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DOI

[10.1126/sciadv.adi8594](https://doi.org/10.1126/sciadv.adi8594)

Publication date

2024

Document Version

Final published version

Published in

Science Advances

Citation (APA)

Feingold, G., Ghate, V. P., Russell, L. M., Blossey, P., Cantrell, W., Christensen, M. W., Diamond, M. S., Gettelman, A., Glassmeier, F., & More Authors (2024). Physical science research needed to evaluate the viability and risks of marine cloud brightening. *Science Advances*, 10(12), Article eadi8594. <https://doi.org/10.1126/sciadv.adi8594>

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ATMOSPHERIC SCIENCE

Physical science research needed to evaluate the viability and risks of marine cloud brightening

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Marine cloud brightening (MCB) is the deliberate injection of aerosol particles into shallow marine clouds to increase their reflection of solar radiation and reduce the amount of energy absorbed by the climate system. From the physical science perspective, the consensus of a broad international group of scientists is that the viability of MCB will ultimately depend on whether observations and models can robustly assess the scale-up of local-to-global brightening in today's climate and identify strategies that will ensure an equitable geographical distribution of the benefits and risks associated with projected regional changes in temperature and precipitation. To address the physical science knowledge gaps required to assess the societal implications of MCB, we propose a substantial and targeted program of research—field and laboratory experiments, monitoring, and numerical modeling across a range of scales.

INTRODUCTION

As the evidence for a warming planet and the concomitant effects on temperature and precipitation extremes mounts (1), there are increasing calls for studying whether deliberate intervention in the climate system could help avoid the worst impacts on human populations and ecosystems (2–4). For example, solar radiation management (SRM) proposes to reduce the amount of energy absorbed into the climate system by increasing the reflection of incoming solar radiation into space (5). SRM does not address the fundamental need to reduce greenhouse gas emissions and carbon dioxide, without which a lasting reversal of global warming cannot be achieved; rather, SRM is proposed as a possible means to temporarily counteract global warming, while these decarbonization efforts are pursued. SRM would not reduce deleterious effects associated with CO₂ such as ocean acidification. The two leading approaches to SRM are stratospheric aerosol injection (SAI), the injection of particles into the stratosphere to reflect some fraction of incoming solar radiation, and marine cloud brightening (MCB), the injection of aerosol particles into low-level, liquid marine clouds that typically cover large

areas of subtropical oceans to increase their reflectance of solar radiation (6). As MCB is generally acknowledged to have greater uncertainty in its technical feasibility than SAI, this assessment addresses MCB.

The underlying physical processes

The microphysical underpinning of MCB is the occurrence of bright, linear features in cloud fields that sometimes appear downwind of ships and volcanoes. Such features were identified in early satellite photos of cloud fields by Conover (7), who attributed them to ship emissions that led to higher aerosol and cloud droplet concentrations. They have since become known as “ship tracks.”

MCB proposals would use saltwater spray instead of plumes of sulfur-rich emissions from ship stacks or volcanoes to increase the aerosol concentration in the boundary layer. The droplets in the saltwater spray would evaporate to produce fine aerosol haze particles (equivalent dry diameter of approximately 50 nm) that would ideally be carried up to the cloud layer by turbulent and convective air motions. In the clouds, the plume of increased aerosol particles would lead to elevated cloud droplet concentrations compared to the surrounding region. As a cloud with higher droplet concentration reflects more sunlight back to space than a similar cloud with lower droplet concentration (8), MCB has the potential to be an effective SRM technique, at least at the local scale (9–11). Figure 1 provides a visual of some of the key small-scale aerosol–cloud–marine boundary layer processes that are at the heart of MCB, and their desired enhancement of cloud brightness (or reflectance).

To date, there is observational, theoretical, and modeling evidence that an increase in aerosol particle concentration can result in higher drop concentrations and brighter clouds. Less clear is how cloud amount (the amount of liquid water, spatial coverage, and cloud persistence) responds to aerosol perturbations. Both positive and negative liquid water adjustments (12–17) and cloud fraction adjustments (18–21) have been documented. Increases in cloud

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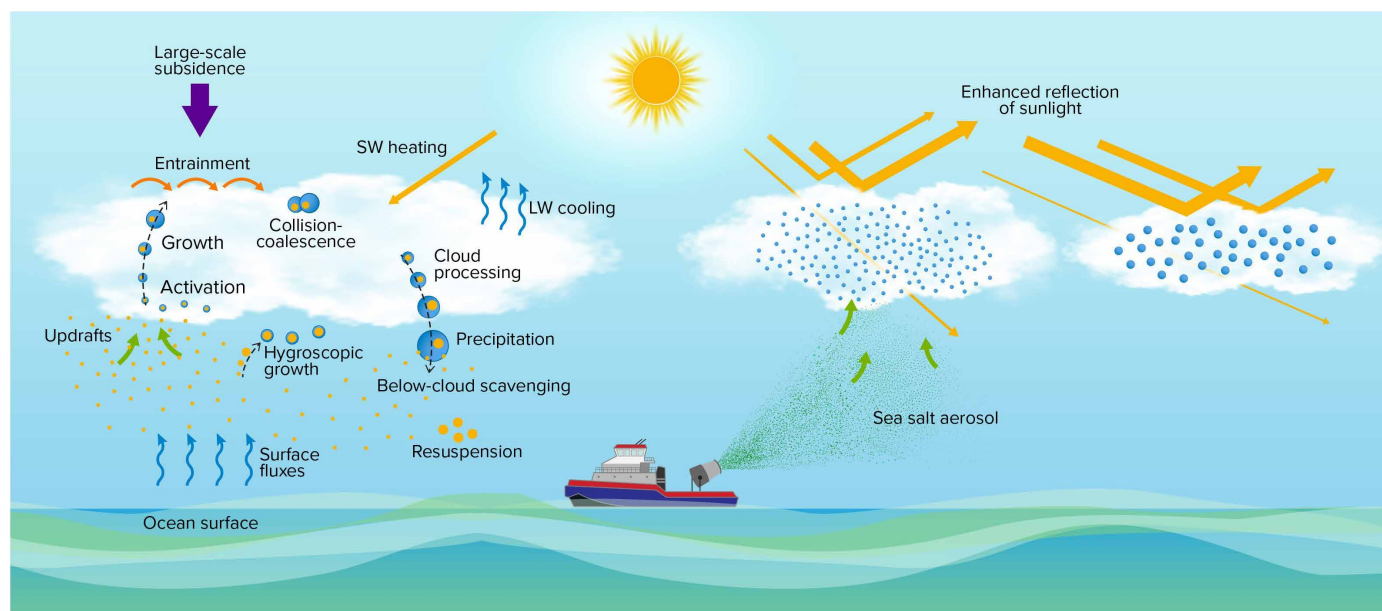


Fig. 1. Marine cloud brightening proposals using ship-based generators. Aerosol particle generators would ingest seawater and produce fine aerosol haze droplets with an equivalent dry diameter of approximately 50 nm. In optimal conditions, many of these haze droplets would be lofted into the cloud by updrafts, where they would modify cloud microphysics processes, such as increasing droplet number concentrations, suppressing rain formation, and extending the coverage and lifetime of the clouds. At the cloud scale, the degree of cloud brightening and surface cooling would depend on just how effectively the droplet number concentrations can be increased, droplet sizes reduced, and cloud amount and lifetime increased. On the left are shown details of the key aerosol, cloud, dynamics, and radiation processes in the marine boundary layer that are the foundation of MCB in shallow liquid clouds that reside close to the Earth's ocean surface (104). The strong coupling between these processes presents interesting challenges and opportunities for understanding the outcomes of seawater haze injections into these clouds.

amount (positive adjustments) are associated with rain suppression (22), while enhanced evaporation of smaller droplets (23) and entrainment feedback (24–26) tend to reduce cloud amount (negative adjustments). These adjustments depend on cloud state, time, and scale and have an inordinately large impact on whether cloud scenes become brighter or darker (19, 27–29). The initial increase in drop concentration and associated brightening, together with these adjustments, determines the “susceptibility” of clouds to aerosol perturbations (defined throughout this document as the change in reflectance for an incremental change in aerosol) and therefore the potential effectiveness of MCB. The problem has long been studied in the context of the climate forcing associated with aerosol-cloud interactions, the least well-understood and most poorly quantified of all climate forcings (30). Figure 2 sketches the key processes depicted in Fig. 1, key geophysical variables, and potential response pathways—both favorable and unfavorable—along which changes in cloud reflectance might occur.

Warm marine boundary layer stratocumulus clouds that blanket vast areas of eastern subtropical oceans have typically been considered ideal candidates for MCB; however, these cloud decks exhibit distinct seasonal and diurnal cycles (31, 32) associated with changes in regional meteorology and covarying aerosol conditions that affect their susceptibility to aerosol injections (33). For example, despite the regularity of ship traffic, ship tracks are not always visible; their manifestation in terms of brightening depends on factors such as meteorological conditions, the characteristics of the cloud, and the background aerosol, to name a few (9, 15, 34–36). Potential changes in the frequency of occurrence, amount, and susceptibility of these clouds in a warmer world and the possibility that MCB activities

might, in turn, modify these properties exemplify the complex multiscale nature of the problem.

Scope of this paper

This paper will focus on the physical science challenges and risks of MCB. It will draw heavily on our broader understanding of aerosol-cloud interactions in the climate system based on many years of marine boundary layer cloud studies and consider the particular knowledge gaps and challenges specific to MCB. Analogs to MCB like ship tracks and effusive volcanic eruptions will be considered, with the recognition that while none of these provide complete evidence for MCB effectiveness, several provide constraints on the magnitudes of the relevant processes.

The broader social, ethical, ecological, economic, and governance aspects of MCB and its research, as outlined in the NASEM Report (2), will not be discussed—primarily because the authors lack expertise in these fields. Furthermore, the recommendation in (2) is for the establishment of an SRM research program conditional on the simultaneous commitment to decarbonization. The absence of treatment of these societal aspects should in no way be construed as a dismissal of their importance.

