**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.

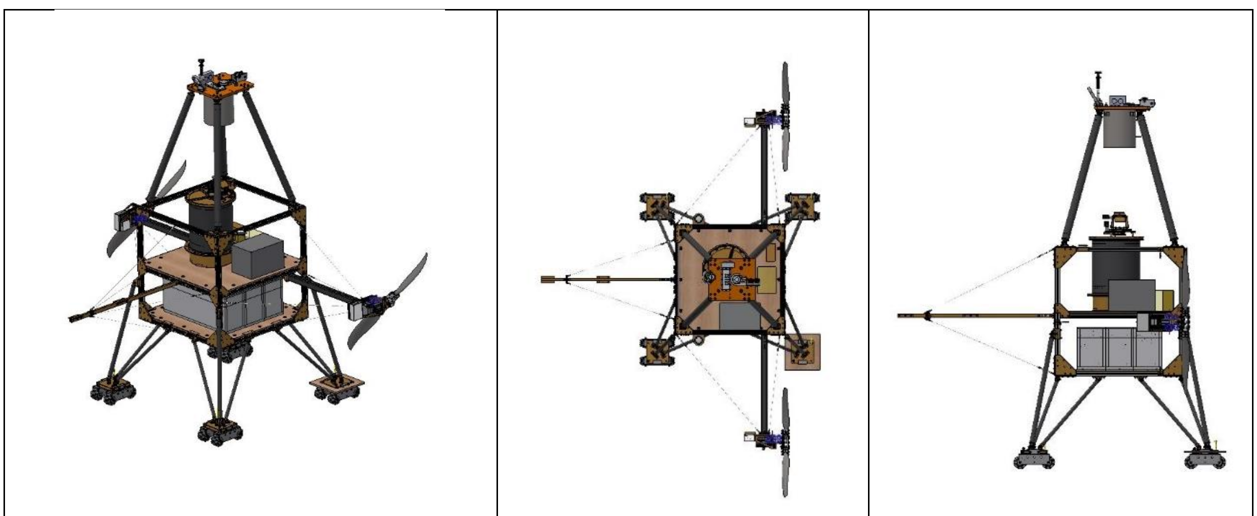

Figure 1: SCoPEx gondola includes boom to place the LITOS turbulence measurement external to the influence of the gondola and propeller aerodynamic wakes. Views are near isometric (left), top (middle), side (right). This model represents the as-built equipment for the crane test described in the appendix.

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.

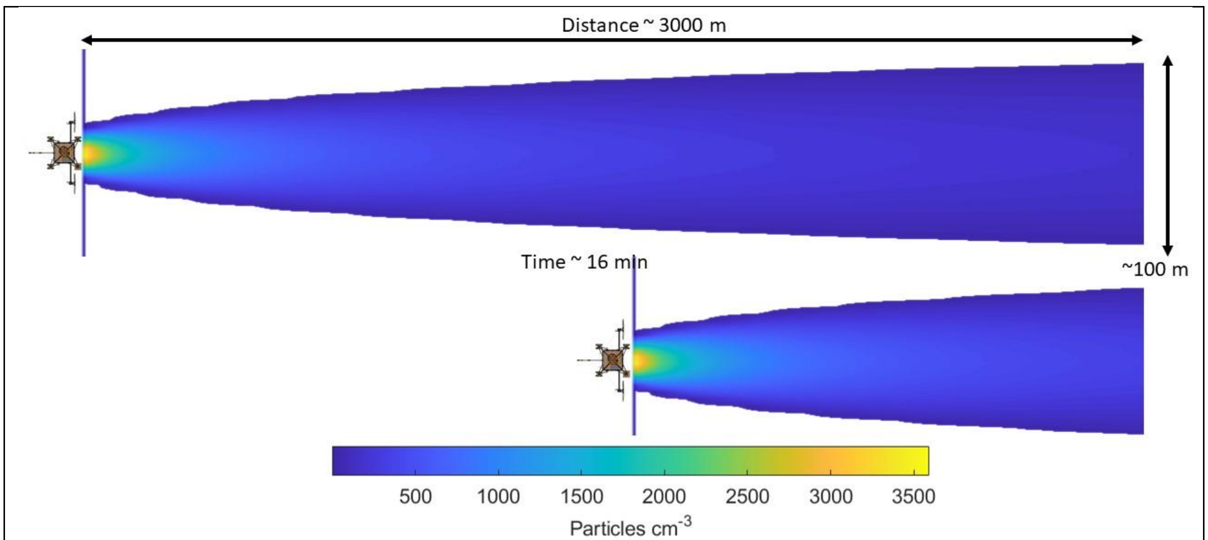


Figure 2: Plume injection (Phase 1) as viewed from above. calcite plume after approximately 8 m of travel and injection (lower plot) and after 16 min of travel and injection (upper plot). The contour colorbar is proportional the number density of monomer calcite particles and is produced by the advection-coagulation 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).

