**Empirical methods to reduce uncertainty about solar geoengineering**

David Keith[1], Frank Keutsch[1], and Cody Floerchinger[1]

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It's possible that the deliberate alteration in the Earth's radiative balance—solar geoengineering—can offer a significant reduction in climate risks when compared to a world with the same emissions trajectory but without geoengineering. Current estimates of the ability of large-scale geoengineering to reduce risks (its efficacy) or the extent to which it introduces new risks (its risks) are almost entirely based on numerical experiments with large-scale models. While it will never be possible to fully determine the efficacy and risks, the combination of models and experiments—both laboratory and outdoors—offers the best pathway to reduce uncertainties.

Here we focus on in situ small-scale experiments that could improve understanding of atmospheric processes and so reduce important process uncertainties in large-scale models. These experiments would thereby reduce overall uncertainty in predicting the efficacy and risks of solar geoengineering. Such process experiments can be completed at space- and time-scales small enough to have negligible climate impacts[2,3].

The importance of empirical methods is disputed, with some arguing that near-term effort should be focused on model-based studies[4]. We offer three lines of argument for an early focus on gathering empirical data.

First, solar geoengineering involves processes for which observational constraints are weak. For example, the far-field size distribution of stratospheric aerosols injected from aircraft will be strongly dependent on linked micro physics, chemistry and plume dispersal, for which very few observational constraints exist, especially for aircraft (the most likely injection method). In addition, solar geoengineering may use alternative materials such as calcium carbonate[5] for which there is *no* stratospheric observational data.

Second, the history of environmental science teaches that model-only analysis tends to build overconfidence. Depending solely on models to inform policymakers about geoengineering methods, their implementation strategy, and their unintended consequences risks inaccuracy and incompleteness in our evaluation and decision process. Models too easily produce *crisp* results with underestimated uncertainties and potentially hidden systematic biases. Experiments—particularly field observations—are *messy* reflecting complexity and unpredictability of natural systems. It is also often the case in earth science that an experimental analysis of a system reveals the importance of processes that were previously unknown or thought to be inconsequential. As such, measurements can provide key constraints to models and models can provide contextual motivation for further experimental research. This relationship spurs a back-and-forth between the experimental and modeling communities that has been an essential driver of innovation in environmental science.

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[1] Harvard University, 12 Oxford St, Cambridge MA. Contact: david_keith@harvard.edu

[2] Parson and Keith. 2013. "End the Deadlock on Governance of Geoengineering Research." *Science* 339: 1278–79.

[3] Keith, Duren, and MacMartin. 2014. "Field experiments on solar geoengineering: report of a workshop exploring a representative research portfolio." Philosophical Transactions of the Royal Society A, 372.

[4] MacMartin and Kravitz, Mission-driven research for stratospheric aerosol geoengineering, 2019

[5] Keith et al., Stratospheric solar geoengineering without ozone loss, 2016

Third, and finally, the development of experiments has lagged use of models to explore solar geoengineering scenarios—and that lag cannot by easily reversed if funding is delayed. Contemporary Earth system models exist (in large measure) from efforts that were funded to explore the climate system responses to anthropogenic forcing. In situ field experiments have not been supported in equal measure. Conducting such experiments is a multi-year task because it takes time to build up the complex hardware and large teams of specialized individuals. Suppose future policy makers want better practical understanding of the possibilities and risks of deployment and decide to initiate a well-funded research program. Modeling teams can respond far faster with existing models than experimental efforts can respond without preexisting programs, instrumentation, and dedicated scientific and technical teams. Increased funding will not be able to compensate for a lack of long-term preexisting experimental efforts because science evolves by building on previous results and because parallel experimental efforts are not as effective as sequential ones. A robust experimental community centered around climate intervention does not now exist. Building such a community takes significant time and resources. The technical innovation, infrastructure development, and personnel ramp-up that is required to deploy strong experimental teams is slow. Experimental investment needs to ramp up now if experimenters are to offer effective and timely response when needed. Delay now means less chance to reduce uncertainty—even if future policy makers sharply increase resources.

**Illustrative Examples**

Here we offer three brief examples of experiments that could fill knowledge gaps. This list is not in any way comprehensive. It is confined to stratospheric aerosols simply because this is the topic the authors know best. Many process experiments relevant to Cirrus Cloud Thinning (CCT) or Marine Cloud Brightening (MCB) that could no doubt offer similar chances to reduce process uncertainties and so improve large-scale models. Moreover, CCT and MCB have uncertainties that are tightly linked to process uncertainties about clouds that are important contributors to uncertainty in climate and weather prediction, so experiments for these solar geoengineering technologies are more likely to yield co-benefits to climate science.

These examples are rough schematics that would no doubt evolve under serious scrutiny. We offer them simply as illustrations to encourage the community and the NAS committee to consider a range of experiments and the role they might play in improving understanding of solar geoengineering. All three examples are small extensions of existing experimental methods using technology that either exists or could be easily developed.