VIABILITY

To assess whether MCB could be technically viable requires quantification of the radiative forcing associated with aerosol-cloud-climate interactions. MCB is built on the idea that a local scale (on the order of 10 km) or regional scale (on the order of 100 km) injection of particles into clouds can influence the reflectance of incoming solar

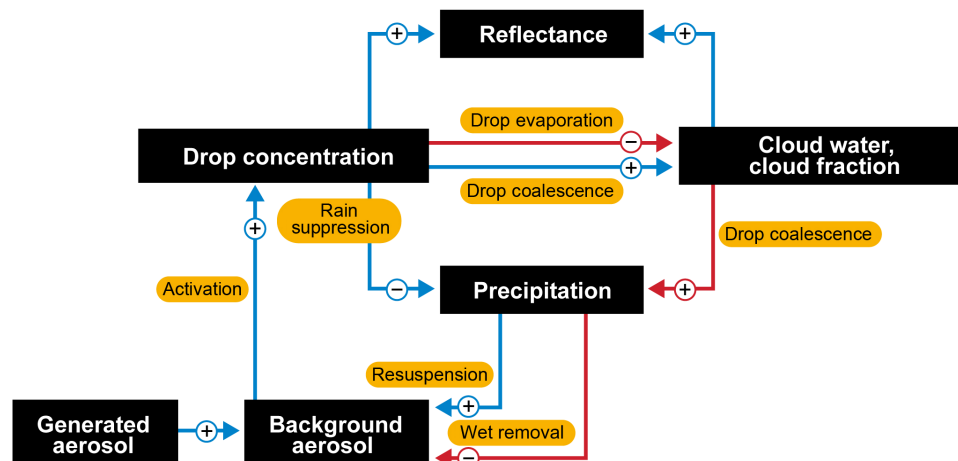


Fig. 2. Primary microphysical pathways between system variables in response to marine cloud brightening. Blue arrows indicate pathways along which clouds are optimally brightened and red arrows indicate counterproductive pathways that offset cloud brightening. Gold text boxes indicate the processes that drive the changes in variables. Plus (+) and minus (−) signs indicate the expected response of the receiving variables. The separation of cloud microphysics (i.e., “drop concentration”) and cloud macrophysics (“cloud water and cloud fraction”) represents the impact of injected aerosol directly via aerosol-cloud interactions and cloud adjustments. The two competing pathways between drop concentration and cloud water and cloud fraction reflect the documented possibility of both desirable (suppression of drop coalescence; blue) and undesirable (droplet evaporation; red) responses. The desirable pathway is characterized by increased drop concentration, larger cloud water and cloud fraction, and, if some precipitation does fall and evaporate below the cloud, resuspension of aerosol into the atmosphere. The undesirable pathway suffers from drop evaporation, precipitation, and removal of aerosol to the surface—all of which reduce drop concentration, cloud water, and cloud fraction. Note that all of these microphysical processes act in clouds, and a major challenge would be to seed in optimal conditions and with optimally sized aerosol particles so as to enhance brightening.

radiation (albedo) at globally relevant scales. [MCB is sometimes also considered as a means to reduce temperatures in sensitive ecosystems at regional scales, such as coral reefs (37–39).] Global relevance requires consideration of MCB within the context of the Earth system. Figure 3 depicts how local-to-regional scale perturbations contribute to the global radiation budget, along with associated feedback to the atmospheric state, which in turn affects the likelihood of the success of MCB. Current assessments of aerosol-cloud-climate forcing based on Earth system models (ESMs) and other lines of evidence suggest that anthropogenic pollution, emitted by large population centers, industries, and other human activities but distributed globally by general circulation, has introduced a global cooling effect of $-1.3 \pm 0.7 \text{ W m}^{-2}$ (30), offsetting approximately one-third of the anthropogenic greenhouse gas warming by CO_2 , CH_4 , and other trace gases via both its direct radiative effect and its influence on clouds (30, 40).

Estimates of the radiative effect of emissions from shipping traffic, the primary analog for proposed MCB activities, vary considerably: Observational studies of manually detected ship tracks suggest a global forcing in the range of -0.0004 to -0.0006 W m^{-2} (41) but such low estimates are likely to be heavily biased toward the less frequently occurring strongly visible tracks (42, 43). Estimates of regional radiative forcing due to emissions from an entire shipping corridor (44, 45) are orders of magnitude larger than those obtained by Schreier *et al.* (41). Global modeling studies suggest a range of -0.06 to -0.6 W m^{-2} (46–50). These high uncertainties are the result of uncertainties in aerosol emissions, pre-industrial aerosol amounts, the susceptibility of cloud reflectance to aerosol perturbations, the difficulty of detecting aerosol signals in noisy background cloud fields, and our imperfect models (e.g., poorly resolved aerosol loading, cloud amount, and aerosol-cloud interactions).

The local-to-regional nature of the proposed MCB intervention presents similar challenges to the quantification of aerosol-cloud-climate forcing. A robust understanding of the global implications of a locally-to-regionally based seeding effort requires far better resolution than is currently achievable in ESMs—and even global storm resolving models (grid spacing of a few km)—so that the extent to which local-to-regional injections scale up to global changes in reflectance is highly uncertain. At the scale of an updraft, microphysically detailed parcel models can resolve the size and composition dependence of aerosol growth to droplet sizes. At scales on the order of 10 to 100 km, large eddy simulation (LES) models that resolve the most important processes and scales (tens to hundreds of meters) are well suited to quantifying aerosol-related cloud brightening and cloud amount adjustments but are unable to represent larger-scale responses in space and time. The ability to capture a range of responses richer than those obtained from large-scale models is foundational for a more robust understanding of local-to-global responses [see, e.g., (51)].

At the observational end, decades of data acquired by surface sites, in situ aircraft missions, and more recently uncrewed aerial systems (UASs, generally known as drones), have taught us much about cloud microphysics: droplet formation, drop growth, and rain formation [see, e.g., (9, 52–55)]. Satellite-based remote sensing that uses microphysical and optical property measurements to measure cloud brightening and dimming [see, e.g., (15)] is our primary tool for monitoring aerosol-cloud interactions from the kilometer scale to the global scale. While global studies have focused on the climate forcing question, studies at the shipping lane (44, 45, 56) or even individual ship-track scale [see, e.g., (17, 57)] have addressed more microphysically oriented questions (Fig. 1). Analytical tools that follow individual ship tracks, even if they are hard to detect, are a recent addition to this line of

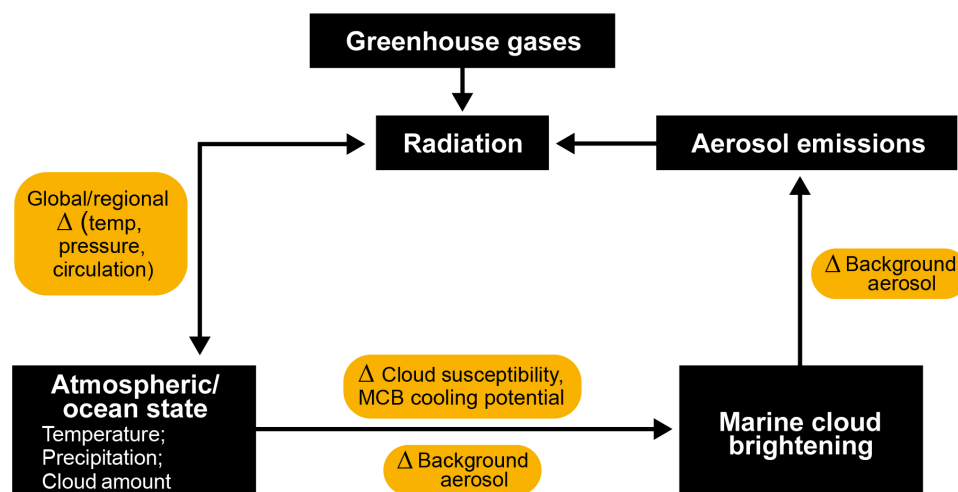


Fig. 3. How MCB fits into the atmospheric component of the Earth system. The MCB box subsumes processes and pathways as depicted in Figs. 1 and 2. Gold boxes indicate the changes (Δ) in key variables along the pathways indicated. MCB modifies the radiative effects of greenhouse gasses. Radiation influences and responds to the atmospheric/oceanic state (Δ in temperature, pressure, and circulation), which together with changes in the background aerosol, sets the stage for changes in cloud susceptibility and the potential for MCB cooling. The feedback loop between radiation and the atmospheric/oceanic state illustrates how MCB might influence regional temperature and precipitation patterns.

research (42, 58). However, detection of these signals against a highly variable regional background presents challenges [see, e.g., (59, 60)].

We, therefore, address the question of whether we could create a carefully designed MCB research program, using our current modeling and observational tools, to establish the technical viability and risks of augmenting aerosol-cloud cooling on a global scale through the application of marine injections and, if not, what would be needed to do so. The question becomes even more pertinent when one considers that, in the interests of public health, anthropogenic aerosol emissions have already been reduced in some regions and are projected to decrease in others in the coming decades (61). Current aerosol emission monitoring (62, 63) suggests less offsetting of warming associated with long-lived greenhouse gasses, exposing the planet and its inhabitants to enhanced climate change.

Thirty-one physical scientists working in the field of aerosol-cloud-radiation interactions gathered at a workshop in April 2022 to review critical issues, assess knowledge and knowledge gaps, and build on the roadmap for MCB research proposed by Diamond *et al.* (64). While the number of participants was limited for practical reasons, the group represented a wide range of skill sets, ideas, and viewpoints on MCB. The following is a distillation of the workshop deliberations and a recasting of some of the ideas addressed in the workshop report (65).

The contingencies

In the course of the workshop and the writing of the report, it became apparent that the technical feasibility of MCB is contingent on a number of factors, all of which translate to specific challenges, knowledge gaps, and research needs. As in (64), the structure of the following discussion tracks broadly with increasing spatial and temporal scale of the processes, with no ranking of their importance implied by their ordering.

Generation of particles with specified size distribution

The generated sea spray droplets injected into the atmosphere would need to be produced with a size distribution that increases cloud drop concentration substantially. This requires consideration of both the seeded particle size and the background aerosol size distribution. Moreover, one would need to avoid seeding with droplets that are small enough to have deleterious effects on the evaporation of cloud water (Fig. 2) (66), or giant cloud condensation nuclei (particles $>2\text{-}\mu\text{m}$ dry diameter) that tend to initiate rainfall (67)—both of which would act to reduce cloud brightness (66, 68).

Delivery of particles to cloud base

Sea spray aerosol particles generated by proposed MCB technology would need to be lofted up to the cloud base to affect cloud microphysics, which, for surface sprayers, would require marine boundary layers with strong enough vertical mixing to counteract the stabilization associated with the evaporating sea spray particles [see, e.g., (69)]. Alternative distribution methods using aircraft or fleets of UASs would provide more targeted delivery but present aerosol-generation challenges (70).

Microphysical-dynamical feedback (“cloud adjustments”)

Because the local adjustments in cloud cover or thickness in response to aerosol injections can be either positive or negative, potential losses in cloud water and cloud cover would have to be small enough for the seeded cloudy scene to be brighter. Alternatively, an ideal case for MCB deployment would be where seeding is effective at suppressing precipitation and increasing cloud amount (along with its ability to sustain itself through longwave radiative cooling), thus enhancing scene brightening even further (Fig. 2, blue arrows). However, microphysical-dynamical boundary layer feedback associated with entrainment and negative adjustments is more complex [see, e.g., (25)] and is, in general, poorly quantified. A visual example of a negative adjustment is the formation of dark (cloud-free) regions that sometimes flank ship tracks; these are associated with a circulation that is generated transverse to the track (18, 71).

Climatological susceptibility of clouds

Clouds with a high susceptibility to seeding would need to exist frequently enough, and with large enough spatial coverage, for local cooling to be globally relevant. Given the dependence of cloud brightening susceptibility on meteorological factors, background aerosol conditions, and meteorological-aerosol covariability, a clear understanding of how susceptibility varies over the diurnal and seasonal cycles in concert with meteorological and aerosol conditions would be essential (33, 72–74).

Detectability

The stratocumulus decks that are considered primary targets for MCB are characterized by high albedo variability (60). Recent sulfur shipping emissions (on the order of 10 Tg sulfate year^{−1} before 2020 regulations) (75) generate weak regional-scale albedo enhancement signals that require years to detect (44, 45, 60), a timescale that may be too long to allow for adjustments to seeding strategies—or for a cessation of activities if undesirable responses begin to emerge. Successful climate-relevant MCB would thus be contingent on the ability to detect its effect at timescales appropriate for decision-making (on the order of 1 year).

Global and regional impacts on temperature and precipitation

The impact of MCB on global temperature and precipitation patterns via its influence on sea surface temperatures carries the potential for serious risks that might arise as a result of unfavorable circulation responses (76, 77). This risk must be weighed against the potential risk of unfavorable circulation responses projected to occur in emission scenarios without MCB. There is no guarantee that the positive impacts of cooling the planet globally would outweigh potential negative regional impacts (78). Effective strategies would need to minimize negative regional impacts that may be far removed from seeding locations, particularly in sensitive ecosystems, and where human populations and other living organisms might be placed under stress (Fig. 3). The distinct possibility that some regions of the planet might benefit from climate intervention while others may suffer quickly enters the realm of governance, social science, and ethics. Although these are not the topics of this review, addressing these issues seriously is crucial to any climate intervention research activity [see, e.g., (2)].