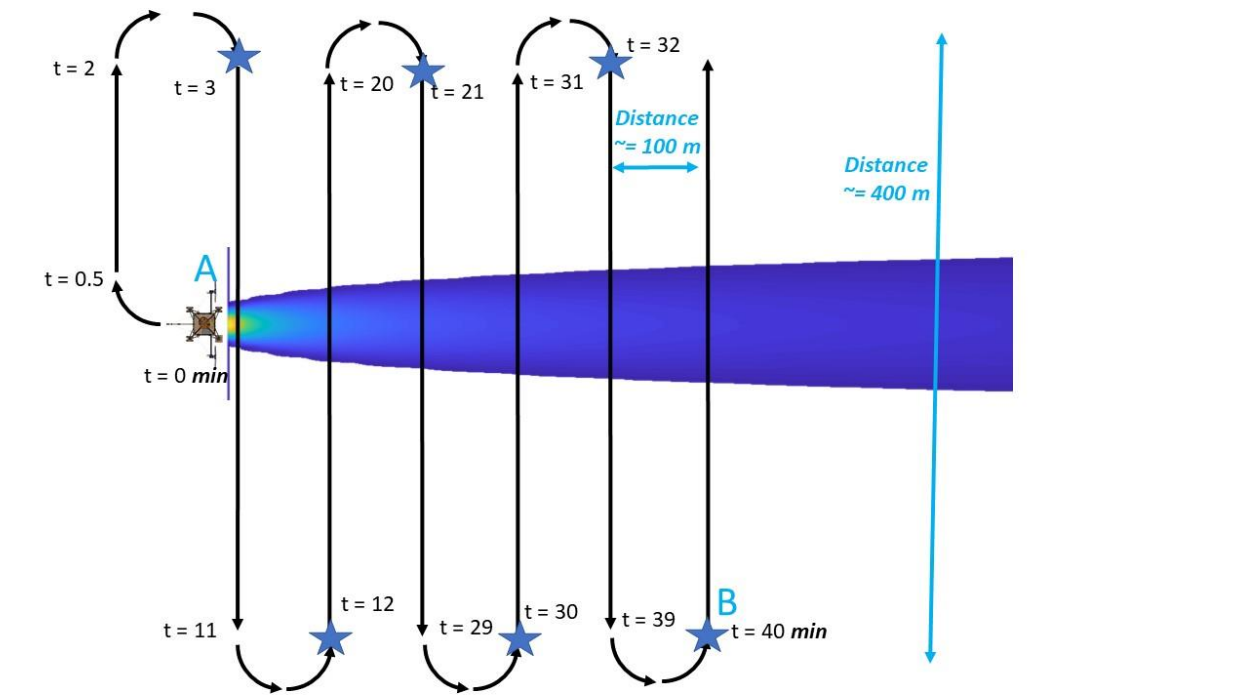


**Figure 3**: Plume sampling maneuvers (Phase 2), shown from an overhead view (gondola only). The repositioning capability of the gondola is used to turn the gondola and allow it to move relative to the plume, while changing its orientation. The approximate estimated time to perform each maneuver are 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) | $3.9\times10^{-5}$ |
| Calcite (plume tangent view, location B) | $1.2\times10^{-5}$ |
| Molecular species | $1.2\times10^{-7}$ |
| Background aerosol | $6.3\times10^{-8}$ |

