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.

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Studies of the impacts of stratospheric geoengineering depend, perhaps implicitly, on processes that would take place in the near field of an aircraft injection plume. 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 nearfield. Models can utilize existing measurements of solid rocket motor plumes, wake sampling of high altitude aircraft, convective overshooting events, and volcanic eruptions as observational analogs to compare some important stratospheric impacts with these model results. Although these bodies of literature are quite robust, they are inadequate for geoengineering because they exclude (a) relevant scales of atmospheric transport, (b) particle microphysics, and (c) chemistry of alternate geoengineering materials specific to stratospheric solar radiation management (SRM). In this document we review existing in situ data sets and describe their utility for SRM as well as areas where they are unable to shed light on the issues described above. We then suggest how new measurements could reduce uncertainty by injecting these materials into the stratosphere and comparing the results of in-situ observations with results obtained by models based on laboratory studies of both their chemical and optical properties.

Introduction

There is growing interest in researching geoengineering to reduce some of the negative effects of anthropogenic climate change . Here we focus on stratospheric injection of aerosols or aerosol precursors (SAI). Most of the rapidly growing body of literature on SAI has used general circulation models (GCMs). Modeling results have shown that SAI, when used in conjunction with emission reduction, may help mitigate some of the detrimental climate effects as compared to a world with emission cuts alone.

Stratospheric aerosols might be created using three distinct methods (a) the release of gas phase SO_2, (b) the injection of Accumulation Mode H_2SO_4 droplets (AM$-H_2SO_4$), or (c) the injection of alternative, solid aerosol (such as calcite, diamond, titanium dioxide, etc.).

Most studies that have sought to research the effects of geoengineering by means of SAI have assumed the addition of aerosol would take place by means of an injection of 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 et al. (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 (1).

Recent work by several groups (Vattioni et al. (2019), Pierce et al. (2010), and Benduhn et al. (2016)) have highlighted the potential benefits of injecting H_2SO_4 aerosol directly into the accumulation mode (i.e., Aerosols with a radius of 0.1–1.0 µm), by potentially emitting H_2SO_4 vapor into an aircraft plume(2–4). This work has suggested that one would be able to better control the resulting aerosol size distribution of the aerosol and thus the radiative forcing per unit sulfur injection, and this would allow 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 injection rates of gas phase SO_2 injection. Such a system would thus minimize the increase in the stratospheric sulfate burden. 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 models used to study the potential for AM$-H_2SO_4$ all rely on a detailed understanding of aerosol microphysics, a quantitative understanding of the interaction between the jet engine exhaust and aerosol precursor, for the particular injection technology to be used, is required. This interaction must be known due to the potential influence it will have on the nucleation and coagulation of a vapor phase

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aerosol injection. Although in situ datasets that exist can help us understand some of the elements of this complex nucleation, there currently exists no dataset with which we can validate the model results described above.

Though sulfur aerosol does exist in the background stratosphere and there are some natural analogs of broad stratospheric sulfate injections (volcanic eruptions), some have suggested that it may not be the optimal candidate for SAI and that an alternative aerosol may be most appropriate in order to mitigate SAI risks. Aerosols such as calcite ($CaCO_3$), alumina (Al_2O_3), diamond (carbon), rutile (TiO_2), and several others, have been proposed as a way to minimize some inherent risks to SAI such as ozone loss and stratospheric heating (5, 6). Although model results of these aerosol species suggest they possess optical properties that make them well suited to be used in a geoengineering scenario, (6) the stratospheric aerosol microphysics of these compounds (especially coagulation on the surface of the aerosol after injection) is poorly understood. As with AM$-H_2SO_4$ and SO_2 injections, there is a profound lack of in situ data to assess the ability to model the microphysics of alternative aerosols, as well as the stratospheric chemistry of these materials, especially with respect to ozone, a lack of which is only 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. When developing a modeling framework to simulate an SAI scenario, one can think of the problem as having two domains **1:** the nearfield domain, meaning the domain proximate to the injection point wherein the injected aerosol or aerosol precursors retain plume-like properties with complex aerosol dynamics and physical morphology and **2:** the global domain, where the initially injected plume of aerosol or aerosol precursors has mixed into the background stratosphere such that it can now be thought of as being homogeneously distributed within the spatial grid boxes in large scale GCMs.