***Near-field aerosol formation in stratospheric aircraft plumes (timescale: minutes to hours)***

*Motivating questions*: Stratospheric aerosol would most likely be deployed from an aircraft. Measurements of stratospheric aircraft wake crossings have shown that the far-field aerosol size distribution is strongly dependent on near-field processes such as ion formation within the plume.[6] While the importance of the near-field may be obvious for solid aerosol injections (such as calcite) or for the injection of accumulation mode $H_2SO_4$ to better control the size distribution of sulfate aerosols, it is also important for the injection $SO_2$. The resulting aerosol size distribution from an $SO_2$ injection depends on the oxidation dynamics of $SO_2$ to $SO_3$, which in turn depends on local chemistry within the plume rather than on the background oxidation rate of $SO_2$ in the well mixed stratosphere[7].

[6] Yu and Turco, The role of ions in the formation and evolution of particles in aircraft plumes, 1997

[7] Pierce et al. Efficient formation of stratospheric aerosol for climate engineering by emission of condensable vapor from aircraft, 2010

*Experiment*: relevant data could be gathered using a high-altitude aircraft to generate and then reenter a plume. Simulations suggest that appropriate size distributions can be generated with an emission rate of about 10 kg $H_2SO_4$ $km^{-1}$. If the minimum length of a plume that can be operationally studied by reentry with a single aircraft is roughly 30 km[8], then the total mass of released aerosol can be small as 100 kg. A single aircraft such as the NASA WB-57 or ER-2 might be used. It might be possible to reenter a plume several times exploring plume expansion and aerosol dynamics over duration from about 5 min to more than an hour. The most important aircraft measurements would include aerosol size distribution and LIDAR along with $CO_2$ and CO that allow measurement of plume dilution from engine exhaust signature.

***Far-field evolution of injected stratospheric aerosol (timescale: days to weeks)***

*Motivating questions*: Introduced aerosols and aerosol precursors such as $SO_2$, will interact strongly with the background chemistry of the stratosphere. Uncertainties in stratospheric chemical transport models will grow larger the further conditions depart from typical stratospheric conditions for which the models were parameterized and validated. Conditions in an aerosol plume will generally (i) have higher concentrations of numerous compounds than those for which the models were parameterized and (ii) may involve compounds for which the models were not calibrated at all; to date, there have been very few chemical transport models that focus on stratospheric aerosol systems. These reasons demand testing model results of the physical and chemical evolution of aerosols in stratospheric air against in situ observations.

Chemical dynamics in the stratosphere have a strong diurnal dependence. Measurement over a significant number of diurnal cycles would greatly improve our ability to understand physical and chemical evolution, particularly the effect of the variation between nighttime and daytime stratospheric chemistry on an injected aerosol plume. While solar geoengineering aerosols will undergo chemical and physical changes over periods of more than a year in the stratosphere, measurements over a period of 1-2 weeks would provide substantial confidence in understanding the longer-term chemical and physical evolution of both stratospheric particles and gases.

*Experiment*: In order to observe an aerosol plume for several days, an aircraft could release a few tons of aerosol in a set of parallel segments with length order 30-100 km creating a structed patch of perturbed air. A super-pressure balloon with propulsion and plume tracking (LIDAR and small aerosol payload) capabilities could then accompany the plume for many days observing its structure with LIDAR. The balloons propulsion system need only have enough control authority to keep position with the plume center-of-mass in the face of wind shear. An aircraft with a full aerosol instrument payload would then, with the guidance of the tracking balloon, transect the aerosol plume as it was transported through the background stratosphere. This would likely need to be a second aircraft if payload capability was too small to carry both instruments and equipment for aerosol dispersal. The tracking balloon would direct the aircraft through the aerosol plume such that it would measure the aerosol composition and properties periodically.

***Aging of solid aerosol in the stratosphere***

*Motivating questions*: Many uncertainties exist in our ability to predict the evolution of the physical and chemical properties of aerosols over their stratospheric lifetime. How do aerosols age in the presence of stratospheric gases and UV exposure? How does that aging alter their optical properties and chemical reactivity? Experimental evidence of aerosol properties at this scale becomes a necessity when dealing with alternative aerosol material (such as calcite) as there exists no analogous measurements of these types of materials in stratospheric conditions.

[8] Fahey et al. In situ observations in aircraft exhaust plumes in the lower stratosphere at midlatitudes, 1995

*Experiment*: Expose aerosol materials to stratospheric conditions over months and then recover them for laboratory analysis. X (formerly known as Google [X]) Project Loon has demonstrated that small super-pressure balloons can be controlled by changing buoyancy using a compressor ballonet for durations of up to roughly one year[9]. A non-reactive substrate could be coated in an aerosol of geoengineering interest and mounted into an exposure system suspended from a super-pressure balloon. Upon command the system would purge the sample with non-reactive gas and retract it into a mechanically robust container designed to be isolate the samples from the atmosphere during recovery. This system could be flown in the stratosphere for many months until commanded to release the payload for recovery. Aerosol coated substrates could be exposed for varying amounts time or under specific conditions. This system would be analogous to the AirCore[10] system for collecting samples of stratospheric air. Analyzing the samples in a laboratory setting would allow for a more comprehensive analysis than could be completed in any plausible airborne sampling payload.

[9] Friedrich et al. A comparison of Loon balloon observations and stratospheric reanalysis prducts, 2017

[10] Karion and Sweeney, AirCore: An Innovative Atmospheric Sampling System, 2010