**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.

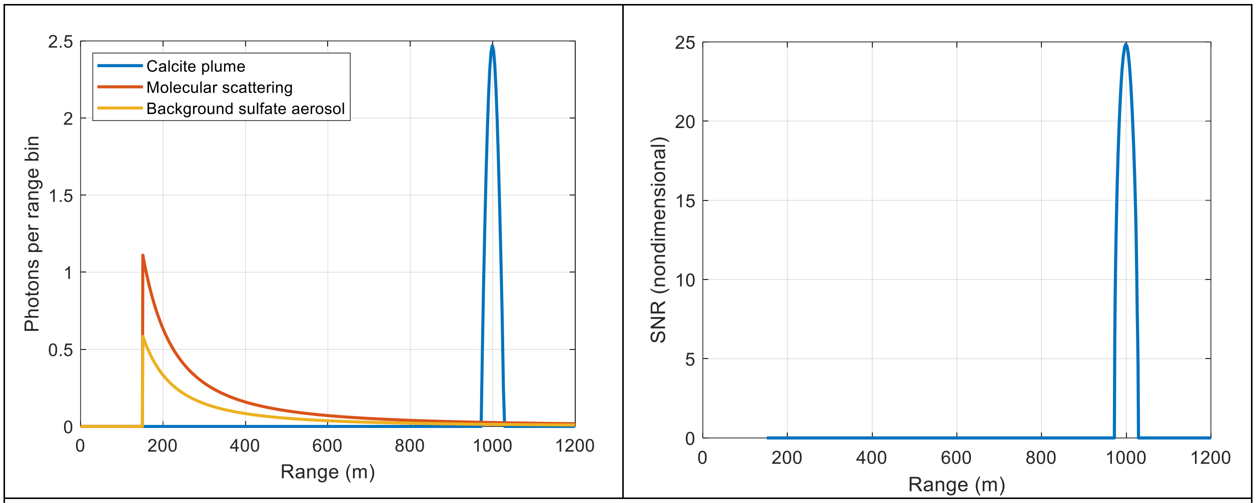


**Figure 4**: Photons per range bin for plume viewed from location B, 1000 m from plume center for a single pulse (left) and SNR for an integration of 250 pulses (right).

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.

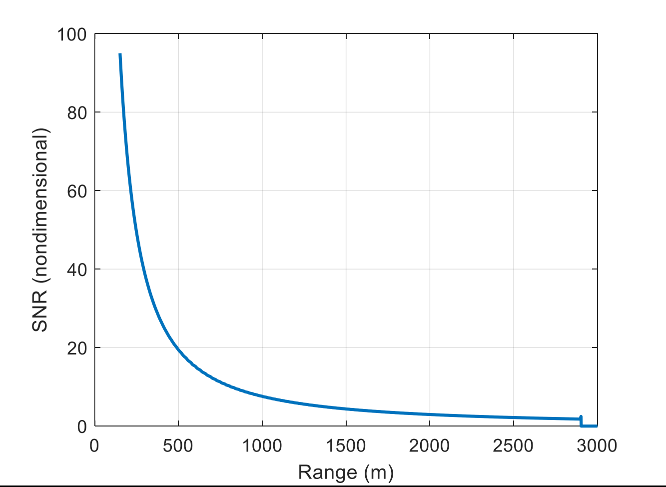


**Figure 5**: SNR of lidar measurement along axis of plume from location A, during injection, for an integration of 25 pulses.

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 $\varepsilon$ 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 $\varepsilon$ 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.

