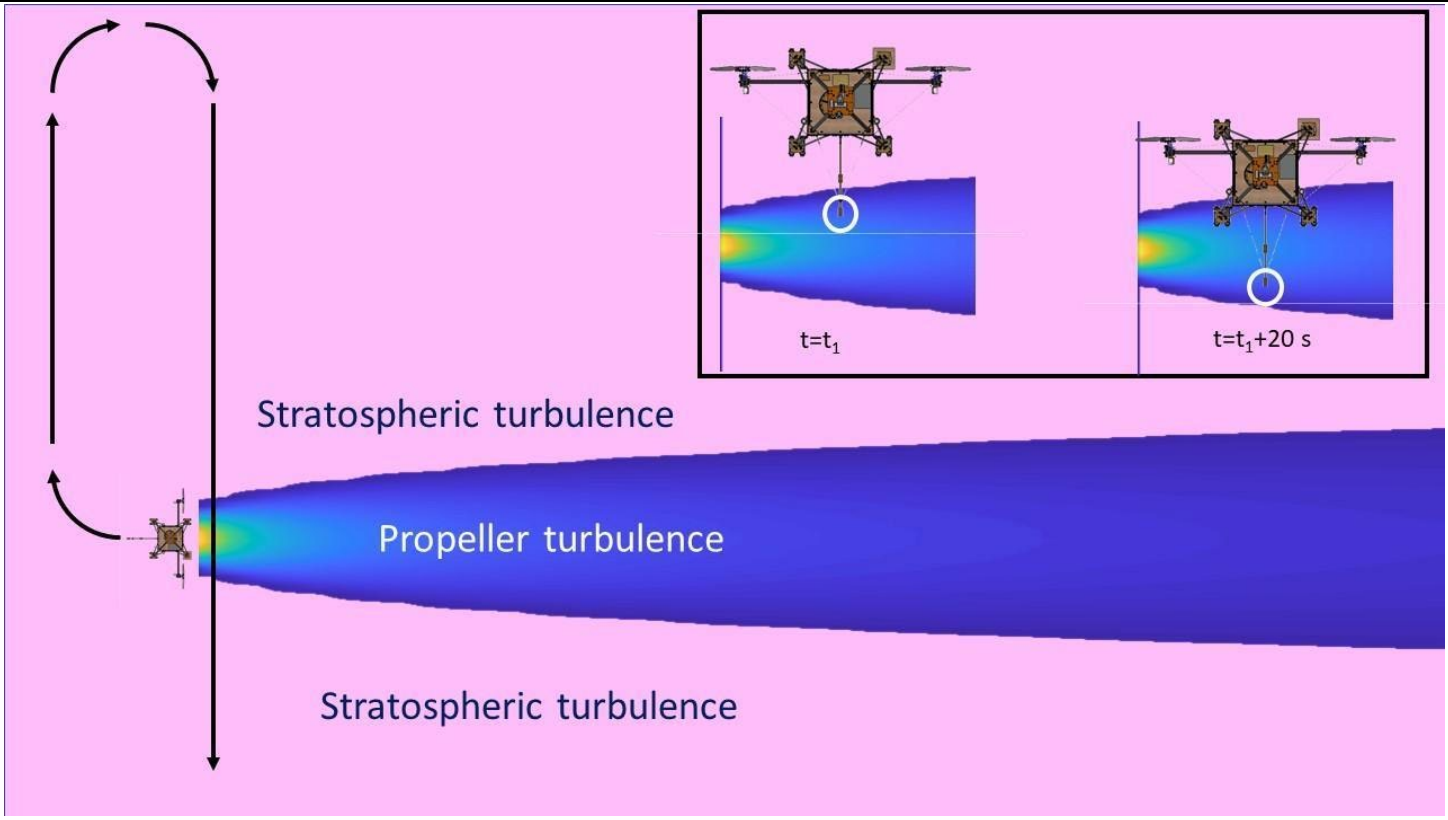

Figure 1: Details of anemometer boom and anemometer position during transects of plume. The extension of the boom in front of the gondola allows the anemometer to intercept the propeller turbulence, eg the aerodynamic wake of the propeller-gondola combination, seconds before the gondola encounters the 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.]