Current global-scale model results, such as those often published by the The Geoengineering Model Intercomparison Project (GeoMIP) (7) or Stratospheric Aerosol Geoengineering Large Ensemble (GLENS) (8) are better suited to answer questions about geoengineering deployment with respect to the global domain as often times GCM chemistry schemes assume well mixed grid boxes and are unable to resolve plume scale transport. Because of massive uncertainties in our understanding of transport and aerosol dynamics in the nearfield, scientists currently using GCMs to study the global effects of various geoengineering scenarios are forced to invoke idealized initial conditions in their models. In this document we will review previous in situ studies of nearfield stratospheric plume processes, show how those datasets have contributed to our current understanding, and demonstrate a need for new experiments 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 three injection scenarios, gas-phase SO_2, AM$-H_2SO_4$, or solid aerosols, proposed above 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. Such models improve understanding in their own right and are consistent with recommendations from the model development community, for example from the Geoengineering Modeling Research Consortium's recommendation to prioritize embedded plume-scale models of aerosol formation and dynamics following injection from aircraft.

We note that sub-grid scale processes do not represent the only unknowns in global climate models that are relevant to high-fidelity simulations of geoengineering scenarios, and that there are many large scale model phenomena which should be further assessed with observational evidence. However, in this document we focus on a need for in situ data to constrain sub-grid scale processes and hope to highlight a need for reducing the uncertainty in transport and aerosol dynamics and chemistry at this scale. Eventually, if society were to design and support a rigorous geoengineering research program with the goal of sometime deploying this technology, larger scale experiments could be done to empirically assess the uncertainty in the GCMs at larger scales with respect to SAI.

Solid Rocket Motor Plumes

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. (9) 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, with the primary halogen source being HCl emitted from the combustion of ammonium perchlorate (NH_4ClO_4) present in the rocket engine. However, in studying the effects on the ozone layer, this era of stratospheric sampling provides us with 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 that the many assumptions 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 then solves for the Cl_2 concentration of a circular nighttime plume as it expands in diameter along an isentropic surface. Aircraft measurements then 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 $Cl_2 + hv \rightarrow 2\,Cl$), 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 ($\sim 28min$)(10).

Ross went on to publish a second study as a part of the RISO project in 1999, although this time looking to quantify the size distribution of alumina aerosols emitted from the rocket engines which contain particulate alumina (Al_2O_3). Ross et al. (1999) 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. While 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 (11). 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 et al. (12) and discussed later in this document . However, the data from Ross et al. (1999) were 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 (13), therefore it is imperative that we understand the coagulation and accumulation properties of these aerosols (and other aerosols of interest) in a stratospheric environment.

Stratospheric Wake Crossing

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 1990's 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 23km. 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 ER-2 sampled its own plume on several occasions by making a hairpin turn and heading into its original path, therefore measuring its own wake (Figure 1). 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 a geoengineering scenario (14).

Fahey et al. (1995) 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. (15)

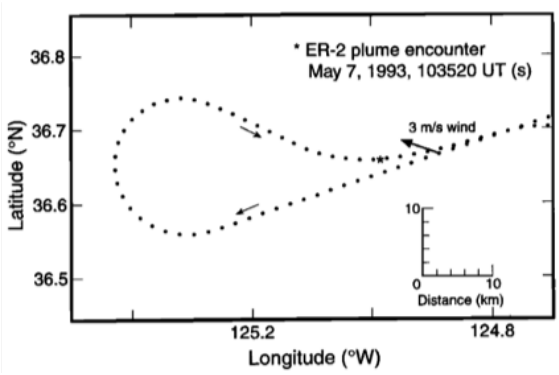

Fig. 1. Adapted from Fahey et al. 1995. (15) Shows the ER-2 flight track on a typical wake crossing trajectory.

In another study published by Fahey et al. (1995) (16) they use a similar wake crossing technique to measure the exhaust of the Concorde aircraft and develop 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 10hr 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 1000hr. The model predicted exponential mean particle diameter growth continuing right until the of the simulation at 1000hr.