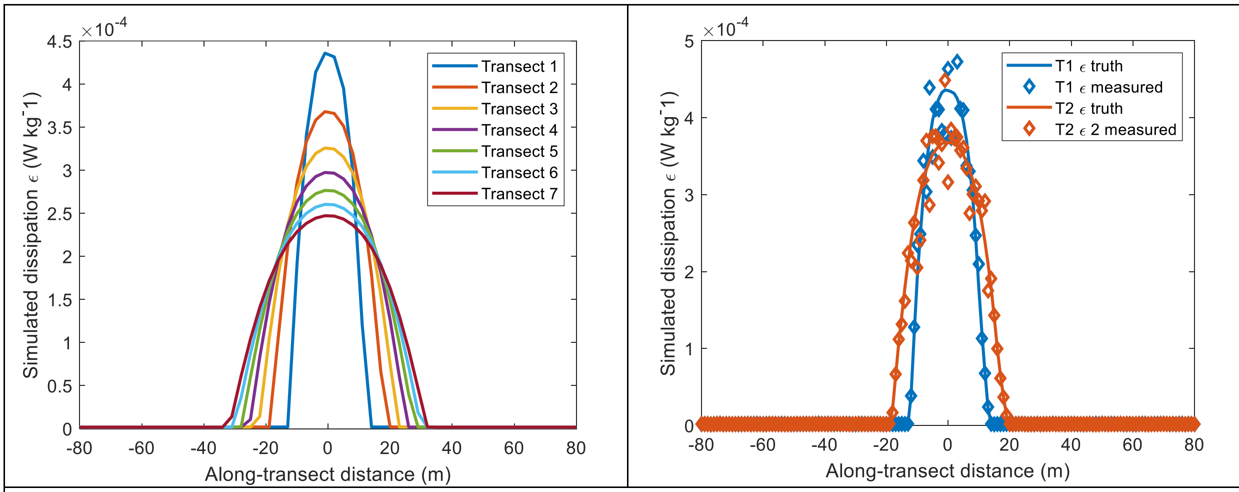


**Figure 6**: Expected profile of turbulent kinetic dissipation based on CFD inputs to Golja *et al*. 2021 for 7 transects spaced at 100 m (left). A Monte Carlo simulation using a conservative estimate of LITOS measurement noise for the first two transects is also shown (right).

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 $\varepsilon$ 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).

POPS particle size distribution measurement:

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**

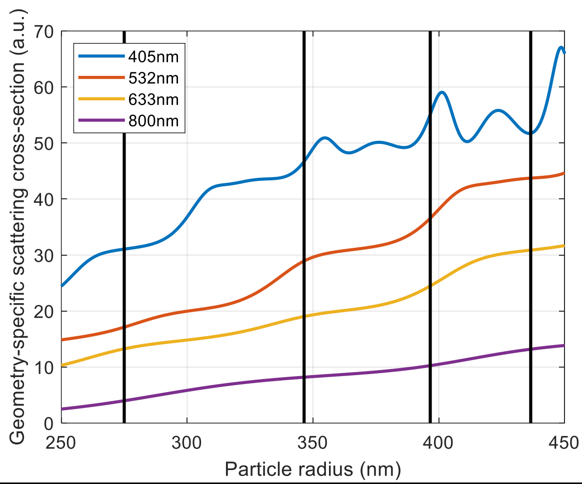


**Figure 7**: Calibration curve for POPS for calcite spheres as a function of laser wavelength. The radii of the equivalent spheres for fractal aggregates composed of N=1…4 aggregates are shown by the vertical black lines.

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.

<u>Turbulence science analysis:</u>

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.

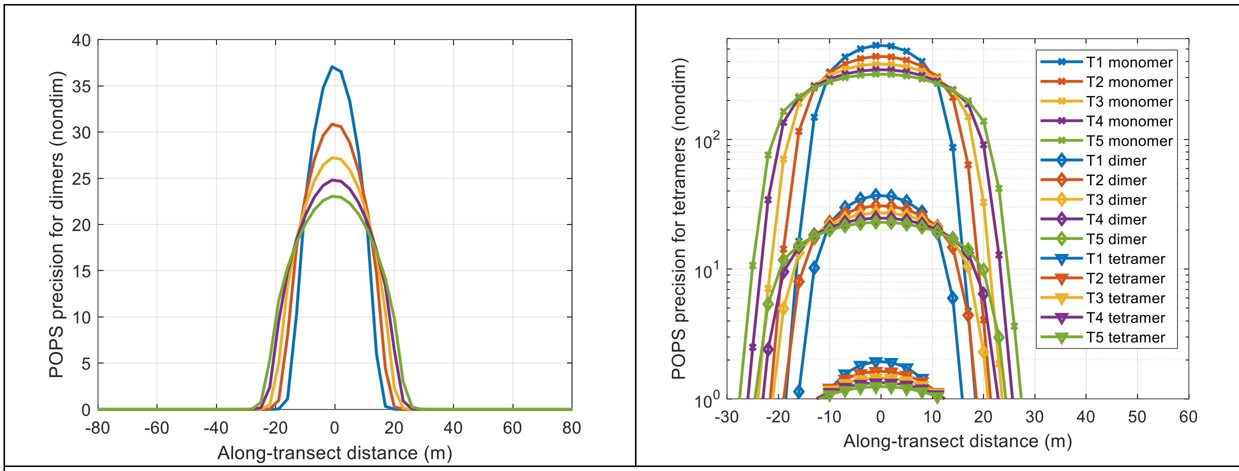


Figure 8: Precision of POPS measurement of number of particles N per 4 m transect (ΔN/N) for a flow rate of 0.1 L min$^{-1}$. The data for dimers are shown on left panel, the right panel superimposes monomers, dimers, and tetramers on a log scale. The SNR exceeds 1 in the core of the plume for all transects 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.

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