A PATH FORWARD

Assessing the physical/technical feasibility of MCB and addressing the challenges outlined above require a greater understanding of MCB-specific aerosol-cloud-radiation interactions, boundary layer turbulence, cloud dynamics, and cloud-circulation coupling at the full range of atmospheric scales (64). Although these topics have been longstanding areas of inquiry within the atmospheric sciences, deliberate seeding carries some more specific challenges. Thus far, LES model simulations and analyses of cloud properties within ship tracks using in situ measurements and remote sensing retrievals have been the primary process-oriented tools for studying MCB [see, e.g., (71, 79, 80)], while climate models have addressed large-scale responses [see, e.g., (81–84)]. Below, we discuss specific actions to provide the tools and data needed to assess the physical science basis, approach, and potential consequences of MCB.

Figure 4 summarizes the elements of a comprehensive and coordinated research effort, which will include a considerable effort in multiscale modeling, with appropriate constraints from laboratory measurements and field observations. The effort will need to be both integrative and iterative. Satellite remote sensing and model reanalysis

products will play a key role in providing a regional-to-global perspective while in situ ship, aircraft, and UAS measurements will provide a more detailed view of aerosol and cloud microphysics, dynamics, and radiation. Modeling efforts will need to balance detailed representation of the small-scale processes, and cover large domains—up to global scales. Details are furnished in the discussion below. Ideas are synthesized thematically, and without prioritization.

Field and laboratory work

Establish a long-running, single-point emission perturbation experiment

This should be at a location where the baseline conditions are amenable to both observations and modeling. This would differ from earlier work that performed short-duration deliberate emission experiments (85). For example, one could envision a site on an island with minimal topography so as to reduce disruptions to the airflow. Such an experiment, complemented by routine LES modeling and ongoing observations from geostationary satellites, as well as in situ sampling by UASs, other airborne platforms, and surface-based instruments, will illuminate the effectiveness of MCB for a range of meteorological and cloud conditions at that location and with a well-defined aerosol source. It would also allow for testing of emissions of varying aerosol particle sizes and concentrations, composition, duration, and intensity, more analogous to proposed MCB approaches than ship emissions. Note, however, that a single-point source would create a distinct track and therefore differ from the suggested implementation of MCB injections in which routine spraying would create a higher sea salt concentration background, making individual fresh plumes less discernible.

Closure experiments

These represent a rigorous test of our understanding of how aerosol perturbations project onto essential aspects of marine boundary layer cloud systems—namely, changes in drop concentration and ultimately upward shortwave flux. To establish confidence in the fundamentals of aerosol growth to droplet sizes, one compares the observed drop concentrations to the drop concentrations calculated based on measurements of aerosol size distribution and composition, in concert with updraft velocities [see, e.g., (86–88)]. This so-called “activation” of an aerosol particle to form a droplet is defined as the point at which a nascent haze particle at equilibrium with its environment reaches a critical size large enough for it to grow freely as a droplet. Some of the larger aerosol particles will lag behind their equilibrium size and, strictly speaking, will not activate. Regardless, their haze sizes will be large enough for them to participate in cloud microphysical processes [see, e.g., (89)] and reflect sunlight. Droplet activation is driven by water vapor supersaturation, which is in turn a function of the local cloud updraft and aerosol particle sizes. For a given updraft, the competition for water vapor between ambient and seeded particles will determine just how many droplets are formed. The combination of parcel model calculations and detailed in situ observations allows for the evaluation of detailed size and composition effects on uncertainty in drop concentration enhancement (90). Equally critical will be closure studies that assess the degree to which aerosol and cloud microphysical properties produce calculated radiative properties similar to those measured directly by surface-based or space-borne sensors [see, e.g., (91, 92)]. These studies will help characterize the degree of brightening for different background aerosol, seeded aerosol, cloud, and meteorological conditions [see, e.g., (73)].

INTEGRATED VIEW OF MARINE CLOUD BRIGHTENING RESEARCH



Fig. 4. An integrated approach to a marine cloud brightening research program comprising laboratory facilities, field experiments, and modeling. Laboratory facilities such as cloud chambers together with observations at a range of scales will help improve the representation of physical processes in models. Parcel models, large eddy models, and cloud-resolving models will inform the global model activities to improve the reliability of regional climate responses. The Earth view image is courtesy of the European Organisation for the Exploitation of Meteorological Satellites (EUMETSAT).

Testing particle spraying technology from an ocean-based platform

The size distribution of injected particles will likely need to be optimized to avoid excessive evaporation of cloud water and/or precipitation formation. A perturbation experiment would provide the opportunity to test the ability of current nozzle designs to produce haze droplets of desired sizes in the marine environment (93).

New laboratory facilities

Such facilities, focused on aerosol and cloud microphysics will address gaps in process understanding. Models, particularly fine-scale models such as microphysically detailed parcel models and LES with embedded cloud parcel models (66), will benefit from improvements in our understanding of activation, droplet growth, entrainment, cloud processing of aerosol, onset of collision-coalescence, and the role of giant cloud condensation nuclei, all in a turbulent environment [see, e.g., (94)]. Few laboratory facilities capable of addressing all of these processes exist at this time. An envisioned cloud chamber facility called the Aerosol-Cloud-Drizzle Convection Chamber (ACDC2) is currently in design for possible future implementation (95). It is larger than the existing Michigan Tech II Convection-Cloud Chamber (96) and will extend droplet lifetimes, enabling the investigation of a wider range of liquid cloud processes that are relevant to MCB: activation, droplet growth by condensation, and the initiation of

collision-coalescence in a turbulent medium, as well as coupling these processes with the aerosol (97). Specific to MCB, cloud chamber experiments plus parcel modeling will lead to a better understanding of the interplay between the background aerosol population and the seeded particle sizes.

Opportunistic experiments

Our primary sources of aerosol-cloud data—in situ airborne, satellite-based remote sensing, and surface remote sensing—provide large volumes of data on MCB-target clouds, some of which include opportunistic seeding experiments associated with natural volcanic emissions, biomass burning, individual ship tracks, designated shipping lanes, urban point sources, and urban plumes (17, 98, 99). For shipping in particular, new regulations that took effect in 2020 have markedly reduced the sulfur content of marine fuels and have already led to notable declines in ship-track occurrence (35, 36) as well as changes in cloud microphysical properties within shipping corridors (45). This presents an opportunity for a better understanding of the changes in cloud response and their implications for MCB. Emissions from large volcanic eruptions have been shown to provide far more extensive regional perturbations to cloud fields than ship tracks, enabling a more robust test of the representation of aerosol-cloud-climate interactions within ESMs [see, e.g., (19, 100, 101)]. However, the scarcity in the observational record of such large-scale events

across a wide range of geographic and meteorological conditions means that, while useful, they are incomplete. In general, interpreting opportunistic experiments in the context of MCB requires taking into account anticipated disparities in aerosol size and composition, duration, and spatial extent. Nevertheless, these events offer important opportunities for observational and modeling studies from microphysical to global scales [see, e.g., (101)]. Dedicated field campaigns could be designed to sample opportunistic experiments using measurements and modeling tools similar to the single-point emission experiments described above. Being prepared to expand data collection in response to unpredictable events such as volcanic eruptions would position the community to collect valuable data without the long lead times typical of field campaigns.

Assessment of vertical mixing in a variety of conditions

A dedicated field experiment would ascertain the degree to which sea spray aerosol emitted by generators near the surface would reach the cloud base in a variety of conditions. Applying proposed real-world injection rates over extended periods and in different atmospheric conditions would provide an evaluation of how many of the generated particles reach clouds and whether evaporation of droplets in the lowest ~100 m might adversely influence the atmospheric mixing state and inhibit vertical transport (69, 102, 103).

Scaling up to global scales

The success of an MCB program depends on whether local cloud brightening will scale sufficiently to provide a globally relevant impact. Assessing the scalability of MCB requires a solid understanding of aerosol-meteorological co-variability, i.e., whether the natural covariation of background aerosol and meteorological conditions presents clouds that will brighten considerably if seeded. The susceptibility of a cloud to brightening depends on the background aerosol loading (if it is high already, brightening will be relatively ineffective) as well as on the meteorological conditions (more stable, shallow marine boundary layers tend to produce more susceptible clouds) (33). Assessing susceptibility over large areas relies heavily on meteorological reanalysis, satellite-based measurements of cloud macro- and microscale properties, and aerosol column measurements. Even considering the numerous field experiments that have taken place over oceans [see, e.g., (55, 104–107)], aerosol measurements in the remote marine environment are not nearly as well sampled in situ as over land. Taking into account geographical location, time of day, and season, the following would need to be assessed.

The robustness of susceptibility in target areas and the potential for brightening

Do conditions favoring susceptible clouds occur frequently enough and over large enough areas to generate cooling of sufficient magnitude? How many of the world's stratocumulus decks would represent viable targets? Do these target areas behave similarly in terms of their ability to generate cooling if seeded? At what time of day and during what season do these targets present themselves for a given stratocumulus deck? Should we consider targeting the extratropical oceans, an area where studies seem to indicate positive liquid water path adjustments (101, 108), or perhaps even Arctic clouds over the open ocean to slow the dangerous positive feedback involving greenhouse gas warming, sea ice, and clouds [see, e.g., (109)]? A globally applied metric based on the product of brightening potential and frequency of occurrence of cloud cover has been proposed [see, e.g., (73)]; other metrics should be explored.

Predictability of liquid water and cloud fraction adjustments and their timescales

Liquid water and cloud fraction adjustments have the potential to determine whether aerosol injections will create brighter or darker cloudy scenes. Aerosol perturbations brighten clouds within 10 min of particles entering the cloud, but the clouds then adjust to these perturbations with a timescale of roughly 1 day as the effects of the initial perturbation propagate through the cloudy boundary layer system (28, 110, 111). The delay in adjustment could have a substantial impact on the offsetting or enhancement of the original perturbation, as brightening only matters during daylight hours and is more effective at small solar zenith angles. Solar absorption is also highest near solar noon and depends on cloud amount as well as cloud drop concentration, factors which also affect adjustment and its timescales (102). Quantifying these timescales is essential.

Amounts (mass of material), size distribution, durations, and intervals for maximum impact of seeding

Very little research exists on the topic of matching seeding amounts and strategies to desired cooling. Largely, this is because quantitative observations of cloud response to ambient marine boundary layer aerosol are lacking. Perturbation studies of the kind discussed above will provide important insights (112).

Modeling at a range of scales

This effort, focused on different research objectives will play a number of important roles in assessing the scale-up. Routine LES modeling of a broad range of real case studies (on the order of 30 per season, per location) at the mesoscale has proven very useful in other settings [see, e.g., (113)]. Initializing LES models with observed initial and boundary conditions (e.g., soundings and surface fluxes) and then comparing the simulated cloud fields to observations for a range of commonly occurring meteorological conditions will improve our confidence in the ability of LES models to reproduce observed cloud fields and their susceptibility to aerosol. Multi-model ensembles will further establish robustness. The relative dearth of surface in situ and surface remote sensing measurements in the remote marine environment will require heavier reliance on reanalysis, ship-based soundings, and satellite retrievals of cloud microphysics, amount, and radiative fluxes.

The need to obtain a robust understanding of responses in global circulation in the context of future climate scenarios is key to MCB deployment. Circulation responses and attendant changes in cloudiness, especially at the spatial resolution of nation states and ecosystems, are uncertain and occur against the background of an uncertain response to anthropogenic greenhouse gases (77). At present, studies evaluating the impacts of MCB on regional climate are limited in scope relative to the rigorous analysis performed to characterize other future scenarios. The implications of this knowledge gap are enormous, not only because of physical science uncertainties but also because of deep concerns about environmental justice that would arise from the inequitable distribution of benefits and risks. For example, the global simulations of Jones *et al.* (81) point to a sharp decrease in precipitation in the Amazon in response to simulated MCB, when seeding occurs in the South Atlantic, leading to a strong localized cooling in the area. These teleconnections between South Atlantic sea surface temperatures and precipitation over northeastern Brazil have been documented in observational analyses (114). Subsequent studies using different models find weaker or negligible drying responses in this region [see, e.g., (83,

115)] suggesting substantial inter-model uncertainty [reviewed in (76)]. Fasullo *et al.* (116) suggest, based on climate modeling studies, that smoke from the 2019/2020 wildfire season in eastern Australia brightened the clouds in the southeastern Pacific and cooled the ocean surface enough to contribute to the establishment of the unusually persistent La Niña event of 2020–2023 [see also (83)].

The Geoengineering Model Intercomparison Project (GeoMIP) climate model intercomparison efforts [see, e.g., (82)] to evaluate inter-model consistency will be valuable as a framework for exploring scenarios that apply idealized albedo perturbations in different target regions to test the robustness of circulation changes. Assessment of differential regional responses to MCB with respect to variables such as temperature, precipitation, water availability, and crop yields are of particular interest. Parallel intercomparison projects of the “no MCB in a warming world” would provide important context for what might happen without an intervention (“a risk-risk assessment”). Expanding the range of intercomparison projects would further help to resolve the provenance of model differences. Models using regionally refined meshes over areas of interest (117) or multimodeling/multiscale frameworks (51) may help to resolve the aerosol-cloud dynamical processes and spatial granularity of climate responses and associated risks to populations (particularly coastal versus inland communities) either within or outside the target seeding or climate impact region.

Detection of MCB

Any MCB program will need systems in place to detect the degree of cloud brightening from space and reduction in downwelling short-wave radiation at the surface. A small number of studies have indicated that perturbations at scales on the order of 100 km in shipping lanes would require several years to detect with existing satellite-based systems (44, 45), depending on the magnitude of the perturbation relative to the natural variability in cloud albedo (60). Speeding up this process is crucial to determining whether MCB is working as intended or whether it might need to be modified to achieve different goals. Approaches include the following.

Refined algorithms or instruments

These are required for retrieving important geophysical variables from space, particularly from geostationary platforms, and leveraging the complementary advantages of polar-orbiting and geostationary satellite measurements. Machine learning is increasingly being used to enhance the quality of such products—e.g., the retrieval of cloud optical depth in broken cloud fields (118) and aerosol optical depth in the aerosol-cloud continuum region close to clouds (119). Commensurate with cloud brightening detected by space-borne sensors, a decrease in downwelling solar radiation (dimming) could be identified by surface-based sensors (120–123).

Undetected tracks

Assessment of the contribution of undetected tracks to brightening has recently been done indirectly, i.e., by tracking the particle source over the duration of the perturbation rather than searching for a weak signal in a noisy background (42). Using this approach, detection of in- versus out-of-track albedo changes can be achieved within months; however, because of the inherently weak signal-to-noise ratio in such situations, detection times are still likely to be multiple years over larger regions, and in- versus out-of-track comparisons may misrepresent adjustments due to the generation of secondary circulation [see, e.g., (71)]. A lack of access to ship position data at a reasonable cost also limits our ability to research undetected tracks.

Other detection methods

Detection methods that leverage knowledge of the seeding plume location over time, such as a passive tracer co-emitted with the seeding agent, a unique pattern of dispersal, and the frequency/intensity of dispersal, might also improve detectability.

STRONG CONSENSUS

In discussions of the research program outlined above, one unifying theme stood out: Routine field activities tightly coordinated with modeling and satellite remote sensing to test critical components of proposed MCB activities. Such an effort would cover a range of activities: particle generation and delivery to clouds, local detection of cloud brightening, regional responses, and global assessments of radiative forcing. At every step of the way, experimental work would be complemented by modeling activities to test all aspects of our understanding of the system. The scope of the problem is broad-sweeping: It requires everything from particle generator development to routine field experiments in a variety of conditions to improvements in the representation of the modeling of the system at a range of scales to enhancement in detection. The research would address what we consider the most serious physical risk, i.e., that circulation changes induced by an MCB intervention could induce heterogeneous regional temperature and precipitation responses that inequitably impact populations and put sensitive ecosystems at risk. Many of the practical aspects are laid out above and in the workshop report (65). Most require sustained decadal-scale support—particularly for routine experimentation and model development. The benefits would be commensurate: The effort would serve both the goals of understanding the viability of MCB, as well as the broader goal of reducing uncertainties in the radiative forcing of aerosol-cloud interactions.

OTHER CONSIDERATIONS

Optimization with a bespoke approach

As noted, clouds are not all created equal—some are more susceptible to aerosol injections than others, depending on their albedo and the meteorological and background aerosol conditions. To the extent that optimal brightening conditions can be robustly established (73), a bespoke approach to MCB, rather than routine spraying under all conditions, might have a higher probability of a more desirable outcome, with the caveat that logistical challenges could increase considerably. More specifically, a bespoke approach will need to address many of the contingencies addressed above as well as those identified in a comprehensive research program. The four key aspects follow.

Optimizing seeded size distributions

There is no single optimal size and seeding rate for all cloud conditions (66). Establishing how to optimally match seeded particle size, injection rates, and durations to ambient aerosol and cloud conditions is very important.

Delivery to cloud

Shallower boundary layers tend to provide opportunities for more efficient mixing of particles into the cloud and more effective brightening. Nevertheless, deep boundary layers with stratiform low cloud coupled locally by cumulus convection may benefit appreciably from seeding: Cumulus detrainment into adjacent open cells has been demonstrated to accelerate the transition to a high cloud fraction, reflective state (124). Conversely, deeper boundary layers tend to have stronger

negative liquid water adjustments, except perhaps in the case of “invisible” tracks (42). It is important to establish the pros and cons of seeding shallower versus deeper boundary layers [see, e.g., (43, 73, 125)].

Targeting the more susceptible clouds

Liquid water and cloud fraction adjustments to aerosol injections have a strong impact on whether cloud fields become brighter. A concerted effort to understand the relationship between the meteorological/aerosol state and these adjustments is at the heart of the optimization of MCB. For example, MCB might be optimized by seeding during particular times of the day (e.g., in the early morning hours before precipitation) (79, 126), and during specific seasons when meteorological and aerosol patterns indicate a higher probability of susceptible target clouds. Ultimately, any targeting of clouds based on their susceptibility would need to consider the balance of susceptibility, frequency of occurrence, and spatial coverage. For example, one might define optimal conditions in a way that includes less-susceptible clouds, provided such clouds occur frequently enough and with sufficient coverage. Note that given the 2- to 3-day lifetime of particles in the marine boundary layer, the required spatial coverage and frequency of MCB injections might create a background haze that will probably make it harder to optimize the timing and location. If such optimization were to be pursued, it would likely rely on a sophisticated real-time optimization system that would use observations, modeling, and forecasting to identify the most susceptible target clouds.

Understanding regional circulation responses

As climate models evolve, and the representation of low clouds and aerosol-cloud interactions in climate models improves, it is imperative that we continue to reevaluate our understanding of MCB impacts on regional circulation patterns and concomitant changes in temperature and precipitation patterns [see, e.g., (83, 116)]. A reliable climate modeling framework will go a long way toward assisting with the optimization of choosing where (location), when (season and diurnal cycle), how much (injected mass for given areal coverage/frequency of occurrence), and for how long to inject to achieve a desirable outcome. The effects of a variable (in time and space) seeding pattern on larger-scale circulation changes, based on optimal conditions rather than persistent seeding in prescribed regions, are unknown, but we speculate that these might mitigate larger-scale uneven circulation responses associated with continuous or continual seeding in the same location. This, in turn, might reduce the probability of spatiotemporal variability in benefits and risks.

If the risks associated with regional circulation responses could be adequately resolved, then a more thoughtful, targeted, and scientifically deliberate approach to seeding might be considered. The outcome of such a bespoke MCB effort might potentially determine the viability of successfully brightening clouds, with the added benefit of doing so in a more efficient manner. The risk, however, is that as more of the contingencies are addressed through strategies that are tailored for particular conditions, the overall coverage of target clouds would diminish, the complexity of implementation would increase, and detection might be more challenging. Here too, the cost-benefit analysis would require a reliable modeling framework, backed by routine satellite-based monitoring of clouds, meteorological reanalysis, and process-based cloud modeling of real-world, well-observed cases.

Marine sky brightening

While not discussed in the workshop, for completeness, we address the relatively unexplored but related idea of marine sky brightening

(MSB), which aims to increase the albedo over oceans via the direct scattering of haze droplets [see, e.g., (126, 127)]. MSB might be considered either a deliberate targeting of clear skies over the oceans, for example, in regions that are home to sensitive corals (37), or as a salutary byproduct of MCB: Particles emitted inadvertently into clear skies or not effectively lofted into partially cloudy skies provide an alternate means of generating a more reflective surface than the darker underlying ocean (128). We note that by not deliberately targeting the clouds themselves, MSB would suffer from fewer of the challenges and uncertainties of MCB—particularly those related to the delivery of haze particles to clouds, cloud susceptibility, and liquid water and cloud amount adjustments. MSB still needs to grapple with haze droplet generation at sizes that maximize light scattering while at the same time contending with sedimentation. The much greater mass of salt required for larger particles would substantially increase the energy cost of spraying (93) and may make a larger-scale deployment impractical. Last, it is also unclear how effective MSB would be at global scales and whether regional MSB-related cooling might affect the general circulation.

Effective communication

On a practical note, the interaction of scientists working on different scales, in different disciplines, and with different approaches will require a common language for MCB, with consistent (and possibly refined) definitions of susceptibility, cloud regimes, and microphysical quantities, among others. In this document, we have consistently defined susceptibility as a change in reflectance for an incremental change in aerosol but other susceptibility metrics such as the change in drop concentration per change in aerosol concentration exist. Unifying and/or modifying the current aerosol-cloud interaction vocabulary will help ensure that the scientific community provides clear messages to decision-makers and the public.

More broadly, communication of MCB should be prefaced by emphasizing that there exist no scenarios in which MCB would replace decarbonization. As in (2), the establishment of a transdisciplinary SRM research program should be considered to be conditional on the simultaneous commitment to decarbonization. Moreover, the value of MCB should be communicated relative to the warming at the same level of decarbonization but without MCB.

SUMMARY

MCB is one of the primary proposed SRM approaches to enhancing the reflectance of incoming solar radiation to space by seeding marine boundary layer clouds. This review considers the physical science knowledge gaps, viability, and risks associated with MCB. It builds on discussions at a recent workshop attended by 31 international scientists to assess the state of knowledge in the field of MCB and to assess research needs toward reducing unknowns in key components of physical science.

Progress in assessing the viability of MCB depends on our ability to address knowledge gaps at a range of spatial and temporal scales concerning aerosol and cloud microphysics, local and large-scale adjustments, aerosol-meteorology covariability, and detection of perturbations and their radiative effect. A consensus viewpoint is that addressing these knowledge gaps would require routine in situ field measurements tightly coordinated with modeling and satellite remote sensing to test critical components of proposed MCB activities. The effort would cover activities across a large range of scales

and disciplines encompassing particle generation and delivery to clouds, local detection of cloud brightening, regional responses, and global assessments of radiative forcing. An integration of experimental work and modeling activities would test all aspects of our understanding of the system. It would focus our attention on what we consider the most serious physical risk associated with MCB, i.e., that circulation changes induced by an MCB intervention could induce heterogeneous regional temperature and precipitation responses that inequitably affect populations and put sensitive ecosystems at risk.

A sustained, substantial, and targeted program of research is required to address major elements of this approach: continued study of natural analogs (e.g., effusive volcanoes and ship tracks), long-duration controlled point source emissions, focused field campaigns applying controlled perturbations, laboratory studies, routine modeling of aerosol-cloud systems at the large eddy scale, model intercomparison efforts across the full range of scales, and analysis of existing observations in the context of reanalysis model data. Many of the knowledge gaps associated with MCB exist in areas of study already familiar to the broader field of climate forcing by aerosol-cloud interactions, and the path forward is clearer though challenging (129). Some, like particle generation and delivery to the cloud, include more specific but seemingly manageable MCB challenges.

Two key themes stand out as particularly challenging: the first is the lack of a clear understanding of the relationship between aerosol and meteorological conditions and liquid water and cloud fraction adjustments and their timescales. These adjustments exert a strong control over whether aerosol injections will brighten cloudy scenes. The second is the need for models that can robustly simulate the influence of local/regional MCB on global circulation patterns. As noted, changes in these circulation patterns have the potential to create regions of the world that benefit from MCB as well as others that might suffer. Regional changes in temperature and rainfall could influence heat stress, water availability, crop productivity (130), and the ability of communities to thrive. Representing such regional responses in global climate models requires a comprehensive and coordinated effort of multiscale modeling, with appropriate constraints from laboratory measurements and field observations as part of an iterative and integrated research program (Fig. 4).

An understanding of these large-scale manifestations of MCB requires modeling tools that do not currently exist. Given the ethical ramifications, models must be able to provide reliable projections of shifts in circulation patterns and accompanying changes in temperature and precipitation before an active MCB program is undertaken. Achieving such advances in models requires expanded in situ observations to constrain and differentiate model processes at small scales, and microphysical specificity that is not retrievable from remote sensing.

This paper has raised the notion of a bespoke approach to MCB implementation, aimed at targeting the more susceptible clouds at locations, times of the day, synoptic periods, and seasons when satellite remote sensing suggests the highest impact. A thoughtful, scientifically deliberate approach like this is appealing. It might mean a higher likelihood of local success if the challenges can be overcome, and possibly improved detection. However, it would come at the expense of reduced spatial and temporal coverage. A bespoke approach will be more difficult to implement since it will require advance notice of meteorological and background aerosol conditions and adjustments to, for example, nozzles and seeding locations. If it were to

be adopted, we speculate that the effects of a variable seeding pattern (in time and space) based on optimal conditions might mitigate the negative impacts on larger-scale circulation associated with continuous or continual seeding in the same location [see, e.g., (81)]. In turn, this might have less chance of creating a strong spatial imprint of response and attendant risks of undesirable outcomes. A rigorous MCB model intercomparison would be required to establish the robustness of outcomes. Understanding the trade-offs will be a considerable challenge.

This paper has focused on the physical science knowledge gaps in assessing the viability and risks of MCB. We reiterate the message in the NASEM Report (2) that the viability of MCB will depend on resolving both the physical science questions as well as the broader social, ethical, ecological, economic, public health, and governance aspects of the problem and that MCB will be ineffective without the simultaneous commitment to decarbonization.

REFERENCES AND NOTES

1. United Nations Environment Programme, "Emissions Gap Report 2022: The Closing Window—Climate Crisis Calls for Rapid Transformation of Societies" (United Nations Environment Programme, 2022); www.unep.org/emissions-gap-report-2022.
2. National Academies of Sciences, Engineering, and Medicine, *Reflecting Sunlight: Recommendations for Solar Geoengineering Research and Research Governance* (National Academies of Sciences, Engineering, and Medicine, 2021).
3. Office of Science and Technology Policy, *Congressionally Mandated Research Plan and an Initial Research Governance Framework Related to Solar Radiation Modification* (Office of Science and Technology Policy, 2023); www.whitehouse.gov/wp-content/uploads/2023/06/Congressionally-Mandated-Report-on-Solar-Radiation-Modification.pdf.
4. United Nations Environment Programme, *One Atmosphere: An Independent Expert Review on Solar Radiation Modification Research and Deployment*. Kenya, Nairobi (United Nations Environment Programme, 2023); <https://wedocs.unep.org/handle/20.500.11822/41903>.
5. M. G. Lawrence, S. Schäfer, H. Muri, V. Scott, A. Oeschies, N. E. Vaughan, O. Boucher, H. Schmidt, J. Haywood, J. Scheffran, Evaluating climate geoengineering proposals in the context of the Paris agreement temperature goals. *Nat. Commun.* **9**, 3734 (2018).
6. J. Latham, Control of global warming? *Nature* **347**, 339–340 (1990).
7. J. H. Conover, Anomalous cloud lines. *J. Atmos. Sci.* **23**, 778–785 (1966).
8. S. Twomey, Pollution and the planetary albedo. *Atmos. Environ.* **8**, 1251–1256 (1974).
9. P. A. Durkee, K. J. Noone, R. T. Bluth, The Monterey area ship track experiment. *J. Atmos. Sci.* **57**, 2523–2541 (2000).
10. P. V. Hobbs, T. J. Garrett, R. J. Ferek, S. R. Strader, D. A. Hegg, G. M. Frick, W. A. Hoppel, R. F. Gasparovic, L. M. Russell, D. W. Johnson, C. O'Dowd, P. A. Durkee, K. E. Nielsen, G. Innis, Emissions from ships with respect to their effects on clouds. *J. Atmos. Sci.* **57**, 2570–2590 (2000).
11. A. Robock, D. G. MacMartin, R. Duren, M. W. Christensen, Studying geoengineering with natural and anthropogenic analogs. *Clim. Change* **121**, 445–458 (2013).
12. Y.-C. Chen, M. W. Christensen, L. Xue, A. Sorooshian, G. L. Stephens, R. M. Rasmussen, J. H. Seinfeld, Occurrence of lower cloud albedo in ship tracks. *Atmos. Chem. Phys.* **12**, 8223–8235 (2012).
13. J. A. Coakley, C. D. Walsh, Limits to the aerosol indirect radiative effect derived from observations of ship tracks. *J. Atmos. Sci.* **59**, 668–680 (2002).
14. M. D. Lebsock, G. L. Stephens, C. Kummerow, Multisensor satellite observations of aerosol effects on warm clouds. *J. Geophys. Res.* **113**, D15205 (2008).
15. E. Gryspeerdt, T. Goren, O. Sourdeval, J. Quaas, J. Mülmenstädt, S. Dipu, C. Unglaub, A. Gettelman, M. Christensen, Constraining the aerosol influence on cloud liquid water path. *Atmos. Chem. Phys.* **19**, 5331–5347 (2019).
16. Q. Han, W. B. Rossow, J. Zeng, R. Welch, Three different behaviors of liquid water path of water clouds in aerosol–cloud interactions. *J. Atmos. Sci.* **59**, 726–735 (2002).
17. V. Toll, M. Christensen, J. Quaas, N. Bellouin, Weak average liquid-cloud-water response to anthropogenic aerosols. *Nature* **572**, 51–55 (2019).
18. W. M. Porch, C.-Y. J. Kao, R. G. Kelley, Ship trails and ship induced cloud dynamics. *Atmos. Environ. A Gen.* **24**, 1051–1059 (1990).
19. Y. Chen, J. Haywood, Y. Wang, F. Malavelle, G. Jordan, D. Partridge, J. Fieldsend, J. De Leeuw, A. Schmidt, N. Cho, L. Oreopoulos, S. Platnick, D. Grosvenor, P. Field, U. Lohmann, Machine learning reveals climate forcing from aerosols is dominated by increased cloud cover. *Nat. Geosci.* **15**, 609–614. (2022).

20. M. W. Christensen, W. K. Jones, P. Stier, Aerosols enhance cloud lifetime and brightness along the stratus-to-cumulus transition. *Proc. Natl. Acad. Sci. U.S.A.* **117**, 17591–17598 (2020).
21. T. Goren, D. Rosenfeld, Satellite observations of ship emission induced transitions from broken to closed cell marine stratocumulus over large areas. *J. Geophys. Res. Atmos.* **117**, D17206 (2012).
22. B. A. Albrecht, Aerosols, cloud microphysics, and fractional cloudiness. *Science* **245**, 1227–1230 (1989).
23. S. Wang, Q. Wang, G. Feingold, Turbulence, condensation, and liquid water transport in numerically simulated nonprecipitating stratocumulus clouds. *J. Atmos. Sci.* **60**, 262–278 (2003).
24. A. S. Ackerman, M. P. Kirkpatrick, D. E. Stevens, O. B. Toon, The impact of humidity above stratiform clouds on indirect aerosol climate forcing. *Nature* **432**, 1014–1017 (2004).
25. C. S. Bretherton, P. N. Blossey, J. Uchida, Cloud droplet sedimentation, entrainment efficiency, and subtropical stratocumulus albedo. *Geophys. Res. Lett.* **34**, L03813 (2007).
26. R. Wood, Cancellation of aerosol indirect effects in marine stratocumulus through cloud thinning. *J. Atmos. Sci.* **64**, 2657–2669 (2007).
27. H. Xue, G. Feingold, B. Stevens, Aerosol effects on clouds, precipitation, and the organization of shallow cumulus convection. *J. Atmos. Sci.* **65**, 392–406 (2008).
28. F. Glassmeier, F. Hoffmann, J. S. Johnson, T. Yamaguchi, K. S. Carslaw, G. Feingold, Aerosol-cloud-climate cooling overestimated by ship-track data. *Science* **371**, 485–489 (2021).
29. J.-Y. Chun, R. Wood, P. Blossey, S. J. Doherty, Microphysical, macrophysical, and radiative responses of subtropical marine clouds to aerosol injections. *Atmos. Chem. Phys.* **23**, 1345–1368 (2023).
30. P. Forster, T. Storelvmo, K. Armour, W. Collins, J.-L. Dufresne, D. Frame, D. J. Lunt, T. Mauritsen, M. D. Palmer, M. Watanabe, M. Wild, and H. Zhang, The Earth's energy budget, climate feedbacks, and climate sensitivity, in *Climate Change 2021: The Physical Science Basis. Contribution of Working Group I to the Sixth Assessment Report of the Intergovernmental Panel on Climate Change*, V. Masson-Delmotte, P. Zhai, A. Pirani, S. L. Connors, C. Péan, S. Berger, N. Caud, Y. Chen, L. Goldfarb, M. I. Gomis, M. Huang, K. Leitzell, E. Lonnoy, J. B. R. Matthews, T. K. Maycock, T. Waterfield, O. Yelekçi, R. Yu, B. Zhou, Eds. (Cambridge Univ. Press, 2021), pp. 923–1054.
31. S. A. Klein, D. L. Hartmann, The seasonal cycle of low stratiform clouds. *J. Climate* **6**, 1588–1606 (1993).
32. R. Eastman, S. G. Warren, Diurnal cycles of cumulus, cumulonimbus, stratus, stratocumulus, and fog from surface observations over land and ocean. *J. Climate* **27**, 2386–2404 (2014).
33. J. Zhang, G. Feingold, Distinct regional meteorological influences on low-cloud albedo susceptibility over global marine stratocumulus regions. *Atmos. Chem. Phys.* **23**, 1073–1090 (2023).
34. J. A. Coakley, P. A. Durkee, K. Nielsen, J. P. Taylor, S. Platnick, B. A. Albrecht, D. Babb, F.-L. Chang, W. R. Tahnk, C. S. Bretherton, P. V. Hobbs, The appearance and disappearance of ship tracks on large spatial scales. *J. Atmos. Sci.* **57**, 2765–2778 (2000).
35. T. Yuan, H. Song, R. Wood, C. Wang, L. Oreopoulos, S. E. Platnick, S. von Hippel, K. Meyer, S. Light, E. Wilcox, Global reduction in ship-tracks from sulfur regulations for shipping fuel. *Sci. Adv.* **8**, eabn7988 (2022).
36. D. Watson-Parris, M. W. Christensen, A. Laurenson, D. Clewley, E. Grypsperdt, P. Stier, Shipping regulations lead to large reduction in cloud perturbations. *Proc. Natl. Acad. Sci. U.S.A.* **119**, e2206885119 (2022).
37. S. A. Condie, K. R. N. Anthony, R. C. Babcock, M. E. Baird, R. Beeden, C. S. Fletcher, R. Gorton, D. Harrison, A. J. Hobday, É. E. Plagányi, D. A. Westcott, Large-scale interventions may delay decline of the Great Barrier Reef. *R. Soc. Open Sci.* **8**, 201296 (2021).
38. J. Latham, J. Kleypas, R. Hauser, B. Parkes, A. Gadian, Can marine cloud brightening reduce coral bleaching? *Atmos. Sci. Lett.* **14**, 214–219 (2013).
39. J. Latham, A. Gadian, J. Fournier, B. Parkes, P. Wadhams, J. Chen, Marine cloud brightening: Regional applications. *Philos. Trans. A Math Phys. Eng. Sci.* **A 372**, 20140053 (2014).
40. N. Bellouin, J. Quaas, E. Grypsperdt, S. Kinne, P. Stier, D. Watson-Parris, O. Boucher, K. S. Carslaw, M. Christensen, A.-L. Daniau, J.-L. Dufresne, G. Feingold, S. Fiedler, P. Forster, A. Gettelman, J. M. Haywood, F. Malavelle, U. Lohmann, T. Mauritsen, D. T. McCoy, G. Myhre, J. Mulmenstadt, D. Neubauer, A. Possner, M. Rugenstein, Y. Sato, M. Schulz, S. E. Schwartz, O. Sourdeval, T. Storelvmo, V. Toll, D. Winker, B. Stevens, Bounding global aerosol radiative forcing of climate change. *Rev. Geophys.* **58**, e2019RG000660 (2020).
41. M. Schreier, H. Mannstein, V. Eyring, H. Bovensmann, Global ship track distribution and radiative forcing from 1 year of AATSR data. *Geophys. Res. Lett.* **34**, L17814 (2007).
42. P. Manshausen, D. Watson-Parris, M. W. Christensen, J.-P. Jalkanen, P. Stier, Invisible ship tracks show large cloud sensitivity to aerosol. *Nature* **610**, 101–106 (2022).
43. A. Possner, H. Wang, R. Wood, K. Caldeira, T. P. Ackerman, The efficacy of aerosol–cloud radiative perturbations from near-surface emissions in deep open-cell stratocumuli. *Atmos. Chem. Phys.* **18**, 17475–17488 (2018).
44. M. S. Diamond, H. M. Director, R. Eastman, A. Possner, R. Wood, Substantial cloud brightening from shipping in subtropical low clouds. *AGU Adv.* **1**, e2019AV000111 (2020).
45. M. S. Diamond, Detection of large-scale cloud microphysical changes within a major shipping corridor after implementation of the International Maritime Organization 2020 fuel sulfur regulations. *Atmos. Chem. Phys.* **23**, 8259–8269 (2023).
46. K. Capaldo, J. J. Corbett, P. Kasibhatla, P. Fischbeck, S. N. Pandis, Effects of ship emissions on sulphur cycling and radiative climate forcing over the ocean. *Nature* **400**, 743–746 (1999).
47. A. Lauer, V. Eyring, J. Hendricks, P. Jöckel, U. Lohmann, Global model simulations of the impact of ocean-going ships on aerosols, clouds, and the radiation budget. *Atmos. Chem. Phys.* **7**, 5061–5079 (2007).
48. K. Peters, P. Stier, J. Quaas, H. Graßl, Corrigendum to “Aerosol indirect effects from shipping emissions: Sensitivity studies with the global aerosol-climate model ECHAM-HAM” published in *Atmos. Chem. Phys.*, **12**, 5985–6007, 2012. *Atmos. Chem. Phys.* **13**, 6429–6430 (2013).
49. M. Righi, C. Klinger, V. Eyring, J. Hendricks, A. Lauer, A. Petzold, Climate impact of biofuels in shipping: Global model studies of the aerosol indirect effect. *Environ. Sci. Technol.* **45**, 3519–3525 (2011).
50. M. Sofiev, J. J. Winebrake, L. Johansson, E. W. Carr, M. Prank, J. Soares, J. Vira, R. Kouznetsov, J.-P. Jalkanen, J. J. Corbett, Cleaner fuels for ships provide public health benefits with climate tradeoffs. *Nat. Commun.* **9**, 406 (2018).
51. C. R. Terai, M. S. Pritchard, P. Blossey, C. S. Bretherton, The impact of resolving simulations. *J. Adv. Model. Earth Syst.* **12**, e2020MS002274 (2020).
52. S. Twomey, J. Warner, Comparison of measurements of cloud droplets and cloud nuclei. *J. Atmos. Sci.* **24**, 702–703 (1967).
53. L. F. Radke, J. A. Coakley, M. D. King, Direct and remote sensing observations of the effects of ships on clouds. *Science* **246**, 1146–1149 (1989).
54. C. Fountoukis, A. Nenes, N. Meskhidze, R. Bahreini, W. C. Conant, H. Jonsson, S. Murphy, A. Sorooshian, V. Varutbangkul, F. Brechtel, R. C. Flagan, J. H. Seinfeld, Aerosol–cloud drop concentration closure for clouds sampled during the International Consortium for Atmospheric Research on Transport and Transformation 2004 campaign. *J. Geophys. Res.* **112**, D10530 (2007).
55. J. Wang, R. Wood, M. P. Jensen, J. C. Chiu, Y. Liu, K. Lamer, N. Desai, S. E. Giangrande, D. A. Knopf, P. Kollias, A. Laskin, X. Liu, C. Lu, D. Mechem, F. Mei, M. Starzec, J. Tomlinson, Y. Wang, S.-S. Yum, G. Zheng, A. C. Aiken, E. B. Azevedo, Y. Blanchard, S. China, X. Dong, F. Gallo, S. Gao, V. P. Ghatge, S. Glienke, L. Goldberger, J. C. Hardin, C. Kuang, E. P. Luke, A. A. Matthews, M. A. Miller, R. Moffet, M. Pekour, B. Schmid, A. J. Sedlacek, R. A. Shaw, J. E. Shilling, A. Sullivan, K. Suski, D. P. Veghte, R. Weber, M. Wyant, J. Yeom, M. Zawadowicz, Z. Zhang, Aerosol and cloud experiments in the Eastern North Atlantic (ACE-ENA). *Bull. Amer. Meteor. Soc.* **103**, E619–E641 (2022).
56. S. Hu, Y. Zhu, D. Rosenfeld, F. Mao, X. Lu, Z. Pan, L. Zang, W. Gong, The dependence of ship-polluted marine cloud properties and radiative forcing on background drop concentrations. *J. Geophys. Res. Atmos.* **126**, e2020JD033852 (2021).
57. M. W. Christensen, J. A. Coakley Jr., W. R. Tahnk, Morning-to-afternoon evolution of marine stratus polluted by underlying ships: Implications for the relative lifetimes of polluted and unpolluted clouds. *J. Atmos. Sci.* **66**, 2097–2106 (2009).
58. E. Grypsperdt, T. W. P. Smith, E. O’Keeffe, M. W. Christensen, F. W. Goldsworth, The impact of ship emission controls recorded by cloud properties. *Geophys. Res. Lett.* **46**, 12547–12555 (2019).
59. K. Peters, J. Quaas, P. Stier, H. Graßl, Processes limiting the emergence of detectable aerosol indirect effects on tropical warm clouds in global aerosol-climate model and satellite data. *Tellus B Chem. Phys. Meteorol.* **66**, 24054 (2022).
60. D. J. Seidel, G. Feingold, A. R. Jacobson, N. Loeb, Detection limits of albedo changes induced by climate engineering. *Nat. Clim. Change* **4**, 93–98 (2014).
61. J. Quaas, H. Jia, C. Smith, A. L. Albright, W. Aas, N. Bellouin, O. Boucher, M. Doutriaux-Boucher, P. Forster, D. P. Grosvenor, S. Jenkins, Z. Klimont, N. G. Loeb, X. Ma, V. Naik, F. Paulot, P. Stier, M. Wild, G. Myhre, M. Schulz, Robust evidence for reversal of the trend in aerosol effective climate forcing. *Atmos. Chem. Phys.* **22**, 12221–12239 (2022).
62. Z. Liu, Z. Deng, S. Davis, P. Ciais, Monitoring global carbon emissions in 2022. *Nat. Rev. Earth Environ.* **4**, 205–206 (2023).
63. J. Rogelj, P. M. Forster, E. Kriegler, C. J. Smith, R. Séférian, Estimating and tracking the remaining carbon budget for stringent climate targets. *Nature* **571**, 335–342 (2019).
64. M. S. Diamond, A. Gettelman, M. D. Lebsock, A. McComiskey, L. M. Russell, R. Wood, G. Feingold, Opinion: To assess marine cloud brightening's technical feasibility, we need to know what to study—and when to stop. *Proc. Natl. Acad. Sci. U.S.A.* **119**, e2118379119 (2022).
65. G. Feingold, V. P. Ghatge, L. M. Russell, “DOE-NOAA Marine Cloud Brightening Workshop” (no. DOE/SC-0207; OAR-ESRL/CSL-1, Oak Ridge National Laboratory, Oak Ridge, TN, Atmospheric Radiation Measurement (ARM) Data Center, 2022).
66. F. Hoffmann, G. Feingold, Cloud microphysical implications for marine cloud brightening: The importance of the seeded particle size distribution. *J. Atmos. Sci.* **78**, 3247–3262 (2021).
67. H. G. Houghton, Problems connected with the condensation and precipitation processes in the atmosphere. *Bull. Amer. Meteor. Soc.* **19**, 152–159 (1938).
68. R. Wood, Assessing the potential efficacy of marine cloud brightening for cooling Earth using a simple heuristic model. *Atmos. Chem. Phys.* **21**, 14507–14533 (2021).
69. A. K. L. Jenkins, P. M. Forster, The inclusion of water with the injected aerosol reduces the simulated effectiveness of marine cloud brightening. *Atmos. Sci. Lett.* **14**, 164–169 (2013).

70. C. Claudel, A. Lockley, F. Hoffmann, Y. Xia, Marine-cloud brightening: an airborne concept. *Environ. Res. Commun.* (2024); <https://doi.org/10.1088/2515-7620/ad2f71>.
71. H. Wang, G. Feingold, Modeling mesoscale cellular structures and drizzle in marine stratocumulus. Part II: The microphysics and dynamics of the boundary region between open and closed cells. *J. Atmos. Sci.* **66**, 3237–3256 (2009).
72. H. Trofimov, P. Post, E. Gryspeerd, T. Toll, Meteorological conditions favorable for strong anthropogenic aerosol impacts on clouds. *J. Geophys. Res. Atmos.* **127**, e2021JD035871 (2022).
73. J. Zhang, X. Zhou, T. Goren, G. Feingold, Albedo susceptibility of northeastern Pacific stratocumulus: The role of covarying meteorological conditions. *Atmos. Chem. Phys.* **22**, 861–880 (2022).
74. E. Erfani, P. Blosser, R. Wood, J. Mohrmann, S. J. Doherty, M. Wyant, K.-T. O, Simulating aerosol lifecycle impacts on the subtropical stratocumulus-to-cumulus transition using large-eddy simulations. *J. Geophys. Res. Atmos.* **127**, e2022JD037258 (2022).
75. K. Bilsback, D. Kerry, B. Croft, B. Ford, S. Jathar, E. Carter, R. Martin, J. Pierce, Beyond SO_x reductions from shipping: Assessing the impact of NO_x and carbonaceous-particle controls on human health and climate. *Environ. Res. Lett.* **15**, 124046 (2020).
76. K. Ricke, J. S. Wan, M. Saenger, N. J. Lutsko, Hydrological consequences of solar geoengineering. *Annu. Rev. Earth Planet. Sci.* **51**, 447–470 (2023).
77. T. Shepherd, Atmospheric circulation as a source of uncertainty in climate change projections. *Nat. Geosci.* **7**, 703–708 (2014).
78. H. Douville, K. Raghavan, J. Renwick, R. P. Allan, P. A. Arias, M. Barlow, R. Cerezo-Mota, A. Cherchi, T. Y. Gan, J. Gergis, D. Jiang, A. Khan, W. Pokam Mba, D. Rosenfeld, J. Tierney, O. Zolina, Water cycle changes, in *Climate Change 2021: The Physical Science Basis. Contribution of Working Group I to the Sixth Assessment Report of the Intergovernmental Panel on Climate Change*, V. Masson-Delmotte, P. Zhai, A. Pirani, S. L. Connors, C. Péan, S. Berger, N. Caud, Y. Chen, L. Goldfarb, M. I. Gomis, M. Huang, K. Leitzell, E. Lonnoy, J. B. R. Matthews, T. K. Maycock, T. Waterfield, O. Yelekçi, R. Yu, B. Zhou, Eds. (Cambridge Univ. Press, 2021), pp. 1055–1210.
79. H. Wang, P. J. Rasch, G. Feingold, Manipulating marine stratocumulus cloud amount and albedo: A process-modelling study of aerosol-cloud-precipitation interactions in response to injection of cloud condensation nuclei. *Atmos. Chem. Phys.* **11**, 4237–4249 (2011).
80. A. H. Berner, C. S. Bretherton, R. Wood, Large eddy simulation of ship tracks in the collapsed marine boundary layer: A case study from the Monterey area ship track experiment. *Atmos. Chem. Phys.* **15**, 5851–5871 (2015).
81. A. Jones, J. Haywood, O. Boucher, Climate impacts of geoengineering marine stratocumulus clouds. *J. Geophys. Res. D Atmos.* **114**, (2009).
82. B. Kravitz, A. Robock, O. Boucher, H. Schmidt, K. E. Taylor, G. Stenchikov, M. Schulz, The geoengineering model intercomparison project (GeoMIP). *Atmos. Sci. Lett.* **12**, 162–167 (2011).
83. S. Hill, Y. Ming, Nonlinear climate response to regional brightening of tropical marine stratocumulus. *Geophys. Res. Lett.* **39**, 15707 (2012).
84. P. J. Rasch, J. Latham, C.-C. J. Chen, Geoengineering by cloud seeding influence on sea ice and climate system. *Environ. Res. Lett.* **4**, 045112 (2009).
85. L. M. Russell, A. Sorooshian, J. H. Seinfeld, B. A. Albrecht, A. Nenes, L. Ahlm, Y. Chen, M. Coggon, J. S. Craven, R. C. Flagan, A. A. Frossard, H. Jonsson, E. Jung, J. J. Lin, A. R. Metcalf, R. Modini, J. Mülmenstädt, G. Roberts, T. Shingler, S. Song, Z. Wang, A. Wonschütz, Eastern Pacific emitted aerosol cloud experiment. *Bull. Am. Meteorol. Soc.* **94**, 709–729 (2013).
86. T. M. Van Reken, T. A. Rissman, G. C. Roberts, V. Varutbangkul, H. H. Jonsson, R. C. Flagan, J. H. Seinfeld, Toward aerosol/cloud condensation nuclei (CCN) closure during CRYSTAL-FACE. *J. Geophys. Res. Atmos.* **108**, 4633 (2003).
87. K. J. Sanchez, L. M. Russell, R. L. Modini, A. A. Frossard, L. Ahlm, C. E. Corrigan, G. C. Roberts, L. N. Hawkins, J. C. Schroder, A. K. Bertram, R. Zhao, A. K. Y. Lee, J. J. Lin, A. Nenes, Z. Wang, A. Wonschütz, A. Sorooshian, K. J. Noone, H. Jonsson, D. Toom, A. M. Macdonald, W. R. Leaitch, J. H. Seinfeld, Meteorological and aerosol effects on marine cloud microphysical properties. *J. Geophys. Res. Atmos.* **121**, 4142–4161 (2016).
88. Y. Peng, U. Lohmann, R. Leaitch, Importance of vertical velocity variations in the cloud droplet nucleation process of marine stratus clouds. *J. Geophys. Res.* **110**, D21213 (2005).
89. J. B. Jensen, R. J. Charlson, On the efficiency of nucleation scavenging. *Tellus* **36B**, 367–375 (1984).
90. L. M. Russell, J. H. Seinfeld, R. C. Flagan, R. J. Ferek, D. A. Hegg, P. V. Hobbs, W. Wobrock, A. I. Flossmann, C. D. O'Dowd, K. E. Nielsen, P. A. Durkee, Aerosol dynamics in ship tracks. *J. Geophys. Res. Atmos.* **104**, 31077–31095 (1999).
91. P. K. Quinn, D. J. Coffman, Local closure during the first aerosol characterization experiment (ACE 1): Aerosol mass concentration and scattering and backscattering coefficients. *J. Geophys. Res.* **103**, 16575–16596 (1998).
92. K. J. Sanchez, G. C. Roberts, R. Calmer, K. Nicoll, E. Hashimshoni, D. Rosenfeld, J. Ovadnevaite, J. Preissler, D. Ceburnis, C. O'Dowd, L. M. Russell, Top-down and bottom-up aerosol–cloud closure: Towards understanding sources of uncertainty in deriving cloud shortwave radiative flux. *Atmos. Chem. Phys.* **17**, 9797–9814 (2017).
93. P. J. Connolly, G. B. McFiggans, R. Wood, A. Tsimas, Factors determining the most efficient spray distribution for marine cloud brightening. *Philos. Trans. A Math. Phys. Eng. Sci.* **372**, 20140056 (2014).
94. F. Yang, F. Hoffmann, R. A. Shaw, M. Ovchinnikov, A. M. Vogelmann, An intercomparison of large-eddy simulations of a convection cloud chamber using haze-capable bin and Lagrangian cloud microphysics schemes. *J. Adv. Model. Earth Syst.* **15**, e2022MS003270 (2023).
95. R. A. Shaw, W. Cantrell, S. Chen, P. Chuang, N. Donahue, G. Feingold, P. Kollias, A. Korolev, S. Kreidenweis, S. Krueger, J.-P. Mellado, D. Niedermeier, L. Xue, Cloud–aerosol–turbulence interactions: Science priorities and concepts for a large-scale laboratory facility. *Bull. Am. Meteorol. Soc.* **101**, E1026–E1035 (2020).
96. K. Chang, J. Bench, M. Brege, W. Cantrell, K. Chandrakar, D. Ciochetto, C. Mazzoleni, L. R. Mazzoleni, D. Niedermeier, R. A. Shaw, A laboratory facility to study gas–aerosol–cloud interactions in a turbulent environment: The II chamber. *Bull. Am. Meteorol. Soc.* **97**, 2343–2358 (2016).
97. S. Thomas, F. Yang, M. Ovchinnikov, W. Cantrell, R. A. Shaw, Scaling of turbulence and microphysics in a convection–cloud chamber of varying height. *J. Adv. Model. Earth Syst.* **15**, e2022MS003304 (2023).
98. T. Yuan, L. Remer, H. Yu, Microphysical, macrophysical and radiative signatures of volcanic aerosols in trade wind cumulus observed by the A-Train. *Atmos. Chem. Phys.* **11**, 7119–7132 (2011).
99. M. W. Christensen, A. Gettelman, J. Cermak, G. Dagan, M. Diamond, A. Douglas, G. Feingold, F. Glassmeier, T. Goren, D. P. Grosvenor, E. Gryspeerd, R. Kahn, Z. Li, P.-L. Ma, F. Malavelle, I. L. McCoy, D. T. McCoy, G. McFarquhar, J. Mülmenstädt, S. Pal, A. Possner, A. Povey, J. Quaas, D. Rosenfeld, A. Schmidt, R. Schrödner, A. Sorooshian, P. Stier, V. Toll, D. Watson-Parris, R. Wood, M. Yang, T. Yuan, Opportunistic experiments to constrain aerosol effective radiative forcing. *Atmos. Chem. Phys.* **22**, 641–674 (2022).
100. D. T. McCoy, D. L. Hartmann, Observations of a substantial cloud-aerosol indirect effect during the 2014–2015 Bárðarbunga-Veiðivötn fissure eruption in Iceland. *Geophys. Res. Lett.* **42**, 10409–10414 (2015).
101. F. F. Malavelle, J. M. Haywood, A. Jones, A. Gettelman, L. Clarisse, S. Bauduin, R. P. Allan, I. H. H. Karset, J. E. Kristjánsson, L. Oreopoulos, N. Cho, D. Lee, N. Bellouin, O. Boucher, D. P. Grosvenor, K. S. Carslaw, S. Dhomse, G. W. Mann, A. Schmidt, H. Coe, M. E. Hartley, M. Dalvi, A. A. Hill, B. T. Johnson, C. E. Johnson, J. R. Knight, F. M. O'Connor, D. G. Partridge, P. Stier, G. Myhre, S. Platnick, G. L. Stephens, H. Takahashi, T. Thordarson, Strong constraints on aerosol–cloud interactions from volcanic eruptions. *Nature* **546**, 485–491 (2017).
102. P. Prabhakaran, F. Hoffmann, G. Feingold, Evaluation of pulse aerosol forcing on marine stratocumulus clouds in the context of marine cloud brightening. *J. Atmos. Sci.* **80**, 1585–1604 (2023).
103. D. C. Hernandez-Jaramillo, L. Harrison, B. Kelahe, Z. Ristovski, D. P. Harrison, Evaporative cooling does not prevent vertical dispersion of effervescent seawater aerosol for brightening clouds. *Environ. Sci. Technol.* **57**, 20559–20570 (2023).
104. A. Sorooshian, B. Anderson, S. E. Bauer, R. A. Braun, B. Cairns, E. Crosbie, H. Dadashazar, G. Diskin, R. Ferrare, R. C. Flagan, J. Hair, C. Hostetler, H. H. Jonsson, M. M. Kleb, H. Liu, A. B. MacDonald, A. McComiskey, R. Moore, D. Painemal, L. M. Russell, J. H. Seinfeld, M. Shook, W. L. Smith, K. Thornhill, G. Selioudis, H. Wang, X. Zeng, B. Zhang, L. Ziemba, P. Zuidema, Aerosol–cloud–meteorology interaction airborne field investigations: Using lessons learned from the U.S. West Coast in the design of ACTIVATE off the U.S. East Coast. *Bull. Am. Meteorol. Soc.* **100**, 1511–1528 (2019).
105. C. R. Mechoso, R. Wood, R. Weller, C. S. Bretherton, A. D. Clarke, H. Coe, C. Fairall, J. T. Farrar, G. Feingold, R. Garreaud, C. Grados, J. McWilliams, S. P. de Szoeke, S. E. Yuter, P. Zuidema, Ocean–cloud–atmosphere–land interactions in the southeastern Pacific: The VOCALS program. *Bull. Amer. Meteorol. Soc.* **95**, 357–375 (2014).
106. P. K. Quinn, T. S. Bates, D. J. Coffman, L. Upchurch, R. Moore, L. Ziemba, T. Bell, E. Saltzman, J. Graff, M. J. Behrenfeld, Seasonal variations in western North Atlantic remote marine aerosol properties. *J. Geophys. Res.* **124**, 14240–14261 (2019).
107. G. M. McFarquhar, C. S. Bretherton, R. Marchand, A. Protat, P. J. DeMott, S. P. Alexander, G. C. Roberts, C. H. Twohy, D. Toohey, S. Siems, Y. Huang, R. Wood, R. M. Rauber, S. Lasher-Trapp, J. Jensen, J. L. Stith, J. Mace, J. Um, E. Järvinen, M. Schnaiter, A. Gettelman, K. J. Sanchez, C. S. McCluskey, L. M. Russell, I. L. McCoy, R. L. Atlas, C. G. Bardeen, K. A. Moore, T. C. J. Hill, R. S. Humphries, M. D. Keywood, Z. Ristovski, L. Cravigan, R. Schofield, C. Fairall, M. D. Mallet, S. M. Kreidenweis, B. Rainwater, J. D'Alessandro, Y. Wang, W. Wu, G. Saliba, E. J. T. Levin, S. Ding, F. Lang, S. C. H. Truong, C. Wolff, J. Haggerty, M. J. Harvey, A. R. Klekociuk, A. McDonald, Observations of clouds, aerosols, precipitation, and surface radiation over the Southern Ocean: An Overview of CAPRICORN, MARCUS, MICRE, and SOCRATES. *Bull. Amer. Meteorol. Soc.* **102**, E894–E928 (2021).
108. D. T. McCoy, P. Field, H. Gordon, G. S. Elsaesser, D. P. Grosvenor, Untangling causality in midlatitude aerosol–cloud adjustments. *Atmos. Chem. Phys.* **20**, 4085–4103 (2020).
109. Y. Wang, J. H. Jiang, H. Su, Y. Choi, L. Huang, J. Guo, Y. L. Yung, Elucidating the role of anthropogenic aerosols in arctic sea ice variations. *J. Climate* **31**, 99–114 (2018).
110. E. Gryspeerd, T. Goren, T. W. P. Smith, Observing the timescales of aerosol–cloud interactions in snapshot satellite images. *Atmos. Chem. Phys.* **21**, 6093–6109 (2021).
111. E. Gryspeerd, F. Glassmeier, G. Feingold, F. Hoffmann, R. J. Murray-Watson, Observing short timescale cloud development to constrain aerosol–cloud interactions (2022); <https://doi.org/10.5194/acp-2022-335>.

112. P. Prabhakaran, F. Hoffmann, G. Feingold, Effects of intermittent aerosol forcing on the stratocumulus-to-cumulus transition. *Atmos. Chem. Phys.* **24**, 1919–1937 (2024).
113. W. I. Gustafson Jr., A. M. Vogelmann, Z. Li, X. Cheng, K. K. Dumas, S. Endo, K. L. Johnson, B. Krishna, T. Fairless, H. Xiao, The large-eddy simulation (LES) atmospheric radiation measurement (ARM) symbiotic simulation and observation (LASSO) activity for continental shallow convection. *Bull. Am. Meteorol. Soc.* **101**, E462–E479 (2020).
114. G. Utida, F. W. Cruz, J. Etourneau, I. Bouloubassi, E. Scheffé, M. Vuille, V. F. Novello, L. F. Prado, A. Sifeddine, V. Klein, A. Zular, J. C. C. Viana, B. Turcq, Tropical South Atlantic influence on Northeastern Brazil precipitation and ITCZ displacement during the past 2300 years. *Sci. Rep.* **9**, 1698 (2019).
115. C. W. Stjern, H. Muri, L. Ahlm, O. Boucher, J. N. S. Cole, D. Ji, A. Jones, J. Haywood, B. Kravitz, A. Lenton, J. C. Moore, U. Niemeier, S. J. Phipps, H. Schmidt, S. Watanabe, J. E. Kristjánsson, Response to marine cloud brightening in a multi-model ensemble. *Atmos. Chem. Phys.* **18**, 621–634 (2018).
116. J. T. Fasullo, N. Rosenblum, R. Buchholz, A multiyear tropical Pacific cooling response to recent Australian wildfires in CESM2. *Sci. Adv.* **9**, eadg1213 (2023).
117. Q. Tang, S. A. Klein, S. Xie, W. Lin, J.-C. Golaz, E. L. Roesler, M. A. Taylor, P. J. Rasch, D. C. Bader, L. K. Berg, P. Caldwell, S. E. Giangrande, R. B. Neale, Y. Qian, L. D. Riihimäki, C. S. Zender, Y. Zhang, X. Zheng, Regionally refined test bed in E3SM atmosphere model version 1 (EAMv1) and applications for high-resolution modeling. *Geosci. Model Dev.* **12**, 2679–2706 (2019).
118. V. Nataraja, S. Schmidt, H. Chen, T. Yamaguchi, J. Kazil, G. Feingold, K. Wolf, H. Iwabuchi, Segmentation-based multi-pixel cloud optical thickness retrieval using a convolutional neural network. *Atmos. Meas. Tech.* **15**, 5181–5205 (2022).
119. C. K. Yang, J. C. Chiu, A. Marshak, G. Feingold, T. Várnai, G. Wen, T. Yamaguchi, P. J. van Leeuwen, Near-cloud aerosol retrieval using machine learning techniques, and implied direct radiative effects. *Geophys. Res. Lett.* **49**, e2022GL098274 (2022).
120. J. Augustine, G. B. Hodges, Variability of surface radiation budget components over the U.S. from 1996 to 2019—Has Brightening Ceased? *J. Geophys. Res. Atmos.* **126**, e2020JD033590 (2021).
121. M. Wild, A. Ohmura, K. Makowski, Impact of global dimming and brightening on global warming. *Geophys. Res. Lett.* **34**, L04702 (2007).
122. J. J. Michalsky, C. N. Long, ARM solar and infrared broadband and filter radiometry. *Meteorol. Monogr.* **57**, 16.1–16.15 (2016).
123. M. D. Shupe, J. M. Comstock, D. D. Turner, G. G. Mace, Cloud property retrievals in the ARM program. *Meteorol. Monogr.* **57**, 19.1–19.20 (2016).
124. G. Feingold, I. Koren, T. Yamaguchi, J. Kazil, On the reversibility of transitions between closed and open cellular convection. *Atmos. Chem. Phys.* **15**, 7351–7367 (2015).
125. A. Possner, R. Eastman, F. Bender, F. Glassmeier, Deconvolution of boundary layer depth and aerosol constraints on cloud water path in subtropical stratocumulus decks. *Atmos. Chem. Phys.* **20**, 3609–3621 (2020).
126. A. K. L. Jenkins, P. M. Forster, L. S. Jackson, The effects of timing and rate of marine cloud brightening aerosol injection on albedo changes during the diurnal cycle of marine stratocumulus clouds. *Atmos. Chem. Phys.* **13**, 1659–1673 (2013).
127. L. Ahlm, A. Jones, C. W. Stjern, H. Muri, B. Kravitz, J. E. Kristjánsson, Marine cloud brightening—As effective without clouds. *Atmos. Chem. Phys.* **17**, 13071–13087 (2017).
128. J. M. Haywood, A. Jones, A. C. Jones, P. Halloran, P. J. Rasch, Climate intervention using marine cloud brightening (MCB) compared with stratospheric aerosol injection (SAI) in the UKESM1 climate model. *Atmos. Chem. Phys.* **23**, 15305–15324 (2023).
129. R. Wood, T. Ackerman, P. J. Rasch, K. Wanser, Could geoengineering research help answer one of the biggest questions in climate science? *Earth's Future* **5**, 659–663 (2017).
130. B. Parkes, A. Challinor, K. Nicklin, Crop failure rates in a geoengineered climate: Impact of climate change and marine cloud brightening. *Environ. Res. Lett.* **10**, 084003 (2015).

Acknowledgments: We thank D. W. Fahey, G. Frost, and S. Nasiri for comments and ongoing support of this effort. We thank the Department of Energy (DOE) Argonne National Laboratory for Figs. 1 to 3, and C. R. Thompson (NOAA/Chemical Science Laboratory) for Fig. 4. The results and conclusions, as well as any views or opinions expressed herein, are those of the authors and do not necessarily reflect the views of NASA, NOAA, NSF, or the Departments of Commerce or Energy. **Funding:** This work was supported by NOAA Climate Program Office Earth's Radiation Budget Grant #03-01-07-001; Atmospheric System Research (ASR) program as part of the Department of Energy (DOE) Office of Biological and Environmental Research (BER); DOE/BER grants DE-SC0022227 (to D.T.M.), DE-SC0021045 (to L.M.R.), and DE-SC0012704 (to A.C.M. and F.Y.); University of Colorado, Cooperative Institute for Research in Environmental Sciences Visiting Fellow Program (to M.S.D.); Pacific Northwest National Laboratory project 76858 (to C.M.K.); Pacific Northwest National Laboratory project 57131 under Battelle Memorial Institute contract DE-A06-76RLO 1830 (to C.M.K., J.M., M.W.C., and A.G.); Argonne National Laboratory DE-AC02-06CH11357 (to V.P.G.); Lawrence Livermore National Laboratory under contract nos. DE-AC52-07NA27344 and LLNL-JRNL-853525 (to X.Z.); Jet Propulsion Laboratory, California Institute of Technology, under a contract with the National Aeronautics and Space Administration (to M.L.); NSF grant AGS-2133229 (to R.A.S. and W.C.); and German Research Foundation under grants HO 6588/1-1 and HO 6588/3-1 (to F.H.) and 500932476 (to A.P.). **Author contributions:** G.F.: Conceptualization, supervision, project administration, and visualization. L.M.R.: Conceptualization and investigation. V.P.G.: Conceptualization, methodology, supervision, and visualization. W.C.: Conceptualization. P.K.Q.: Conceptualization. M.S.D.: Conceptualization, investigation, validation, supervision, and visualization. R.W.: Methodology and resources. P.P.: Conceptualization. J.Z.: Conceptualization. J.M.: Conceptualization and validation. X.Z.: Conceptualization. C.M.K.: Conceptualization. M.W.C.: Conceptualization. D.T.M.: Conceptualization. A.C.M.: Conceptualization and project administration. F.H.: Conceptualization. A.P.: Conceptualization and resources. M.L.: Conceptualization. A.G.: Conceptualization. Y.M.: Conceptualization and methodology. F.G.: Conceptualization. K.S.S.: Conceptualization. All authors contributed to the writing and reviewing of the manuscript. **Competing interests:** The authors declare that they have no competing interests.

Submitted 6 September 2023

Accepted 14 February 2024

Published 20 March 2024

10.1126/sciadv.adi8594