Yu et al. (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". (12) 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.

As a part of the SPADE project Anderson et al. computed the flow field and chemical kinetics of the ER-2 aircraft exhaust using the Aerodyne Research Inc. UNIWAKE model to address the effects of complex plume morphology on in-plume chemistry as a function of dilution time since emission from

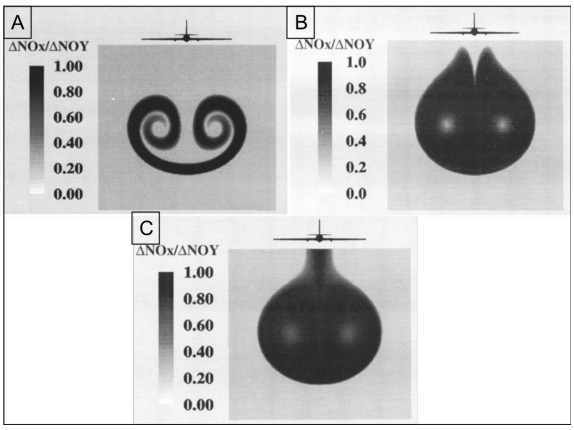

Fig. 2. Figure 2 Adapted from Anderson et al. (1996)(17) shows the chemical and morphological evolution of an ER-2 plume during SPADE at 1.7km (A), 4.8km (B), and 7.9km (C).

the aircraft engine. Anderson et al. showed that the plume morphology is highly variable out to about 5km post emission (Figure 2) and estimated that the stability of the wing vortex pair begins to break up at roughly 20km post emission. (17) Although this study was completed in the mid 1990's 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.

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→100km. 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 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.

Observations of Convective Overshoots and Volcanic Eruptions

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. Several studies have used stratospheric observations of these tropospheric air parcels by high altitude research aircraft to study the transport dynamics, and occasionally the chemical evolution, of discrete stratospheric airmasses after their injection.

Stratosphere-Troposphere Analyses of Regional Transport (START08) deployed on the National Center for Atmospheric Research GV in 2008 and intercepted a tropospheric filament north of the subtropical jet several kilometers above the subtropical tropopause. Homeyer et al. (2011) used Lagrangian back trajectories in order to determine that the tropospheric airmasses originating in the upper tropical troposphere and had an injection to measurement time on the order of 5-6 days (18). Similar observations were made of tropospheric filaments that had penetrated into the stratosphere from the polar vortex during the SPADE campaign roughly 15 years prior, however they did not have the computational capabilities to simulate the transport using backtrajectories. In these cases they were able to parameterize a model of the strain diffusion component of the plume transport and, assuming the initial filament morphology, determine a relationship between plume width and age since injection into the troposphere. (19) This work has showed us that filaments or plumes in the stratosphere are able to maintain their composition as they are transported on timescales of days to weeks. These plume morphology schemes highlight another lacking component in global climate models. Climate models often use an Eularian framework to simulate transport which can cause a sub-grid scale plume to be diluted too rapidly (often in the first timestep) into the model gridbox framework where it would have otherwise maintained its independence from the background stratosphere for days or weeks.

Nominally studies of stratospheric filament transport have been designed to assess transport on the scale of 3 to 10 days. There have also been instances where research aircraft have encountered convective overshooting events in the nearfield and sampled them out to transport distances of roughly 50km. In the 2012 Deep Convective Clouds and Chemistry experiment (DC3) the DC8 aircraft encountered a convective overshoot from a storm over the Oklahoma and sampled the outflow at four distances attempting to understand how these events can inject different soluble and non-soluble trace gasses into the lower stratosphere. (20) These data not only show us how storms can transport chemicals into the stratosphere but also how those tropospheric compounds settle along the isentropes as they get transported away from the turbulent zones near the core of the storm.

Measurements of convective systems and upper troposphere-lower stratosphere exchange as a means to interrogate stratospheric plume transport have arguably provided one of the most valuable in situ datasets we have to help us understand mid-field (10 to >1000km) plume dynamics in the lower stratosphere. Although these data are applicable in some sense to the transport of an SAI plume after its initial injection, because of the turbulent nature of a convective storm, it is difficult to measure these events at points near their injection source and 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.

