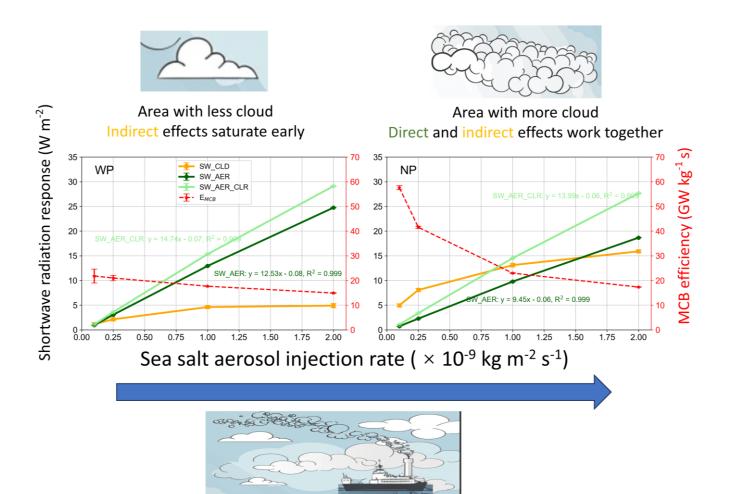
1	The effectiveness of solar radiation management for marine cloud brightening
2	geoengineering by fine sea spray in worldwide different climatic regions
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26 Abstract

27 Marine Cloud Brightening (MCB) geoengineering aims to inject aerosols over oceans to brighten clouds and reflect more sunlight to offset the impacts of global warming or to achieve localized climate 28 cooling. There is still controversy about the contributions of direct and indirect effects of aerosols in 29 implementing MCB and the lack of quantitative assessments of both. Here, we design model simulations 30 with injected sea-salt aerosols in the same framework for five open oceans around the globe. Our results 31 show that a uniform injection strategy that does not depend on wind speed captures the sensitive areas of 32 the regions that produce the largest radiative perturbations during the implementation of MCB. When the 33 injection amounts are low, the sea-salt aerosols dominate the shortwave radiation mainly through the 34 indirect effects of brightening clouds, showing obvious spatial heterogeneity. As the indirect effect of 35 aerosols saturates with increasing injection rates, the direct effect increases linearly and exceeds the 36 indirect effects, producing a consistent increase in the spatial distributions of top-of-atmosphere upward 37 shortwave radiation. This study provides quantifiable radiation and cloud variability data for multiple 38 regional MCB implementations and suggests that injection strategies can be optimized by adjusting 39 injection amounts and selecting sensitive areas in the simulations of regional models. 40

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Keywords: marine cloud brightening; solar radiation management; fine sea spray; climatic ocean regions;
 geoengineering

45 **1. Introduction**

As global temperatures continue to rise, the international community is facing an unprecedented challenge 46 to achieve the ambitious goal set in the Paris Agreement of limiting global warming to within 1.5 ^{0}C 47 (Mengel et al., 2018). One of the key outcomes of the recently concluded 28th Conference of the Parties 48 (COP28) was the completion of the first Global Stocktake (GST), a mid-term assessment of the progress 49 made by countries toward achieving the climate goals of the Paris Agreement. However, the report 50 highlighted that current efforts to reduce emissions had fallen short of the intended targets 51 (https://www.cop28.com/). Against this backdrop, scientists are turning their attention to more innovative 52 geoengineering methods by attempting to reduce or offset the impacts of climate change through artificial 53 54 interventions in the climate (Visioni et al., 2023). Some geoengineering methods seek to capture or remove CO₂ from the atmosphere to increase carbon sinks, while others focus on modifying solar 55 radiation, reducing incoming solar shortwave radiation, or reflecting more sunlight to cool the earth, 56 known as solar radiation management (SRM) (Lenton and Vaughan, 2009). Among these, marine cloud 57 58 brightening (MCB) has a certain realistic basis and is considered the most likely SRM method for regional applications (Latham et al., 2014). It has been observed that exhaust emissions from ocean-going vessels 59 can lead to brighter clouds, with clear ship tracks also visible from satellites, and MCB aims to replicate 60 this effect by spraying sea-salt aerosols (Chen et al., 2012). 61

Aerosol-cloud interactions and their impacts on climate are complex (Rosenfeld et al., 2014, 2019). 62 Injected sea-salt aerosols affect clouds through indirect effects (Paulot et al., 2020). In the case of a 63 constant liquid water content, an increase in cloud droplet number concentration (CDNC) decreases the 64 cloud droplet size, increases the total surface area of cloud droplets, thereby enhancing the cloud albedo, 65 forming brighter clouds, and reflecting more sunlight back to space (the first indirect effect or Twomey 66 effect) (Twomey, 1974). At the same time, the decrease in cloud droplet size suppresses precipitation, 67 thereby increasing the cloud's lifespan and optical thickness (the second indirect effect of aerosols) 68 (Albrecht, 1989). Reducing the cloud drop size induces a faster evaporation and loss of cloud water. 69 However, the effect of the coarse sea spray aerosols has an opposite effect that offsets the loss of liquid 70 71 water path (Liu et al., 2022). In addition, those aerosols that are not injected into the clouds scatter more sunlight back into space through the direct scattering effect (Ahlm et al., 2017; Partanen et al., 2012; Zhao 72 et al., 2021). Therefore, this method is also called marine sky brightening (MSB), which can work even 73 when there are no clouds. Here, we collectively refer to the practice of injecting sea-salt aerosols as MCB. 74 Compared to other geoengineering schemes, such as stratospheric aerosol injection (SAI), MCB has 75

unique advantages. For example, the sprayed aerosols have lower environmental risks and can be applied locally to change the regional climate (Latham et al., 2008). Their deployment costs are relatively low and flexible (Kravitz et al., 2014; Latham et al., 2012, 2014). However, despite these potential advantages, the long-term effects and potential risks of MCB are not fully understood, and there are significant uncertainties as well as ethical, political, and environmental risks. Therefore, most of the current literatures examine the environmental and climate impacts of MCB implementation through modeling.

Table S1 summarizes the results of current modeling simulations on MCB with sea-salt aerosols, as 82 well as their implementation strategies. Most MCB studies use Earth-System Models to assess the impacts 83 of the implementation of MCB on climate. Early MCB studies assumed the effects of MCB 84 implementation by setting a fixed CDNC or directly modifying the cloud effective radius ($r_{\rm e}$), ignoring 85 the processes such as generation, transport, dry and wet deposition, and activation of injected sea-salt 86 aerosols, and not including the direct radiative effect of aerosols. With the development of models, 87 researchers started to conduct more detailed studies by injecting aerosols or increasing sea-salt aerosol 88 emissions, taking into account the post-injection processes of aerosols mentioned above. The 89 implementation region of MCB is crucial. Existing studies have focused on the impacts of MCB 90 91 implementation in three key areas: open oceans globally, the equatorial region (between 30°S and 30°N), and coastal areas with widespread marine stratocumulus clouds. Alterskjær et al. (2012) used the cloud-92 weighted susceptibility function to find the most sensitive regions to the injection of sea-salt aerosols. 93 94 Similarly, Jones and Haywood (2012) determined the 10% of the marine regions globally most suitable for implementing MCB through an iterative method. The contributions of direct and indirect effects of 95 aerosols during the implementation of MCB are still controversial and quantitative assessment of both is 96 97 lacking.

Here, we use the two-way coupled Weather Research and Forecasting - Community Multi-scale Air Quality model (WRF-CMAQ), combined with previous studies on the region and injection strategies, to implement MCB in five open oceans worldwide. This study simulates the regional radiation and cloud responses caused by injecting sea-salt aerosols. This aims to explore the commonalities and differences in MCB implementation in different regions and to seek the optimal strategy for MCB injection.

103

104 **2. Experiments and methods**

105 **2.1 Model configuration**

106 The two-way coupled WRF (v3.4) - CMAQ (v5.0.2) model that considers both direct and indirect effects

107 of aerosols was used in this study (Yu et al., 2014). In the two-way coupled model, aerosols predicted by 108 CMAQ are able to affect clouds, radiation, and precipitation simulated by WRF in a consistent online 109 coupled manner (Wong et al., 2012). Yu et al. (2014) further extended the two-way coupled WRF-CMAQ 110 model by incorporating the aerosol indirect effects (including the first, second, and glaciation aerosol 111 indirect effects), improving the ability of the WRF-CMAQ model to predict clouds and radiation. Wang 112 et al. (2021) validated this model.

The physical schemes of the WRF model are the same as those set in Yu et al. (2014), including the 113 asymmetric convective model (ACM2) for a planetary boundary layer (PBL) scheme (Pleim, 2007), the 114 Morrison 2-moment cloud microphysics scheme (Morrison et al., 2009), the Kain-Fritsch (KF2) cumulus 115 cloud parameterization, the Rapid Radiative Transfer Model for General Circulation Models (RRTMG) 116 117 longwave and shortwave radiation schemes, and the Pleim-Xiu (PX) land-surface scheme. The meteorological initial and boundary conditions are provided by the National Center for Environmental 118 Prediction (NCEP) final analysis dataset (FNL) with a spatial resolution of 1°×1° and temporal resolution 119 of 6 h. The carbon bond gas-phase chemical mechanism (CB05) and aerosol module of AERO6 were 120 121 used in the CMAQ model. The anthropogenic emissions were taken from the Hemispheric Transport of 122 Air Pollution (HTAP V2) projects (Janssens-Maenhout et al., 2015). The biogenic emissions were estimated by the Biogenic Emissions Inventory System version 3.14 (BEISv3.14) model (Carlton and 123 Baker, 2011). Sea salt emissions were calculated online in CMAQ and were divided into open-ocean and 124 125 surf-zone emissions. In the open ocean, Gong (2003) extended the sea-salt aerosol parameterization of Monahan et al. (1986) to submicron sizes, with the emission flux being linearly proportional to the ocean 126 area covered by whitecaps. CMAQ represents the atmospheric particle distribution as the superposition 127 of three log-normal modes, the Aitken, Accumulation, and Coarse modes (Binkowski and Roselle, 2003). 128 129 The particle size distribution and the geometric standard deviation of the emitted sea-salt aerosols are adjusted to the local relative humidity before mixing with the ambient particle modes (Zhang et al., 2005). 130 The geometric mean diameter of accumulation mode sea-salt aerosols in the CMAO ranged from 0.2651 131 132 to 0.8187 µm, with the geometric standard deviation constrained between 1.76 and 1.83. Surf-zone 133 emissions are calculated using the open ocean-source function of Gong (2003), with a fixed whitecap coverage of 100% and a surf-zone width of 50 m. Kelly et al. (2010) provided a detailed description of 134 these processes. In the CMAQ model, the number concentration emission rate was calculated from the 135 136 mass emissions rate as follows:

137
$$E_{3n} = \left(\frac{6}{\pi}\right) \left(\frac{E_n}{\rho_n}\right) \tag{1}$$

$$E_0 = \frac{\sum_n E_{3n}}{D_{gv}^3 \exp\left(-\frac{9}{2}\ln^2 \sigma_g\right)}$$
(2)

where En was the mass emissions rate for species n and ρ_n was the density for that species. The sum $\sum_n E_{3n}$ was taken over all emitted species. The geometric mean diameter for mass or volume, D_{gv} , was given by $D_{gv} = D_g \exp(3 \ln^2 \sigma_g)$ from the Hatch-Choate relations for a lognormal distribution (Binkowski and Roselle, 2003). This study uses Geographic Information System software (ArcGIS) to obtain the open-ocean and surf-zone fractions for each grid within the modeling domain from shoreline information. The modeling domains of the five regions were almost entirely open ocean, with surf-zone fractions of less than 0.01%.

146 **2.2 Experimental setup**

138

147 As summarized in Table S1, the MCB geoengineering implementation areas included the globe, the 148 equator (30°S-30°N), regions with extensive coverages of marine stratocumulus clouds, and so on. Therefore, based on previous experimental designs, we use the WRF-CMAQ model to simulate the 149 injections of sea-salt aerosols in the five open ocean regions (Fig. 1c). These regions are WP and NP, 150 located in the western and northern Pacific Ocean; Equa, located in the Philippine Sea along the equator; 151 and SP and SA, located in the south Pacific and south Atlantic, respectively. The three regions, NP, SP 152 and SA, are located along the western coast of continents, are considered to have extensive coverage of 153 marine stratocumulus clouds and were the most suitable areas for implementing MCB (Alterskjær et al., 154 2012; Hill and Ming, 2012; Jones et al., 2009; Partanen et al., 2012; Stuart et al., 2013). 155

The grid numbers of WRF and CMAQ are 190×190 and 173×173, respectively, and both have a horizontal resolution of 12 km, with 29 vertical layers from the surface to about 21 km altitude. The simulation period for the WP, Equa, and NP regions in the northern hemisphere is from July 24, 2018, to September 1, 2018, while for the SP and SA regions in the southern hemisphere, the simulation period is from February 24, 2023, to April 1, 2023. The first 8 days of the model simulations are considered as the spin-up period to minimize the impacts of initial chemical conditions.

The results of the Base simulations with the model settings described above and default sea salt emissions (no aerosol injection) were obtained. As can been seen, there are significant differences in the cloud distributions for the five ocean regions in the Base simulations during the study period, with wider distributions of liquid clouds in the NP, SP, and SA regions, but fewer clouds in the WP and Equa regions (Fig. 2, first column). Cloud heights are distributed between 500–2000 m, centered at 1000 m (Fig. S1,
first column). The cloud fraction, CDNC, liquid water path (LWP), and sea-salt aerosol concentrations in
the Base simulations for each region are summarized in Table 1.

We test four different sea-salt aerosol injection strategies, wind-speed-dependent Natural×5, Windadjusted, Fixed at 10⁻⁹ kg m⁻² s⁻¹ and Fixed-wind-adjusted. All additional injected sea-salt aerosols are in the accumulation mode. In this study, the geometrical mean dry diameter of sea-salt aerosols injected into the five regions is about 0.11–0.15 μ m, and is similar for all emission scenarios.

173 **Natural**×5: Increase the emission rates of accumulation mode sea-salt aerosols by a factor of 5 (Hill 174 and Ming, 2012). This is a simple wind-speed-dependent increase. The injection rates in the five regions 175 are equivalent to $0.031-0.085\times10^{-9}$ kg m⁻² s⁻¹ (Table S2).

Wind-adjusted: Salter et al. (2008) designed a spray vessel for injecting sea-salt aerosols with a spray efficiency that was dependent on wind speed and was expected to achieve maximum spray outputs at wind speeds between 6–8 m s⁻¹. The threshold wind speed was set to 7 m s⁻¹ and the spray efficiency at lower wind speeds raised to the power of 1.5. We use the source function of Partanen et al. (2012) as follows, where *u* is the 10 m wind speed. For example, at wind of 7 m s⁻¹ the injection rate will be 0.26 × 10^{-9} kg m⁻² s⁻¹.

182
$$F_{\rm m, \, baseline} = \begin{cases} 5 \times 2.8 \times 10^{-12} \times \left(\frac{u}{1 \,{\rm m \, s^{-1}}}\right)^{1.5} \,{\rm kg \, m^{-2} s^{-1}}, & u < 7 \,{\rm m \, s^{-1}} \\ 5 \times 2.8 \times 10^{-12} \times 7^{1.5} \,{\rm kg \, m^{-2} s^{-1}}, & u \ge 7 \,{\rm m \, s^{-1}} \end{cases}$$
(3)

Fixed at 10⁻⁹ kg m⁻² s⁻¹: Unlike the previous two injection methods, the injections of sea-salt aerosols 183 at a fixed rate of 10⁻⁹ kg m⁻² s⁻¹ are not dependent on wind speed and increased uniformly over all ocean 184 grids. Injecting sea-salt aerosols at a fixed rate identified the geographic areas that were most sensitive to 185 increased sea-salt aerosols and produced the largest top-of-atmosphere (TOA) radiative perturbations 186 187 (Alterskjær et al., 2012). Many other studies have used this method (Goddard et al., 2022; Horowitz et al., 2020; Mahfouz et al., 2023). Uniform injections of sea-salt aerosols throughout the region ignored 188 aerosol transports and dispersion at the boundary. Therefore, based on the results of a fixed 10⁻⁹ kg m⁻² s⁻ 189 ¹ injection rate, we identify the geographical regions (30×50 grid points, approximately 360 km $\times 600$ 190 191 km, away from the domain boundary) in five ocean areas where the TOA radiative perturbations caused by uniform injection are the largest, and the most sensitive. Table S3 shows the locations of these sensitive 192 regions. The injection amount in the sensitive region at a fixed 10⁻⁹ kg m⁻² s⁻¹ injection rate is found to be 193 194 about 1/20 of those in the full domain.

195 Fixed-wind-adjusted: To rule out differences in radiative and cloud response due to wind

- 196 variabilities on spray rates, we perform an additional adjustment. Similar to Natural×5, the injections of
 - 197 sea-salt aerosols were also dependent on the wind speed but the integrated amounts in the region are set 198 to be equal to the case that all area had a fixed rate of 10^{-9} kg m⁻² s⁻¹ (Fixed).

199 2.3 Calculations

The calculation method related to radiation, cloud properties, and cloud radiation forcing is based on 200 201 Goddard et al. (2022), briefly described here as follows. This study focuses on the shortwave radiative flux responses at the TOA due to the injections of sea-salt aerosols, which is consistent with the definition 202 of effective radiation forcing (ERF) (Forster et al., 2007). The sea surface temperature in the model is 203 preset by NCEP-FNL, so the model's surface temperature and upward longwave radiation would not 204 respond to the increased sea-salt aerosols. The total upward shortwave radiation flux (SW TOT) at the 205 TOA is under the all-sky conditions. The responses of SW TOT to the injections of sea-salt aerosols 206 could be divided into the cloud radiation effects (SW CLD, excluding the direct effect of the aerosols) 207 208 and direct scattering effects when clouds are present (SW AER).

209

$$SW_TOT = SW_CLD + SW_AER$$
 (4)

The diagnosis of CLEAN-SKY (no aerosols) is not considered in the previous WRF-CMAQ model. So in this study, we extend this feature in the WRF-CMAQ model using the methodology of Ghan et al. (2012) by performing a double radiative call at each time step to calculate radiation variables related to CLEAN-SKY (SW_CLD). We also study the impacts of injecting sea-salt aerosols on the upward shortwave radiation flux at the TOA under the clear-sky conditions (SW_AER_CLR). At this time, only the direct scattering effect of aerosols existed, which is considered to be the maximum MSB potential generated by injecting sea-salt aerosols when there is no cloud.

Due to the different amounts of sea-salt aerosols injected in different ways, it results in different SW_{TOT} responses. Therefore, we propose the concept of MCB efficiency (E_{MCB}) to measure the relationships between the amount of sea-salt aerosol injections and the resulting radiation flux responses (Table S2).

221
$$E_{MCB} = \frac{\text{SW}_{\text{TOT} \text{ response due to injection of sea-salt aerosol (W m-2)}}{\text{Sea-salt aerosol injections (kg m-2 s-1)}}$$
(5)

This is a measure of the mass efficiency of MCB implementing in different regions, that is, how much SW_TOT responses are expected to be generated by injecting sea-salt aerosols at a rate of 1 kg m⁻² s⁻¹. $E_{MCB} = 1$ means that injecting 1 kg of sea-salt aerosols per unit time in the current study area is expected

- to produce a 1 GW (10^9 W) SW_TOT response. Note that this value (E_{MCB}) is based on model calculations under specific atmospheric conditions within the study region and is only used to analyze the sensitivities of the radiative flux to different injection methods and injection amounts.
- This study focuses on the changes in liquid clouds and evaluate the responses in cloud condensation nuclei (CCN), cloud fraction, CDNC, r_{e} , LWP, cloud optical thickness (COT), and cloud albedo due to the injections of sea-salt aerosols. These calculations are shown in Supplementary Text S1.
- Cloud radiation forcing (CRF) parameters can be used to quantify the responses of SW_CLD to changes in cloud cover or cloud albedo, defined as follows (Goddard et al., 2022):
- $CRF_{param} = \alpha_c f \tag{6}$

234 where α_c is mean cloud albedo and *f* is mean cloud fraction.

The CRF parameters can be approximated using the perturbation method as follows (Goddard et al., 2022):

237

$$\operatorname{CRF}_{param} = \alpha'_c \bar{f} + \bar{\alpha}_c f' + \alpha'_c f' \tag{7}$$

where the first term on the right-hand side indicates the changes in CRF_{param} driven by the perturbation of cloud albedo, the second term indicates the changes driven by the perturbation of cloud fraction, and the third term denotes the changes driven by the interactions of the two. The horizontal bars on α_c and fare defined as the monthly means of the Base, and the prime (') defines the monthly mean differences between the sensitivity experiments and Base. The fourth column of Fig. S23 shows that the differences between CRF_{param} and CRF'_{param} are small enough that the perturbation method can be used to approximate the CRF'_{param} .

The changes in cloud albedo are driven by multiple processes. Based on Quaas et al. (2008) and Christensen et al. (2020), Goddard et al. (2022) established the following equation to assess the relative effects of CDNC, LWP, and mean cloud fraction on the responses of SW_CLD due to the injections of sea-salt aerosols:

249
$$\frac{\Delta \alpha}{\Delta \ln \text{AOD}} = f \Delta \alpha_c (1 - \alpha_c) \left(\frac{1}{3} \frac{\Delta \ln \text{CDNC}}{\Delta \ln \text{AOD}} + \frac{5}{6} \frac{\Delta \ln \text{CLWP}}{\Delta \ln \text{AOD}} + \frac{\Delta \ln f}{\Delta \ln \text{AOD}} \right)$$
(8)

where α is the planetary albedo, Δ represents the difference in monthly average results between sensitivity experiments and Base simulations, and α_c is the cloud albedo. The three terms inside the right parenthesis represent the relative contributions of Twomey effect, LWP effect, and cloud fraction effect, respectively, with the latter two related to the second aerosol indirect effect (Albrecht, 1989).

254 The perturbations by generating three ensemble members for each experiment in each region were

added. The results of all sensitivity experiments are compared to those of Base simulations. Unless otherwise specified, all results in this study are shown as overall regional monthly averages of the ensemble.

258

259 **3. Results**

260 **3.1** The impacts of different injection strategies on shortwave radiation at the TOA.

In modeling studies, variations in methods used to increase sea-salt aerosols may lead to different 261 262 conclusions, and these variations may be one of the reasons for differences in the assessments of MCB potentials in the previous studies. In this study, sea-salt aerosols injected in different strategies (with dry 263 diameters of about 0.11–0.15 μ m, Fig. 1a) increase the SW_TOT at the TOA by 0.07–25 W m⁻² in the 264 five ocean regions (Fig. 3a). The Natural×5 and Wind-adjusted strategies, which rely on wind speeds. 265 inject sea-salt aerosols of 0.031–0.085 and 0.18–0.21 \times 10⁻⁹ kg m⁻² s⁻¹ into the five regions, respectively, 266 and result in SW TOT variations of 0.07–2.1 and 1.4–8.4 W m⁻², respectively (Fig. 3a and Table 2). 267 Uniformly injections of sea-salt aerosols at a fixed rate of 10⁻⁹ kg m⁻² s⁻¹ results in SW TOT changes of 268 11-25 W m⁻² in the five regions. The three continental west coast stratocumulus regions of NP, SP, and 269 SA have the most significant SW_TOT responses, all exceeding 20 W m⁻², while the SW_TOT responses 270 in the WP and Equa regions are 18 and 11 W m⁻², respectively. 271

Injecting the same amount of sea-salt aerosols results in substantial variations in SW TOT responses 272 across the different regions (Fig. S2). The sea-salt aerosols sprayed in the Fixed-wind-adjusted 273 experiments are also dependent on wind speed, but the amount of emission rate integrated in the full 274 domain is consistent with the fixed rate of 10⁻⁹ kg m⁻² s⁻¹, ruling out the differences caused by the amount 275 276 of injected sea-salt aerosols. Although both strategies inject the same amounts of sea-salt aerosols, the 277 SW TOT responses they produce are significantly different. The Fixed-wind-adjusted strategy results in SW TOT changes of 5.0–20 W m⁻² in the five regions (Fig. 3a), indicating that the shortwave radiation 278 flux changes caused by wind-speed-dependent injections are smaller than those caused by uniformly 279 280 injections, and showed regional differences.

Figure 3b shows the E_{MCB} values of different sea-salt injection strategies in the five regions. Overall, MCB implementation is more efficient in the NP, SP, and SA regions, while it is less efficient in the WP and Equa, which is similar to the previous SW_TOT response results. E_{MCB} also varies for different injection strategies. In the NP, SP, and SA regions, the E_{MCB} values of the Natural×5 and Wind-adjusted strategies with relatively small injection amounts are higher than the other two strategies with large injection amounts. At the same injection amount, injecting at a fixed rate shows higher E_{MCB} relative to injections depending on wind speed, as consistently shown in all five regions (Fig. 3b). Since the number flux of aerosols increased with the decreases of the injected aerosol particle size for the same mass flux, we examined the MCB efficiency in units of aerosol number concentration (Fig. S3). The results show that the number efficiency of MCB is proportional to the injection rate of aerosol number (Fig. S3c). In the same quality injected, the aerosol number varies greatly (Fig. S3d).

The productions of sea-salt aerosols in nature are strongly correlated with wind speed, and most 292 293 models associated sea-salt aerosol emissions with wind speed (Ahlm et al., 2017; Grythe et al., 2014). Injection strategies depending on wind speed make the distributions of added sea-salt aerosols closer to 294 295 the natural distributions. In natural environments, sea-salt aerosol emissions in strong-wind areas (e.g., storm or typhoon areas) and surf zones are usually much larger than in weak-wind areas. Therefore, 296 injection strategies depending on wind speed concentrate the added sea-salt aerosols in strong-wind areas 297 and surf zones, while the weak-wind regions increase relatively little sea-salt aerosols (Fig. S4). Injecting 298 299 uniformly at a fixed rate in the model will result in a large increase of sea-salt aerosols in places with 300 originally low aerosol concentrations (e.g., weak-wind regions). Therefore, when using models to simulate the injections of sea-salt aerosols by increasing the emission rate, it is necessary to consider the 301 impacts of different injection methods on the distributions of sea-salt emissions. Using a uniformly 302 303 increasing method independent of wind speed can not only avoid the situation of a smaller increase in sea-salt emissions in regions with lower wind speeds, but can also identify the geographical areas most 304 sensitive to the increased sea-salt aerosols and producing the largest TOA radiation perturbations 305 (Alterskjær et al., 2012). 306

Injecting sea-salt aerosols in the sensitive areas with the same uniform injections $(10^{-9} \text{ kg m}^{-2} \text{ s}^{-1})$, the 307 injection rate is about 1/20 of the full domain injection) results in changes of 0.49-3.4 W m⁻² in SW TOT 308 in the five ocean regions (Table S2). The SW TOT responses are the largest in the SP region, at 3.4 W m⁻ 309 ², and 2.7 and 1.7 W m⁻² in the NP and SA regions, respectively, while they were only 0.49 and 0.83 W 310 m⁻² in the WP and Equa regions, respectively. The injected sea-salt aerosols produced SW TOT changes 311 of 5.11–14.3 W m⁻² in the sensitive areas (Fig. 1b). Similarly, the increases in SW TOT in the SP, SA, 312 and NP regions all exceeded 9 W m⁻², with the highest in the SP region at 14.3 W m⁻². In the WP and Equa 313 regions, although the increases in SW TOT are only 5.11 and 5.26 W m⁻², respectively. Considering that 314 the original intents of MCB or MSB design are regional application (hurricane mitigation, coral reef 315

- 316 protection and polar sea ice recovery) (Latham et al., 2014), choosing to inject sea-salt aerosols in the 317 sensitive areas could achieve the corresponding cooling goals within the region, and also affected larger
 - areas through the diffusions and transports of aerosols.

319 **3.2** Characterization of the radiation responses.

SW TOT responses are defined as the sum of the upward shortwave radiation flux response at the TOA 320 321 generated by the combined effects of the direct scattering effect of aerosols (SW AER) and cloud radiative effect (SW CLD) after injecting sea-salt aerosols. Figure 4 shows the contributions of SW AER 322 and SW CLD responses in the SW TOT produced by different injection strategies in the five ocean 323 regions. The majority of the SW TOT radiative flux response due to the lower mass injection Natural×5 324 and Wind-adjusted strategies is caused by the SW CLD response (Fig. 4a). In the NP, SP, and SA regions, 325 326 the contributions of SW CLD exceed 70%, suggesting that sea-salt aerosols injected at these locations increase the SW TOT mainly by affecting clouds through indirect effects. In the Equa, the responses of 327 SW TOT are entirely caused by SW AER. The proportion of SW AER produced by the uniform 328 injection of sea-salt aerosols at a fixed rate of 10⁻⁹ kg m⁻² s⁻¹ continued to increase (Fig. 4c). In the WP, 329 Equa, and SP regions, the proportion of SW AER exceeded that of SW CLD. In the SA region, SW CLD 330 and SW AER are almost equal, while in the NP region, the SW CLD response is 13 W m⁻², still greater 331 than SW AER (9.8 W m⁻²). This is because there is a saturation phenomenon in the cloud response to 332 aerosols injections (discussed below), and the NP, SP, and SA regions provide more SW CLD responses, 333 334 while the cloud responses in the WP and Equa regions saturate and no longer increase. The results of Fixed-wind-adjusted case show that, at the same injection amount, the SW AER responses caused by the 335 injection strategy relying on wind speed is significantly smaller than those of the method with fixed-rate 336 uniform injection, while the disparity in SW CLD responses is minimal. This is because the injection 337 strategy relying on wind speed distributed most of the increased sea-salt aerosols to areas with already 338 high emissions, such as strong-wind areas and surf zones, where the excess marine aerosols have already 339 saturated the cloud responses, resulting in minor changes in SW CLD. In areas with weak winds, the 340 potentials for direct aerosol scattering are not fully exploited due to the relatively small amounts of sea-341 342 salt aerosols injected, leading to a lower SW AER response.

Figures S5 and S6 show the spatial distributions of SW_CLD and SW_AER responses resulting from different injection methods in the five ocean regions. The SW_CLD responses are stronger in the three regions of NP, SP, and SA, while they are weaker in the regions of WP and Equa, and in some grids they even lead to a reduction of the upward shortwave radiation (Fig. S5). The spatial distributions of the SW_CLD responses exhibit noticeable discontinuity, reflecting significant regional differences in the non-uniform distributions of clouds and their impacts on shortwave radiation at the TOA. The effect of cloud properties on SW_CLD will be shown in Section 3.5. Due to the influences of various complex factors on cloud formations and distributions, simulation results related to clouds show significant spatial variabilities. This might be the result of the combined effects of local meteorological conditions and changes in cloud physical properties caused by sea-salt aerosol injections.

In contrast, the spatial distributions of the SW AER response are smoother, leading to consistent 353 354 increases in upward shortwave radiation at the TOA in all ocean regions (Fig. S6). This indicates smaller spatial limitations in the distributions of aerosol particles, allowing direct scattering effects to take place 355 356 everywhere. The direct scattering effect of aerosols is primarily related to the concentrations and physical properties of the particles (discussed below), unlike clouds, which are influenced by multiple variables. 357 These results suggest that when implementing geoengineering measures, it is essential to 358 comprehensively consider the interactions between aerosols and clouds, as well as their different response 359 360 patterns in various regions. Furthermore, the high spatial variabilities of cloud radiation effects emphasize 361 the need for improved resolution in future model studies of cloud-aerosol interactions.

The SW CLD response resulting from the injection of sea-salt aerosols in the sensitive areas of five 362 ocean regions exhibits significant spatial differences. The SW CLD response is larger than the SW AER 363 response in the sensitive areas of NP, SP, and SA, indicating that the changes in SW TOT are mainly 364 driven by the cloud radiation response (Fig. 5). In contrast, the SW CLD response is smaller in the WP 365 and Equa regions. This regional difference is similar to that observed with uniform injection across the 366 entire region. The SW AER response shows consistent results in all areas, resulting in a radiation 367 response change of 3.58–5.44 W m⁻² within the injection areas. In the WP and Equa, the variations in 368 SW TOT are primarily driven by the direct scattering effects of aerosols. Aerosols can have a greater 369 impact on radiation responses outside the sensitive areas through transports and diffusions, reaching up 370 to three times the total radiation within the sensitive areas (Fig. 6). In all regions except WP, the total 371 372 SW CLD response outside the sensitive region was about 270%–408% higher than inside. In WP, the SW CLD response outside the sensitive area has a negative effect. The SW CLD responses in NP, SP, 373 and SA extend to the west and northwest of the injection by the prevailing winds, indicating that clouds 374 375 in these areas are affected by the injection of sea-salt aerosols (Fig. 5). Changes in cloud microphysical properties will be presented later. The SW CLD variations in other directions are not uniform, and there 376

is negative SW_CLD responses in some grids, which again reflects the spatial complexities of cloud radiation effects. The direct scattering effects of aerosols on areas outside the sensitive region is reflected in a widespread increase in upward shortwave radiation at the TOA. The total SW_AER responses outside the sensitive areas in the five ocean regions are approximately 160%–281% higher than inside, but lower than the impacts of SW_CLD responses outside the sensitive areas. There are consistencies in the spatial distributions of SW_AER and SW_CLD responses.

383 3.3 Saturation of the cloud radiative responses.

Figure 7 shows that under low levels of sea-salt aerosol injections, radiation response changes are mainly 384 driven by SW CLD responses. As the injected sea-salt aerosols increased, the SW CLD responses 385 gradually reach saturation. After reaching a certain injection level, the increases of SW CLD responses 386 387 stabilize at its maximum value and no longer increases with further injections. The SW CLD responses show large differences in the five ocean regions, and the different shapes and slopes of the curves indicate 388 389 that the cloud radiative forcing responses to the sea-salt aerosol injections are different in each region. This might be due to variations in cloud types, cloud amounts, and atmospheric conditions in the different 390 regions. In the NP, SP, and SA, the SW CLD responses exceed 10 W m⁻², while in WP, it saturates at 5 391 W m⁻². In Equa, when the sea-salt aerosol injection rate is 10^{-9} kg m⁻² s⁻¹, the SW CLD response is 0.5 392 W m⁻², and even when the injection doubled, the SW CLD response remains at 0.5 W m⁻². This implies 393 that the SW TOT at Equa was almost exclusively from the contributions of the direct scattering effects 394 395 of aerosols.

In contrast to SW CLD, the SW AER responses increase linearly with the injections of sea-salt 396 aerosols ($R^2 > 0.99$). As the injection increases, the contributions of SW_AER to SW_TOT gradually 397 increase, surpassing the SW CLD responses, and show the same trends across the five regions. This 398 implies that at higher injection levels, the contributions of SW CLD to total radiation change saturated, 399 and cloud properties no longer significantly change. At this point, sea-salt aerosols primarily affect 400 radiation through direct scattering effects, and the aerosol particles' ability to scatter solar radiation 401 continues to increase with the increases in aerosol quantities. In some cloud-free regions or weather 402 403 conditions, injected sea-salt aerosols are still able to function through direct scattering.

There exists a specific injection level at which the SW_CLD and SW_AER responses are equal. In the NP region, when the injection level is approximately 1.55×10^{-9} kg m⁻² s⁻¹, both SW_CLD and SW_AER responses are 15 W m⁻². In the SP and SA, these levels are about 0.67×10^{-9} kg m⁻² s⁻¹ and 1×10^{-10} ⁹ kg m⁻² s⁻¹, respectively. While in WP, the responses were already equal when the injection amount was 0.15×10⁻⁹ kg m⁻² s⁻¹. Since there is a saturation of the cloud radiation effects, E_{MCB} decreases with the increases in sea-salt aerosol injection amounts (Fig. 7, red dashed line). This can also explain the higher E_{MCB} of the Natural×5 and Wind-adjusted strategies with relatively low injection amounts (Fig. 3b). Therefore, wind-dependent injection strategies lead to the injection of large amounts of sea-salt aerosols in certain areas with high wind speeds, leading to saturation of cloud radiation effects, which might affect the performances of MCB in the simulations of regional and global models.

When less sea-salt aerosols are injected, both SW_CLD and SW_AER responses contribute to the changes of SW_TOT. As the injection amounts increase, the SW_CLD responses saturate, and the increases in SW_TOT depends on the increases in SW_AER responses, leading to a decrease in E_{MCB} (Fig. 7) Therefore, implementing geoengineering with sea-salt aerosol injections requires considering local atmospheric conditions and balancing the relationships between cooling goals and sea-salt injection efficiencies.

Under clear and cloudless conditions, injecting sea-salt aerosols could still increase the SW TOT 420 421 through direct scattering, and this effect exceeds those of aerosol direct scattering when clouds are present. 422 The variation of the upward shortwave radiation flux at the TOA under the clear-sky conditions (SW AER CLR) does not exhibit significant regional heterogeneity across the ocean areas (Figs. 5 and 423 S7), suggesting that the contribution of direct aerosol scattering is more uniform globally when 424 considering the effects of sea-salt injections on the Earth's radiation budget. The SW AER CLR 425 responses are also linearly correlated with the injection of sea-salt aerosols ($R^2 > 0.99$), and it exceeds the 426 SW AER responses (Fig. 7). This is because cloud layers also scatter and absorb solar radiation, so this 427 scattering effect is more significant under clear sky conditions. It is reflected that in regions with strong 428 429 cloud radiation effects, such as the NP, SP, and SA regions, the differences between the SW AER and 430 SW AER CLR responses are also larger (Fig. 7). When injecting sea-salt aerosol in sensitive areas, the spatial distributions of SW AER CLR and SW AER responses are highly consistent (Fig. 5). Therefore, 431 injecting sea-salt aerosol under conditions of low cloud covers or clear skies also increases the upward 432 433 shortwave radiation flux at the TOA.

434 **3.4 Factors affecting the radiation effects.**

Uniform injections of 10^{-9} kg m⁻² s⁻¹ sea-salt aerosols lead to an increase in aerosol optical depth (AOD) of 0.20–0.37 in all regions (Fig. 8). The distributions of AOD within the regions are not uniform due to

aerosol transports and diffusions, with some areas showing an increase in AOD of over 0.6. Injecting sea-437 salt aerosols in sensitive areas lead to an AOD increase of 0.077–0.12, while outside the injection areas, 438 439 AOD gradually decreases as the aerosols transport and disperse. With the increases in sea-salt aerosol injections, AOD shows a linear increase within a certain range in all five ocean regions ($R^2 > 0.997$, Fig. 440 9a). There is a strong correlation between the AOD changes caused by sea-salt injection and the SW AER 441 responses. When sea-salt aerosols are uniformly injected across the entire region, the correlation 442 coefficients between AOD and SW AER responses in the five ocean areas are greater than 0.94, and 443 when injected in sensitive areas, the correlation coefficients are greater than 0.99 (Fig. S8). The optical 444 445 properties of injected aerosols are described in Supplementary Text S2. In general, the injected sea-salt aerosols scatter sunlight more efficiently than absorb it, causing solar radiation to be reflected back into 446 447 space and tend to scatter more uniformly or backward rather than forward.

In the regions with higher cloud cover, such as NP, SP, and SA, injected sea-salt aerosols significantly 448 increases cloud fraction (Fig. 2, third column and Table 1), leading to the formations of more clouds or 449 expanding the coverage, vertical thickness and lifetime of existing clouds (Goddard et al., 2022). The 450 451 injection of sea-salt aerosols in sensitive areas have similar results, where cloud fractions increase both 452 inside the injection areas and in the regions affected by aerosol transports and diffusions (Fig. S13). Taking the SP region as an example, Fig. 10 demonstrates that uniformly injections of 10⁻⁹ kg m⁻² s⁻¹ sea-453 salt aerosols significantly increases the CDNC. More cloud droplets capture more water vapor, leading to 454 455 an increase in LWP. Additionally, the increases in cloud thickness also contribute to the increase in LWP. The increase in CDNC decreases the mean r_e by 8.9 μ m (~ -37%), increases the COT by more than 220%, 456 and ultimately increases the mean cloud albedo over the region by 0.19 (~64%). Similarly, injecting sea-457 salt aerosols in the NP and SA regions lead to average cloud albedo increases of 0.17 and 0.20, respectively, 458 459 while in the WP and Equa, the increases are 0.15 and 0.13, respectively (Figs. S14–S17). The injection of 460 sea-salt aerosols within the sensitive areas has less effect on cloud microphysical properties than the whole region injections. This is because when sea-salt aerosols are injected across the entire region, the 461 462 surrounding sea-salt aerosols affect the sensitive areas through transports, resulting in an enhanced 463 cumulative effect on cloud microphysical properties in the sensitive areas. Injecting sea-salt aerosol in the sensitive area of the SP affected clouds in the surrounding region through transports, increases the 464 average cloud albedo across the entire area by 0.032 over the entire region and by 0.12 within the sensitive 465 466 regions, which is less than the effects of injection across the entire area (Fig. S18). Similarly, injecting sea-salt aerosols in the sensitive areas of other ocean regions lead to average cloud albedo increases of 467

0.015–0.024 across the entire area, with increases of 0.11 in the sensitive areas of the SP and SA regions,
and increases of 0.090 and 0.10 in the WP and Equa, respectively (Figs. S19–S22).

470 **3.5 Drivers of SW_CLD responses.**

The cloud radiation forcing (CRF) parameters are used to calculate the effects of changes in cloud cover 471 and cloud albedo on the SW CLD responses due to the injections of sea-salt aerosols. Figure S23 472 illustrates the increase in the CRF parameter coinciding with the increases in the SW CLD responses 473 474 after uniform injection of sea-salt aerosols in the five regions (Fig. S5, third row). The results are similar for injections in the sensitive areas (Fig. S24, third column, and Fig. 5, first column). The CRF'_{param} 475 calculated using the perturbation method indicates that in the five ocean regions, CRF'_{param} is primarily 476 driven by perturbations in cloud albedo (Fig. S25, first column), and it significantly surpasses the changes 477 478 in cloud fractions and their interactions. Cloud albedo changes explain over 70% of the CRF'_{param} in all five regions except the Equa. The contribution of cloud fraction changes ranges from 13.9% to 23.7%, 479 480 while the interactions between the two factors account for only about 10% (Fig. S25, second and third columns). The results are similar for injections in sensitive regions, where changes in cloud albedo 481 account for 58.8%–99.4% of the CRF'_{param}, followed by changes in cloud fractions, with the smallest 482 contributions from their interactions (Fig. S26). 483

Figure 11 evaluates the relative effects of Twomey, LWP, and cloud fractions on the SW CLD 484 responses after uniformly injecting sea-salt aerosols in five ocean regions. The results indicate that 485 changes in CDNC (Twomey effect) and LWP are the main drivers of SW CLD responses, while changes 486 487 in cloud fraction contribute minimally to the SW CLD responses. Except for the Equa region, changes in CDNC and LWP account for 48.4%–52.5% and 39.0%–41.7% of the SW CLD changes, respectively, 488 with cloud fraction changes contributing to less than 10.0% (Fig. 11). The results are similar for injections 489 490 in sensitive areas, with changes in CDNC and LWP contributing similarly and more than changes in cloud fractions to SW CLD (Fig. S27). The changes in SW CLD responses after aerosol injections in the 491 sensitive areas of Equa are mainly contributed by LWP effects (~70%). 492

493 Uniform injections of sea-salt aerosols at a rate of 10^{-9} kg m⁻² s⁻¹ produce susceptibilities ($\frac{\Delta \alpha}{\Delta \ln AOD}$) 494 ranging from 0.00030 to 0.0035 in the five regions, with corresponding spatial distributions shown in Fig. 495 11. NP, SP, and SA regions exhibit cloud responses that are more sensitive to aerosol injections in most 496 of the region, with susceptibilities ranging from 0.0028 to 0.0035. The Equa shows the lowest 497 susceptibility, indicating that the system is less responsive to variations in aerosol injections. It is

noteworthy that although the average susceptibility in the WP region is 0.0013, the higher susceptibility 498 values are concentrated in the north of 35°N, where the average susceptibility is 0.0026, similar to those 499 500 of the SP region, suggesting that clouds here are more susceptible to aerosol injections. Injecting sea-salt aerosols in sensitive areas mostly results in cloud that are located outside the sensitive areas (Fig. S27). 501 Injecting sea-salt aerosols in the sensitive areas of SP and SA have a greater impact on the northwest. In 502 the sensitive areas of NP, injecting sea-salt aerosols have a larger impact on the west. In the WP, the 503 injection of sea-salt aerosols into the sensitive area does not fully reflect its susceptibility because we 504 choose to calculate the sensitive areas away from the boundary, and the greatest susceptibilities in the WP 505 506 region happens to be in the northern part of the region near the boundary.

507 **4. Discussions and conclusions**

508 Many studies have discussed the contributions of both the direct and indirect effects of MCB. Some studies suggest that MCB primarily relies on the indirect effects, as originally conceived, i.e., injecting 509 510 aerosols to brighten clouds (Jones and Haywood, 2012; Latham et al., 2012). Other studies proposed that the direct scattering effects of aerosols may be more important (Ahlm et al., 2017; Kravitz et al., 2013; 511 Mahfouz et al., 2023; Niemeier et al., 2013; Partanen et al., 2012). Our results indicate that the 512 513 importances of both aerosol direct and indirect effects during MCB implementation depend on the injection strategies and the choice of injection regions. In cases of low sea-salt aerosol injections or the 514 early stage of MCB implementations, changes in radiative response are mainly driven by indirect effects, 515 516 causing clouds to brighten easily. As the injection of sea-salt aerosol increases, the radiative effect on 517 clouds saturates, and the clouds are difficult to brighten. In contrast, the direct effect continues to increase linearly, leading to a subsequent decrease in the efficiencies of MCB. Partanen et al. (2012) first 518 considered the relative importance of aerosol direct and indirect effects in MCB and preliminarily found 519 the saturated non-linear phenomenon of indirect effects at high CDNC, as well as the linear relationships 520 between direct effects and injection amounts. Haywood et al. (2023) also found a decrease in MCB 521 efficiency with increasing aerosol injections. Regions initially susceptible to modification gradually 522 became less susceptible, and aerosol direct radiation effects dominated. Other use General Circulation 523 524 Model (GCM) studies also found similar results (Alterskjær and Kristjánsson, 2013; Rasch et al., 2024; 525 Stjern et al., 2018). This study highlights and quantifies these findings in a regional model for the first 526 time, showing the changing trends of direct and indirect effects with injection amounts in the different ocean regions. Also due to the higher resolution of the regional model, this study provides more detailed 527

cloud component changes due to sea-salt aerosol injection. The best results are obtained in regions with 528 persistent stratocumulus clouds (e.g., the oceans along the west coast of the continent), where the injected 529 530 sea-salt aerosols work together through both direct and indirect effects. However, in cloud-free or less cloudy regions, MCB implementation can achieve the goal of reflecting more sunlight through the direct 531 scattering effect of aerosols. Considering the uncertainty in the model's resolution of clouds and the fact 532 that, in reality, the cloud distributions are also greatly influenced by the local meteorological conditions, 533 the direct scattering effects of sea-salt aerosols on MCB contributions are relatively certain. Therefore, in 534 cloud-free or less cloudy regions, the direct effect of aerosols becomes more important. 535

536 In the early stages of Earth-System modeling studies, the MCB processes are often simulated by presetting CDNC = 375 or 1000 cm⁻³ in the lower regions of the ocean (Jones et al., 2009; Latham et al., 537 2008; Rasch et al., 2009). However, many follow-up studies have suggested that injections of sea-salt 538 aerosols were difficult to produce a uniform CDNC field due to aerosol dilutions, depositions, and the 539 dependences of the spray rate on wind speed. The CDNC is highly variable spatially, and studies have 540 even reported reductions in CCN and CDNC caused by the injections of sea-salt aerosols (Alterskjær et 541 542 al., 2012; Korhonen et al., 2010; Pringle et al., 2012). In this study, after injecting accumulation mode sea-salt aerosols at a rate of 10⁻⁹ kg m⁻² s⁻¹, the average CDNC concentrations for five ocean regions range 543 from 60.2 to 100 cm⁻³, and the spatial distributions are uneven (Fig. 10 and Figs. S14–S17). Figure 9b 544 indicates that the CCNs in the five regions increase linearly ($R^2 = 1$) with increasing sea-salt aerosol 545 injections, but not all of the CCNs are converted to cloud droplets. After doubling the injection amounts, 546 the regional average CDNC is 84.8–130 cm⁻³, with only some grid points exceeding 200 cm⁻³ within the 547 regions. This implies that injecting more sea-salt aerosols at this point does not result in more cloud 548 droplets, and the conversion of CCN into cloud droplets is less efficient, which slows the CDNC growths 549 550 and tends to saturation (Fig. 9c). Alterskjær et al. (2012) similarly injected sea-salt aerosols at a rate of 10⁻⁹ kg m⁻² s⁻¹ and found that despite emitting sea-salt mass 70 times larger than suggested by Latham et 551 al. (2008), the average CDNC over the ocean was below their assumed value of 375 cm⁻³. This is mainly 552 due to increased competitive effects, decreased maximum supersaturations, inhibitions of aerosol 553 554 activations, and closures of SO₄ nucleation, resulting in reduced effectiveness of sea salt injections. Notably, however, Wood (2021) found that decreased activation due to competition may be overestimated 555 in the Abdul-Razzak and Ghan activation parameterization used in many GCMs relative to a parcel model. 556 557 When Partanen et al. (2012) injected sea-salt aerosols in a Wind-adjusted way (injection amount different from this study), they found the CDNC values of 596, 650, and 784 cm⁻³ in the NP, SP, and SA regions, 558

respectively. Injecting smaller-sized sea-salt aerosols even yields CDNC values exceeding 1000 cm⁻³. 559 They conclude that such high values are mainly due to the model's overestimation of the sizes and 560 561 solubilities of accumulated mode particles, with some non-activated particles forming cloud droplets. Hill and Ming (2012) increased the concentrations of sea-salt aerosols by a factor of five, resulting in an 562 average CDNC increasing from 68 to 148 cm⁻³ between 850–925 hPa. It is noteworthy that Hill and Ming 563 (2012) increased all modes of sea-salt aerosols. Many studies have reported that selecting the appropriate 564 injection particle size is crucial for MCB (Andrejczuk et al., 2014; Hoffmann and Feingold, 2021; 565 Partanen et al., 2012), and injecting Aitken and coarse modes may even lead to a positive forcing with 566 567 CDNC decreasing (Alterskjær and Kristjánsson, 2013). However, Wood (2021) argued that particles with a geometric mean dry diameter of 30-60 nm were most effective in brightening cloud layers, and Goddard 568 569 et al. (2022) similarly found that injecting Aitken mode sea-salt aerosols generated larger radiative flux changes compared to accumulation mode (8.4 W m⁻² versus 3.1 W m⁻²). There are still considerable 570 discussions about choosing the appropriate aerosol particle sizes during the implementation of MCB, with 571 different models and parameterization schemes providing different recommendations. The sensitivity of 572 573 MCB to particle size is not considered in this paper and was left for future research.

In this study, the injection of 10⁻⁹ kg m⁻² s⁻¹ accumulation mode sea-salt aerosols increases cloud 574 albedo in the five ocean regions by 0.13-0.20, with a maximum of more than 0.3. After doubling the 575 576 injection amounts, the regional average cloud albedo could reach 0.45-0.55, representing a cloud albedo 577 change of 0.15–0.24 (Fig. 9d). These values achieve the targeted cloud albedo change as envisioned in previous studies. Bower et al. (2006) suggested that to compensate for the warming associated with 578 doubling atmospheric CO₂ concentrations, a cloud albedo change of 0.16 was needed in three 579 stratocumulus cloud regions (off the west coast of Africa and North and South America, representing 3% 580 581 of global cloud cover). Wood (2021) proposed seeding Aitken mode particles in approximately 9% of the 582 ocean to achieve a corresponding cloud albedo increase of 0.16. It was also suggested that injecting seasalt aerosols in a clean, undisturbed state would produce more brightening. Fig. 9d confirms this finding, 583 indicating that clouds are more likely to brighten in the early stages of sea-salt aerosol injection, and the 584 585 efficiency of cloud brightening decreases with increasing injection amounts. Goddard et al. (2022), simulating injecting accumulation mode sea-salt aerosols in the central Gulf of Mexico, achieved a 586 simulated cloud albedo change of approximately 0.1 in the main impact region, while switching to Aitken 587 588 mode injection resulted in a cloud albedo change of up to 0.35. For the global implementation of MCB, global cloud albedo increases of 0.02 (Bower et al., 2006), 0.062 (Latham et al., 2008), or 0.074 (Lenton 589

and Vaughan, 2009) were estimated.

The contributions of the change in cloud fractions to the SW CLD responses in this study are small, 591 592 which is consistent with the results of Goddard et al. (2022). However, many observational studies indicate that the contribution of cloud fraction to the shortwave radiative forcing should be similar to 593 those of the CDNC and LWP (Chen et al., 2014; Rosenfeld et al., 2019). Goddard et al. (2022) believe 594 that this was due to the fact that the regional atmosphere was wetter during the simulation periods and 595 that the relative contributions of changes in cloud fraction to the SW CLD response would be expected 596 to increase in drier months. Three of the five ocean regions in this study, SA, SP, and NP are much drier 597 and more stable than the Gulf of Mexico simulated by Goddard et al. (2022) (Fig. S28). Furthermore, 598 when we switch to conducting the experiments again in the dry months of the same year, the contribution 599 600 of cloud fraction to SW CLD did not change much, remaining at ~10% (Fig. S28). We believe that this might be a difference due to the parameterization scheme or resolution of the model. Liu et al. (2020) 601 simulated with WRF-Chem model and found that the cloud fraction susceptibilities to aerosols in 602 Morrison scheme and the Lin scheme were only about half of those observed by Moderate Resolution 603 Imaging Spectroradiometer (MODIS). The neglected sub-gridded clouds in the 12-km resolution 604 605 simulations might lead to an underestimation of the radiative effects of clouds (Yu et al., 2014). In addition, cloud fractions are more commonly underestimated in the model (Glotfelty et al., 2019), and using an 606 updated parameterization scheme that accounts for sub-grid condensation might improve the model's 607 608 ability to resolve clouds (Zhao et al., 2023). The effects of finer resolution and more parameterization schemes on aerosol-cloud interactions still need to be verified. Considering the difficulties of modeling 609 to accurately capture the effects of cloud fractions on radiation, the actual effects of MCB may be 610 611 underestimated.

612 This study provides quantifiable data on cloud and radiation changes for the implementation of MCB over the regional oceans, and an optimization scheme on the injection strategy by adjusting the injection 613 614 amounts and selecting sensitive areas. It is noteworthy that different parameterization schemes, models, 615 and resolutions can influence results, especially the cloud feedback on the injected sea-salt aerosols, 616 which is a major reason for discrepancies between models (Stjern et al., 2018). In Earth-system model studies, there has been a rich discussion of the climate and ecological impacts of the MCB with the same 617 framework under the Geoengineering Model Intercomparison Project (GeoMIP) (Rasch et al., 2024). 618 619 However, there is still a lack of a unified framework for mid-scale MCB research.

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622 Data and code availability

- 623 The computational code for cloud and radiation can be found in the code publicly available from Goddard
- et al. (2022). The model results are available upon request.
- 625 Supplemental information.
- 626 The supplementary information related to this article is available online.
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637 **References**

- Ahlm, L., Jones, A., Stjern, C. W., Muri, H., Kravitz, B., and Kristjánsson, J. E.: Marine cloud brightening as effective
 without clouds, Atmospheric Chemistry and Physics, 17, 13071–13087, https://doi.org/10.5194/acp-17-13071-2017,
 2017.
- Albrecht, B. A.: Aerosols, Cloud Microphysics, and Fractional Cloudiness, Science, 245, 1227–1230,
 https://doi.org/10.1126/science.245.4923.1227, 1989.
- Alterskjær, K. and Kristjánsson, J. E.: The sign of the radiative forcing from marine cloud brightening depends on both
 particle size and injection amount, Geophysical Research Letters, 40, 210–215, https://doi.org/10.1029/2012GL054286,
 2013.
- Alterskjær, K., Kristjánsson, J. E., and Seland, Ø.: Sensitivity to deliberate sea salt seeding of marine clouds –
 observations and model simulations, Atmospheric Chemistry and Physics, 12, 2795–2807, https://doi.org/10.5194/acp12-2795-2012, 2012.
- Andrejczuk, M., Gadian, A., and Blyth, A.: Numerical simulations of stratocumulus cloud response to aerosol
 perturbation, Atmospheric Research, 140–141, 76–84, https://doi.org/10.1016/j.atmosres.2014.01.006, 2014.
- Binkowski, F. S. and Roselle, S. J.: Models-3 Community Multiscale Air Quality (CMAQ) model aerosol component 1.
 Model description, Journal of Geophysical Research: Atmospheres, 108, https://doi.org/10.1029/2001JD001409, 2003.
- Bower, K., Choularton, T., Latham, J., Sahraei, J., and Salter, S.: Computational assessment of a proposed technique for
 global warming mitigation via albedo-enhancement of marine stratocumulus clouds, Atmospheric Research, 82, 328–

- 655 336, https://doi.org/10.1016/j.atmosres.2005.11.013, 2006.
- Carlton, A. G. and Baker, K. R.: Photochemical Modeling of the Ozark Isoprene Volcano: MEGAN, BEIS, and Their
 Impacts on Air Quality Predictions, Environ. Sci. Technol., 45, 4438–4445, https://doi.org/10.1021/es200050x, 2011.
- Chen, Y.-C., Christensen, M. W., Xue, L., Sorooshian, A., Stephens, G. L., Rasmussen, R. M., and Seinfeld, J. H.:
 Occurrence of lower cloud albedo in ship tracks, Atmospheric Chemistry and Physics, 12, 8223–8235,
 https://doi.org/10.5194/acp-12-8223-2012, 2012.
- Chen, Y.-C., Christensen, M. W., Stephens, G. L., and Seinfeld, J. H.: Satellite-based estimate of global aerosol–cloud
 radiative forcing by marine warm clouds, Nature Geosci, 7, 643–646, https://doi.org/10.1038/ngeo2214, 2014.
- 663 Christensen, M. W., Jones, W. K., and Stier, P.: Aerosols enhance cloud lifetime and brightness along the stratus-to-664 cumulus transition, Proceedings of the National Academy of Sciences, 117, 17591-17598, 665 https://doi.org/10.1073/pnas.1921231117, 2020.
- Forster, P., Ramaswamy, V., Artaxo, P., Berntsen, T., Betts, R., Fahey, D. W., Haywood, J., Lean, J., Lowe, D. C., Raga,
 G., Schulz, M., Dorland, R. V., Bodeker, G., Etheridge, D., Foukal, P., Fraser, P., Geller, M., Joos, F., Keeling, C. D.,
 Keeling, R., Kinne, S., Lassey, K., Oram, D., O'Shaughnessy, K., Ramankutty, N., Reid, G., Rind, D., Rosenlof, K.,
 Sausen, R., Schwarzkopf, D., Solanki, S. K., Stenchikov, G., Stuber, N., Takemura, T., Textor, C., Wang, R., Weiss, R.,
 Whorf, T., Nakajima, T., Ramanathan, V., Ramaswamy, V., Artaxo, P., Berntsen, T., Betts, R., Fahey, D. W., Haywood,
 J., Lean, J., Lowe, D. C., Myhre, G., Nganga, J., Prinn, R., Raga, G., Schulz, M., and Dorland, R. V.: Changes in
 Atmospheric Constituents and in Radiative Forcing, 2007.
- Ghan, S. J., