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Key Points:

- Diffusion of sunlight by stratospheric aerosols can substantially increase cloud albedo
- The radiative effect of stratospheric aerosol injection would be dominated by inadvertent cloud brightening at many locations and times
- This diffusion-brightening effect has important implications for solar radiation management

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Stratospheric Aerosol Injection Would Change Cloud Brightness

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Abstract Intentional modification of the sunlight reflected back to space by Earth has received increasing attention as a potential tool to combat the current climate crisis. Two approaches have emerged as most viable: Stratospheric Aerosol Injection (SAI) and Marine Cloud Brightening. This study identifies a substantial but unrecognized source of cloud brightening caused by SAI, which we call “diffusion-brightening”; essentially, diffusion of the radiation field that would accompany SAI can result in sunlight entering clouds at steeper angles, which increases cloud albedo without actively injecting aerosols into clouds. We present idealized calculations that suggest the diffusion-brightening effect can lead to clouds reflecting about 10% more of the incoming sunlight depending on stratospheric aerosol, cloud, and Sun conditions. We show that the radiative effect of diffusion-brightening could exceed that of stratospheric aerosol reflection in many cloudy scenes, which has global relevance given that clouds cover around two-thirds of the planet.

Plain Language Summary The deliberate release of particles in the stratosphere—to reflect a small amount of sunlight back to space and thereby cool the surface—is a controversial but increasingly common topic of debate in the face of ongoing global warming. One known side-effect is that the sunlight below these particles would be spread out into different directions. We demonstrate that this spreading of sunlight into different directions can make low-level marine clouds substantially brighter. The importance of this cloud brightening does not appear to have been appreciated in previous studies on the topic. Yet, this cloud brightening is crucial to understand the overall amount of sunlight that would be reflected and has profound implications for potential implementation of any such approach in the future.

1. Introduction

Recent climate change is unprecedented over many centuries to many thousands of years and the human influence is unequivocal (IPCC, 2021). Consequently, efforts to limit further human-induced climate changes are at the forefront of international politics. Perhaps most notably, the landmark Paris Agreement (UNFCCC, 2015)—a legally binding international treaty on climate change—has a goal of limiting global-mean near-surface temperature increase to 2°C above pre-industrial levels while also pursuing efforts toward a limit of 1.5°C (IPCC, 2018). Early indications are that current mitigation efforts and existing future commitments to emission reductions will be inadequate to accomplish this goal (e.g., Dunstone et al., 2024; Huang & Zhai, 2021; Rogelj et al., 2016; Smith et al., 2018; UNEP, 2023a; UNFCCC, 2024; Xu et al., 2018).

The monumental challenges associated with drastic emissions reductions have given rise to increased discussions regarding direct intervention in the climate system (e.g., Climate Overshoot Commission, 2023; Lawrence et al., 2018; MacMartin et al., 2018). One such approach, solar radiation management (SRM), involves deliberately altering the Earth’s atmosphere to reflect more sunlight back to space (e.g., NASEM, 2021; NRC, 2015; UNEP, 2023b). Two of the strategies considered most viable for SRM are Stratospheric Aerosol Injection (SAI)—the injection of aerosol particles into the upper atmosphere to directly reflect sunlight (e.g., Crutzen, 2006; Irvine & Keith, 2020; Robock, 2016), and Marine Cloud Brightening (MCB)—the injection of aerosol particles into near-surface oceanic clouds to indirectly increase cloud albedo (e.g., Diamond et al., 2022; Feingold et al., 2024; Latham, 1990; Wood, 2021). It is important to note that neither SAI nor MCB replace the need for emission reductions; rather, they are being considered as potential options to temporarily shave the peak of the most severe temperature changes thereby providing more time to achieve decarbonization (Horton, 2015; Long & Shepherd, 2014).

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Thus far, the potential climate impacts of SAI and MCB have been assessed and compared almost entirely independently (e.g., Haywood et al., 2023; Visioni et al., 2024). One exception is Boucher et al. (2017) who found a quasi-additivity in SAI and MCB radiative effects using climate model simulations. Here, instead of focusing on the overall radiative effects, we focus on the underlying mechanisms. It is well-known that SAI would generate diffusion of the radiation field (Kravitz et al., 2012), with implications for solar renewable energy (Baur et al., 2024; Murphy, 2009) and agriculture (Greenwald et al., 2006; Gu et al., 2003; Proctor et al., 2018; Roderick et al., 2001; Xia et al., 2016). However, changes in cloud brightness associated with diffusion of the radiation field, which we coin “diffusion-brightening,” appears to have been largely overlooked. The purpose of the present study is to highlight the potential importance of this diffusion-brightening effect and quantify how its magnitude depends on a variety of conditions.

Section 2 provides a theoretical argument for why cloud brightness can increase upon diffusion of the solar radiation field by SAI. Section 3 presents simulation results, beginning with an idealized SAI scenario to quantify the diffusion-brightening effect and then exploring sensitivities to scene conditions to allow more general interpretation. Section 4 summarizes the results and provides concluding remarks.

2. Theoretical Argument for Diffusion-Brightening

Consider a theoretical cloud under the following assumptions: homogeneous and unbounded in the horizontal (so-called “plane-parallel”), non-absorbing (so-called “conservative-scattering”), and infinitely deep (so-called “semi-infinite”). The albedo of this theoretical cloud must be unity regardless of the direction of incident radiation; a photon entering the cloud cannot escape from the cloud sides or base and cannot be absorbed, so its only eventual fate is to emerge at cloud top. However, when examining trajectories of photons through this theoretical cloud, more nuanced behavior emerges.

An individual photon entering this theoretical cloud will travel a random distance before interacting with a cloud droplet, with the mean distance traveled by a large number of photons following the Bouguer-Beer-Lambert law (Mayerhöfer et al., 2020). Upon interacting with a cloud droplet under conservative-scattering, the photon is scattered into a random direction, with the probability of any given direction determined by the scattering phase function (inset Figure 1a). An important property of the phase function for solar radiation interacting with cloud droplets, and a key factor for this study, is that it is far more likely that the photon will be scattered into the forward direction. After being scattered, the photon again travels a random distance before interacting with another cloud droplet, and the multiple-scattering process ensues until the photon eventually emerges from cloud top. It follows from the strong preference for forward scattering that photons initially entering the cloud from directly overhead will, on average, penetrate deeper into this theoretical cloud than photons entering at larger incidence angles (Figure 1a).

Tracking the journey of photons given the optical properties of the atmosphere is precisely the underlying principle of Monte-Carlo radiative transfer models. Hence, we use Monte-Carlo radiative transfer, with 100,000 photons at incidence angles every 10° from 0 to 80° , to quantify the average photon penetration depth for our theoretical cloud (Figure 1b). Assuming arbitrary but physically reasonable cloud and radiation properties (see details in Figure 1 caption), it is clear that the median photon penetration depth is indeed a strong function of incidence angle. For example, a photon entering at a normal incidence angle will penetrate, on average, more than twice as deep as a photon entering at 60° .

Having established the relationship between incidence angle and photon penetration depth for our theoretical cloud, we can now contemplate the implications of relaxing the initial assumptions. Most importantly in this context, dropping the semi-infinite assumption implies that the cloud has a finite depth and therefore photons can emerge from cloud base. Photons that were more likely to penetrate deeper into the semi-infinite cloud, that is, those entering at close-to-normal incidence angles, are now more likely to emerge at cloud base. In other words, it is more difficult for multiple scattering to “turn around” photons that enter at close-to-normal incidence angles. It follows that the cloud albedo is increased at steeper incidence angles. Analytically, the photons emerging at cloud top travel a mean horizontal distance of $h/\sqrt{(1-g)\tau}$ (Marshak et al., 1995) and have a transport mean free path of $h/[(1-g)\tau]$ (Davis et al., 1997) where h is the cloud geometric depth, τ is the cloud optical depth (COD), and g is the asymmetry parameter.

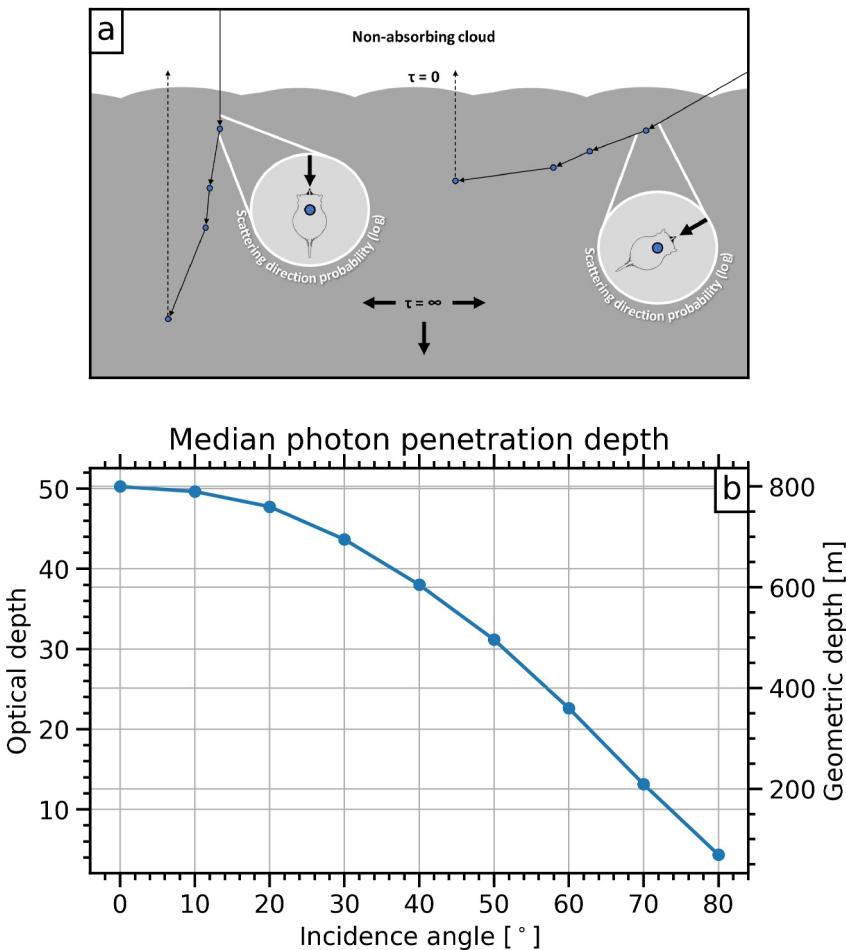


Figure 1. Photon penetration depth dependence on incidence angle into a plane-parallel, semi-infinite cloud with conservative scattering. (a) Schematic of two representative photon trajectories that are identical other than the incidence angle (left photon: 0° ; right photon: 60°). Eventually, the photon must exit at the cloud top as indicated by the dashed arrows. Inset are Mie-derived representations of the scattering phase function for a size parameter of 100 ($2\pi r/\lambda$ where r is the particle radius and λ is wavelength of the incident radiation), which is typical of cloud droplets and solar radiation, and is plotted as the logarithm of the probability of scattering direction (adapted from Petty (2006)). (b) Median photon penetration depth derived from Monte-Carlo radiative transfer. The geometric depth on the right axis corresponds to a cloud droplet number concentration of 100 cm^{-3} and a cloud droplet effective radius of $10 \mu\text{m}$. A Henyey-Greenstein phase function with an asymmetry parameter of 0.85 is assumed.

The connection of the preceding thought experiment to SAI and cloud albedo is obvious. Since stratospheric aerosols diffuse the radiation field, they can change the average incidence angle at which radiation enters clouds below, therefore changing cloud albedo. This is particularly relevant for marine clouds because photons exiting at cloud base are very likely to be absorbed by the dark ocean surface below. While this effect follows from basic radiative and geometric principles and is seemingly known (Kokhanovsky, 2004), it has not been quantified in the context of SRM; this effect was not even noted in a recent comprehensive review of the impacts of SAI (Huynh & McNeill, 2024). To address this shortcoming, we next proceed to quantify the potential importance of this diffusion-brightening effect under an idealized SAI scenario.

3. Simulation of the Diffusion-Brightening Effect

3.1. Idealized SAI Scenario

In order to set up an idealized simulation that is meaningful for quantifying the radiative interactions of stratospheric aerosols and clouds, we loosely target low altitude clouds over ocean at low-latitudes. This is because low altitude clouds are the most frequent clouds over ocean, and ocean is the most widespread surface type at low

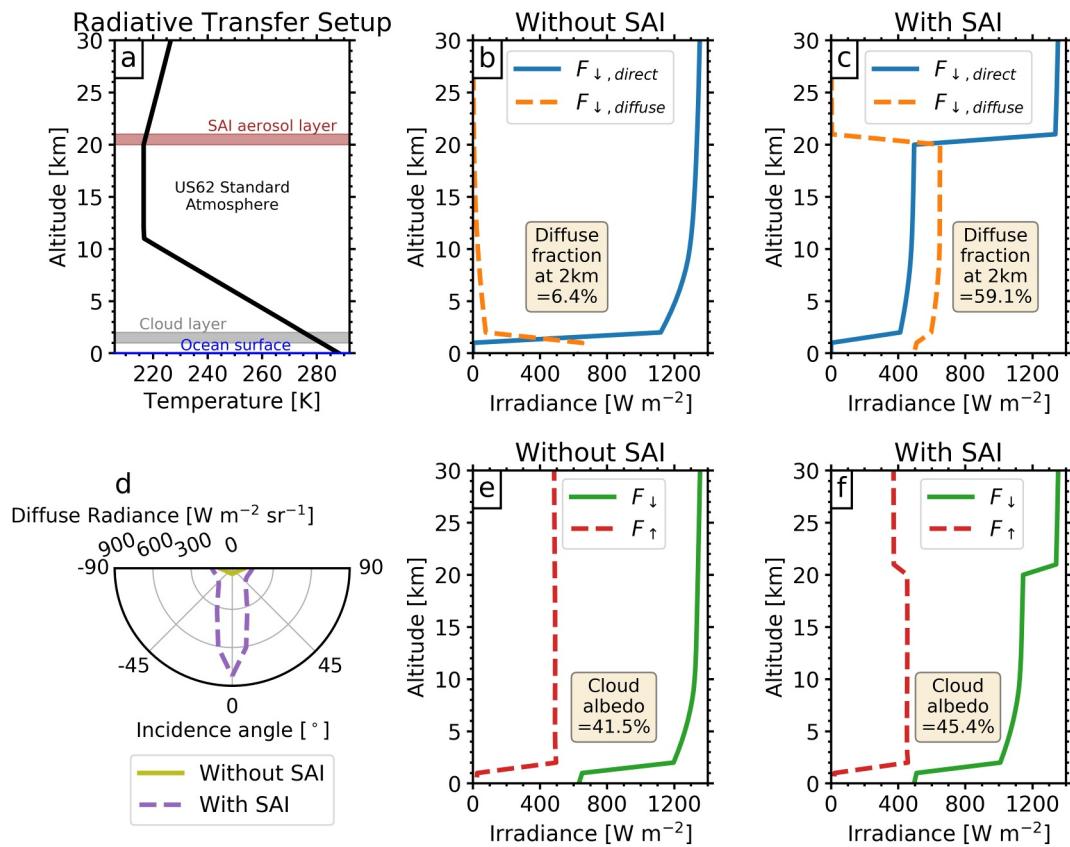


Figure 2. Simulation of the diffusion-brightening effect under an idealized SAI scenario. (a) Schematic of radiative transfer model setup. (b) Downward direct ($F_{\downarrow, \text{direct}}$) and diffuse ($F_{\downarrow, \text{diffuse}}$) irradiance profiles without SAI. (c) Same as (b) but with SAI. (d) The angular distribution of downward diffuse radiance at cloud top with and without SAI. (e) Downward (F_{\downarrow}) and upward (F_{\uparrow}) irradiance profiles without SAI. (f) Same as (e) but with SAI.

latitudes, and low latitudes are the region where most solar energy is input to the Earth system because the Sun is often high in the sky. We represent these conditions in the Santa Barbara DISORT Atmospheric Radiative Transfer (SBDART) code (Ricchiazzi et al., 1998), as shown schematically in Figure 2a. We include a liquid cloud in the 1–2 km layer, which is the closest layer to the surface that is not in contact with the ground. The cloud is initially defined to have a COD of 10 and an effective radius of 8 microns with an assumed gamma droplet size distribution. From SBDART's standard input options, we select the Lambertian ocean surface model and the US62 standard atmosphere that provides vertical profiles of radiatively active trace gas concentrations. Solar zenith angle (SZA) is initially set to 0° . Calculations are run every 5 nm from 0.2 to 5.0 microns followed by spectral-integration to obtain the broadband solar radiation. We use 20 streams to ensure that the angular distribution of the radiation field is resolved well.

An idealized SAI scenario is emulated by adding aerosols in the 20–21 km layer with SBDART's "fresh volcanic" stratospheric aerosol model, which provides optical properties for a typical size distribution and composition of fresh volcanic aerosol. We chose this aerosol model because SAI is often inspired by the impacts of a strato-volcanic eruption, but we note that this model includes non-negligible aerosol absorption that is addressed later. The initial aerosol optical depth (AOD) is set to 1, which is an intentionally heavy aerosol loading to maximize signal detection. For context, SAI scenarios in global models indicate a global-mean AOD of about 0.15 per K cooling, although AOD would not be evenly distributed and major deployment could be associated with AOD approaching 1 depending on the latitude and implementation strategy (Visioni et al., 2021, 2024). The sensitivity of results to stratospheric AOD, COD, and SZA will be examined in Section 3.3.

To demonstrate the extent to which stratospheric aerosol can diffuse the radiation field, we first consider profiles of downward diffuse and direct solar irradiance. Without stratospheric aerosol (Figure 2b), the downward

irradiance extending down to the cloud layer is almost entirely confined to the direct solar beam, with only a small diffuse component associated with molecular scattering. In contrast, including stratospheric aerosols (Figure 2c) extinguishes most of the direct beam in the aerosol layer. Since the majority of the extinguished radiation is scattered downward, the direct beam is essentially converted into diffuse radiation by the aerosols. At the cloud top altitude of 2 km, the downward irradiance without stratospheric aerosol is composed of 6.4% diffuse radiation, but with stratospheric aerosol this drastically increases to 59.1% diffuse radiation. Note that the diffuse radiation field is not isotropic; the sharp forward scattering peak in the phase function, combined with the fact that the stratospheric aerosol layer is dominated by single-scattering, dictates that the diffuse radiation is dispersed mostly around the direction of the direct beam (Figure 2d).

Since diffusion of the direct beam by stratospheric aerosols causes most of the photons to enter the cloud at off-normal angles, cloud albedo—the ratio of upward to downward shortwave irradiance at cloud top—increases (see Section 2). Without stratospheric aerosol, the cloud albedo is 0.415 (Figure 2e). With stratospheric aerosol, the cloud albedo increases to 0.454 (Figure 2f). Therefore, diffusion of the radiation field by stratospheric aerosol resulted in a 9.3% increase in cloud albedo.

It is worth noting that the top-of-atmosphere (TOA) upward irradiance is smaller in the simulation with stratospheric aerosol than without stratospheric aerosol (compare Figures 2e and 2f), which is somewhat surprising given that the primary intent of SAI is to increase the TOA upward irradiance. This occurs in our simulations largely because SBDART's fresh volcanic aerosol model includes non-negligible absorption. Since the simulated cloudy scene is already bright in the absence of aerosol, adding an aerosol layer with an absorbing component actually makes the scene darker overall (Haywood & Shine, 1997). It follows that the diffusion-brightening mechanism is partly counteracting an overall scene darkening due to stratospheric aerosol absorption in this case. This result highlights an important tangential point: the single scattering albedo of any candidate SAI aerosol needs to be extremely close to one to increase the TOA upward irradiance in cloudy scenes. Aerosols with even a small absorbing component, which have been proposed for practical reasons (Gao et al., 2021), will interfere with and potentially absorb radiation that would have otherwise been reflected by bright clouds. This would not be a rare occurrence given that clouds cover about two thirds of the Earth (King et al., 2013).

3.2. Relative Importance of Stratospheric Aerosol Reflection and Diffusion-Brightening

To avoid the complicating influence of aerosol absorption discussed above, we repeat our simulations but set the single scattering albedo of the stratospheric aerosols to one at all wavelengths. This represents a “perfect” SAI aerosol composition and therefore an upper bound on SAI scattering efficiency. We use this simulation to test the relative contributions to the TOA radiative effect of SAI from the (primarily intended) reflection directly from the stratospheric aerosols to the (inadvertent) cloud reflection via diffusion-brightening. To isolate the contributions from these mechanisms, we run a further set of simulations where we scale the COD in the SAI scenario such that the cloud albedo matches its value without stratospheric aerosols. The difference in TOA upward irradiance between this calculation, and the calculation with the unscaled COD is taken to be the contribution from diffusion-brightening. The remaining TOA radiative effect under the SAI scenario is assigned to reflection directly from stratospheric aerosols.

The contribution of stratospheric aerosol reflection (Figure 3, blue dashed line) is largest for optically thin clouds and rapidly decreases with increasing COD because aerosol reflection becomes less effective at brightening a scene as the scene itself becomes brighter. Interestingly, the aerosol reflection contribution becomes positive (i.e., a darkening contribution) for large CODs beyond about 17, which may seem counter-intuitive for perfectly scattering aerosol. There are at least two reasons for this: (a) diffusion of the radiation field by aerosols increases the path length of the photons between the aerosol layer and the cloud layer thereby inducing more absorption by water vapor and other gases that absorb at shortwave wavelengths, and (b) small SZAs combined with bright clouds can cause aerosols to trap more sunlight below the cloud layer than they reflect even at non-absorbing shortwave wavelengths (Boucher et al., 1998; Haywood & Shine, 1997). These effects are included in the stratospheric aerosol reflection calculation because it is defined here as the residual of the total TOA radiative effect and the diffusion-brightening.

The contribution of the diffusion-brightening effect (Figure 3, orange dotted line) initially increases in magnitude with increasing COD, peaking at a COD of about 5, and decreases in magnitude thereafter. The “sweet spot” at intermediate CODs represents a trade-off between the increasing amount of reflected sunlight with increasing

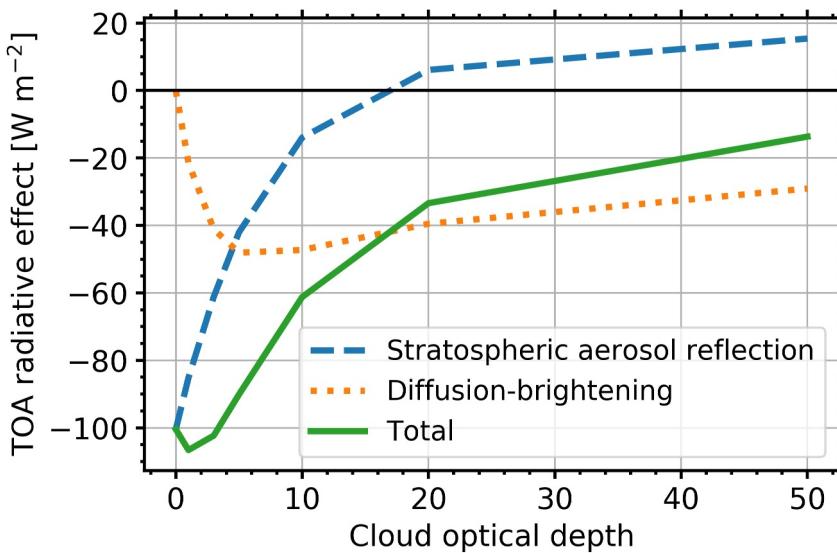


Figure 3. Simulated contributions of stratospheric aerosol reflection and the diffusion-brightening effect to the total TOA radiative effect of SAI. Specifically, this is the TOA upward irradiance for simulations without stratospheric aerosol minus the TOA upward irradiance for simulations with stratospheric aerosol. Plotted as a function of cloud optical depth for the idealized SAI scenario with perfectly scattering stratospheric aerosols.

COD and the decreasing efficiency of the diffusion brightening effect in percentage terms (see Figure 4b and related discussion in Section 3.3). Despite the decreasing magnitude of diffusion-brightening beyond CODs of about 5, this mechanism continues to dominate the overall radiative effect (Figure 3, green solid line). Since only 20% of detectable global daytime clouds have COD below 3, with 40% in the range 3–10 and 40% above 10 (Delgado-Bonal et al., 2024), the dominance of diffusion-brightening may be ubiquitous.

3.3. Sensitivity to Aerosol, Cloud, and Sun Conditions

In Section 3.1 we fixed several important scene conditions to arbitrary (but realistic) values. In this section we test the sensitivity of the diffusion-brightening effect to three key scene conditions: the stratospheric AOD, the COD, and the SZA.

The enhancement in cloud albedo associated with diffusion-brightening increases with increasing stratospheric AOD (Figure 4a). This is expected behavior because a larger aerosol loading leads to greater diffusion of the radiation field and therefore a steeper mean incidence angle at cloud top. Since the relationship is close-to-linear, even relatively small AODs of 0.1–0.2 are associated with cloud albedo enhancements of approximately 1%–2%.

The cloud albedo enhancement decreases rapidly with increasing COD up to the COD in our idealized case of 10, and decreases more gradually thereafter (Figure 4b). This is because at increasingly larger CODs there is a higher likelihood that photons will be turned around and reflected out of the top of the cloud by multiple scattering regardless of incidence angle. At smaller COD, the incidence angle has a much larger impact because of the higher likelihood of photons exiting at cloud base. This explains the “sweet spot” seen in Figure 3 after accounting for the magnitude of reflected energy.

The cloud albedo enhancement decreases with increasing SZA, and even turns negative around 50° (Figure 4c). This potential cloud darkening due to diffusion of the radiation field at high SZAs is relevant for high-latitude regions where the Sun is consistently lower in the sky, or for any location close to sunrise and sunset. However, it is important to note that the energy input to the Earth system is disproportionately larger at small SZAs due to the cosine dependence of TOA downward irradiance on SZA. Furthermore, the expansive marine stratocumulus decks that would likely be key contributors to the diffusion-brightening effect are located in the sub-tropics and typically have maximum cloud coverage in spring or summer when small SZAs are common (Klein & Hartmann, 1993). Therefore, when weighted by energy, it is reasonable to anticipate that cloud albedo enhancement (i.e., brightening) would dominate globally. However, a global analysis is needed to quantify this because

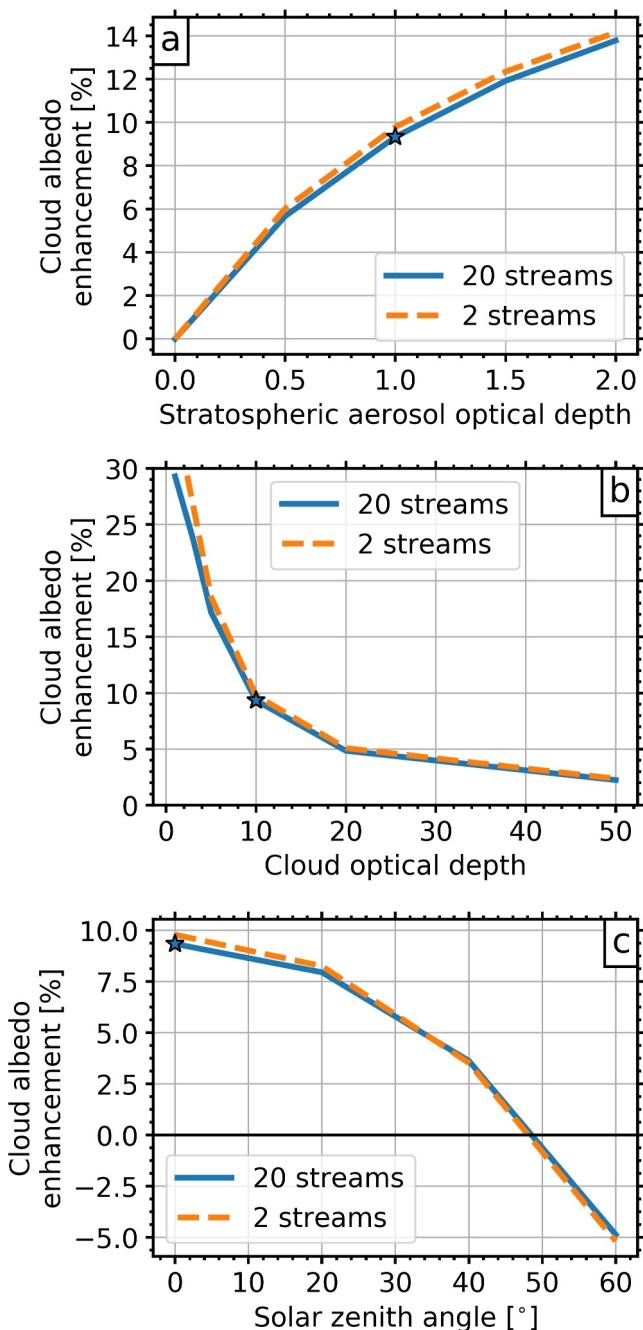


Figure 4. Sensitivity of cloud albedo enhancements from the diffusion-brightening effect to (a) stratospheric aerosol optical depth, (b) cloud optical depth, and (c) solar zenith angle. Each plot includes the main calculation with 20 streams and a sensitivity test with 2 streams. The star on each plot indicates the default value in the idealized SAI scenario.

important contributions to the shortwave cloud radiative effect also occur in other regions such as the inter-tropical convergence zone and the mid-latitude storm tracks (Allan, 2011). Quantification of the global average effect of diffusion-brightening under different SAI scenarios clearly requires convolution of stratospheric aerosol, cloud, and SZA distributions across diurnal and seasonal cycles, which is beyond the scope of this study but could be explored using satellite observations or global models in future work.

For all three scene properties explored in Figure 4, the difference between simulations with 20 streams and 2 streams is rather small and the general behavior is captured in both cases. This indicates that global models that typically employ 2-stream radiation calculations are likely already, implicitly, representing the diffusion-brightening effect. Even though the diffusion-brightening effect is not necessarily missing in global models, explicit quantification of this effect provides a markedly different interpretation of the overall aerosol radiative effect. This interpretation is crucial for solar radiation management because optimizing any potential implementation strategy requires a complete understanding of the underlying processes.

4. Summary and Conclusions

This study highlights an important radiative connection between stratospheric aerosols and cloud brightness, which we call “diffusion-brightening.” The diffusion-brightening effect occurs because stratospheric aerosols diffuse the radiation field below, which can cause sunlight to enter clouds at steeper angles, increasing cloud brightness. Our simulations show that this effect can increase marine low-cloud albedo by about 10% from its baseline value under an idealized SAI scenario. Sensitivity tests show that this result depends on the cloud, aerosol, and Sun conditions, all of which would need to be convolved in global analyses that consider this effect. We also demonstrate that the contribution to the instantaneous TOA radiative effect of SAI from diffusion-brightening could exceed the contribution from direct reflection by stratospheric aerosols in many cloudy scenes.

While global models most-likely represent the diffusion-brightening effect implicitly, explicit appreciation of this effect has profound practical implications. For example, injection of aerosols above clouds may present a remote implementation strategy for brightening of clouds. Compared to traditional MCB, that is, microphysical brightening of clouds, remote cloud brightening could be simpler to implement and avoids the large uncertainties associated with cloud lifetime effects. Furthermore, since the diffusion-brightening effect does not strictly require aerosols to exist in the stratosphere but only above clouds, it can be speculated that aerosols injected above marine boundary layer clouds in the troposphere would initially generate diffusion-brightening followed by traditional MCB if the aerosols are later entrained into the clouds. We also note that the idealized SAI scenario simulated in this study resulted in a change in shortwave absorption within the cloud layer (not shown), which could impact cloud evolution (Boers & Mitchell, 1994; Zhang et al., 2024). Future work is encouraged to explore these concepts.

Alternatively, viewed from the perspective of SAI implementation, stratospheric aerosols that more efficiently diffuse the radiation field may provide a substantial and previously under-appreciated leverage to increase the overall effectiveness of SAI. Given the decreased impact of diffusion-brightening at large CODs, it is also evident that SAI will be associated with diminishing returns if traditional MCB is indeed successful at brightening clouds. Overall, the diffusion-brightening effect identified here demonstrates a crucial radiative connection between

stratospheric aerosols and marine clouds and calls for further work on possible combinations and combined effects of SAI and MCB.

Data Availability Statement

All data used in this study and the code used to produce the figures has been archived at <https://csl.noaa.gov/groups/csl9/datasets/data/2024-Gristey-Feingold/>. The SBDART software can be downloaded at <https://github.com/paulricchiazz/SBDART>.

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