Similar to convective overshooting events, volcanic erup-

tions have provided an immense amount of in situ data that had informed us about regional and even global transport of point source stratospheric injections. While these data have perhaps provided one of the greatest tests of synoptic scale plume transport, we do not discuss them here as we emphasize the need for nearfield measurements.

A Need For Small Scale Perturbative Experiments

In 2014, Dykema et al. proposed an in situ experiment coined the Stratospheric Controlled Perturbation Experiment (SCoPEx) (21). SCoPEx proposes to release a small amount (a few kg or less) of aerosol into the lower stratosphere in the altitude range of 18 to 23km from an instrumented gondola suspended beneath a high altitude balloon. This platform is well-suited to conduct detailed measurement of turbulence of the background stratosphere. The gondola will be equipped with two independently controlled propellers and be able to generate enough thrust in order to steer the payload back into the aerosol plume and intercept it over km transit lengths. Optical measurements of total aerosol count and size distribution from this experiment would provide a dataset that would offer unparalleled in situ measurements for the community to test their models of nearfield transport, aerosol nucleation and coagulation, and aerosol injector dynamics, without the complication of IMN.

Multiple methods of aerosol release have been discussed for the SCoPEx platform in order to generate the desired number of aerosols in a size distribution that maximizes the radiative effects of the injection. Potential aerosol types that are candidates for SCoPEx have been discussed in detail the literature (5, 6, 13). Initially one can imagine releasing gas phase H_2SO_4 in order to nucleate sulfate aerosol or injecting a solid aerosol such as calcium carbonate ($CaCO_3$). Vattioni et al. discusses the advantages and disadvantages of stratospheric injection of gas phase SO_2 and accumulation-mode H_2SO_4 droplets ($AM-H_2SO_4$). These modeling results suggest that $AM-H_2SO_4$ injection could provide a greater amount of radiative forcing for a given mass of sulfur injection(2). However, there is currently a total lack of literature to suggest how one would design and validate a delivery method that would produce a predictable aerosol size distribution (in the accumulation mode) under relevant stratospheric conditions. Downstream measurements of the particle distribution from the nozzles used in SCoPEx would provide important experimental validation of the injection method. Furthermore, conventional sulfur nucleation and coagulation schemes such as Vehkamaki et al. fail to provide valid parameters at extremely high aerosol precursor concentrations, such as those that would exist at the injection nozzle (22). Updates to this scheme have recently been proposed by Maattanen et al. but have yet to been validated by experimental results (23).

The ability of the SCoPEx gondola to re-position itself into the downwind plume at a chosen transport distance and time will also provide one-of-a-kind in situ data allowing a much needed test of aerosol nucleation and coagulation models at different spatial and temporal scales within the nearfield. By simplifying the initial conditions of the injection to a single aerosol precursor, as opposed to the myriad of compounds that are chaotically injected during a solid rocket motor burn or by an aircraft jet turbine, it could be determined how analogous the core dynamics of aerosol schemes are to stratospherically relevant environments and at high injection concentrations. This would provide the first plume measurements in the unperturbed, quiescent, stratosphere, as, until now, stratospheric plumes have only been generated and observed by platforms that induce violent chemical and turbulent disruptions to the system.

A further critical uncertainty relevant to realistic simulations of hypothetical SAI deployments relates to the details of the injection environment achieved for real deployment technologies: how quiescent will this environment be, and what will the chemi-ion distribution look like? Because any injection of stratospheric aerosol or aerosol precursors into the stratosphere will most likely be done using high altitude stratospheric aircraft, we must be able to predict the effect of chemi-ions (generated from the turbines) in the plume in order to ensure that the resulting size distribution is predominately those aerosols that will have the the desired radiative effects. In order to do this the ability to predict the aerosol size distribution in the absence of chemi-ions, which can only be done from a balloon borne platform that will not induce such electrostatic effects must be predicted. SCoPEx will help to elucidate the fundamental processes separate from the development of a deployment technology demonstration.

In this document we describe three fields that have produced in situ data that can begin to inform us about stratospheric plume processes and nearfield aerosol microphysics. These experiments have forced us to better understand physical processes (IMN, wake vortexes, stratospheric filament transport) that were not parts of our models before the experiments were done. Despite the fact that these datasets are invaluable to the community, we must be able to isolate and measure the nearfield aerosol microphysics and plume dynamics in an environment that is less chaotic than an aircraft exhaust plume. An experiment like SCoPEx has the ability to provide some basic tests to our foundational understanding of these scales and provide experimental constraints to models that have yet to be evaluated empirically.

In fact, the capability of SCoPEx to position precision instrumentation into an ambient stratosphere with only mild perturbations to the background wind and chemical fields is an asset for undertaking other essential scientific investigations. For example, investigators in Europe (24) and the United States (25) have developed novel high-speed wind sensors for scientific balloons to study stratospheric energy fluxes associated with atmospheric waves and the mean flow. The magnitude and variability of these energy fluxes are an important uncertainty in understanding the large-scale circulation of the middle atmosphere (26), and play an essential role in mixing tracers within the stratosphere (27).

Successful deployment of one of these sensors in the stratosphere revealed that in a conventional ascending balloon, the wake created by the balloon's rising motion contaminates the wind field (28) necessary to infer the scientific conclusions. The balloon and experiment gondola for SCoPEx are designed so that the air sampled by the experimental instruments is isolated from the balloon wake. Therefore, the SCoPEx platform, fitted with a high-speed, high-sensitivity wind sensor, would provide an ideal platform to measure stratospheric winds and infer the magnitude and mechanism of energy fluxes due to wave-mean flow interactions. Undertaking this scientific objec-

tive with the SCoPEx experimental platform would not require injection of any tracers into the stratosphere. This application of the SCoPEx platform would therefore constitute a non-perturbative means to obtain necessary 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.

The dynamical response of the stratosphere to stratospheric heating or cooling is also not well understood. For example, increasing greenhouse gases and the resulting climate change, including stratospheric cooling, are predicted to result in a speeding up of stratospheric circulation. However, current observations cannot confirm this result, potentially due to a lack of appropriate observations. There is still intense research by the stratospheric science community into improving the overall understanding of stratospheric circulation. Whereas, the direct impact on stratospheric ozone from increasing sulfate aerosol is fairly well understood, the impact on stratospheric dynamics (via heating of the stratosphere) represents a perturbation of the system that still is poorly understood.

Geoengineering model scenarios often employ the same GCMs used for climate change predictions and hence have similar uncertainties. However, a case can be made that stratospheric SRM (via sulfate injection) includes a perturbation that climate models are not optimized or specifically evaluated for, which is the stratospheric heating resulting from high levels of sulfate in the stratosphere, which may result in a modified stratospheric circulation and thus increased uncertainty compared to standard climate change model runs. It therefore is important to investigate alternative materials that will result in minimal heating of the stratosphere, i.e., just the impact from increased short-wave up-scatter (as well as minimal impact on ozone). As such materials do not exist naturally in the stratosphere the only way to quantify their behaviour is by injecting these materials in the stratosphere and comparing the chemical and physical evolution of these particles as well as the surrounding gas-phase with models based on laboratory studies

Further challenges include addressing the temporal scales that would be required to understand the effects of long term exposure of the stratosphere to the foreign aerosol. These questions may require more complex study platforms such as long duration balloons or aircraft flights with larger releases in addition to the small scale perturbation experiments described above. Larger scale experiments would allow the community to begin studying the complex morphology of stratospheric plumes as they evolve for days to weeks and would allow for much finer measurement constraints (given a known source) than can be achieved from measuring filaments of tropospheric air injected into the stratosphere. These types of studies could also reveal some of the potentially complex local chemical phenomena (such as local OH depletion) that could evolve along with a plume of high concentration aerosol precursors.

We ask the Academy take these points into consideration when developing recommendations for geoengineering research. We believe that Academy, as well as others in the field, should be stressing a need for new in situ data to understand the fundamental processes that occur at small scales and often get overlooked in the scope of global models. While the nearfield in itself may not provide the global output of climate models, sub-grid processes can dominate the initial conditions that, in-turn, determine the results of global-scale simulations.

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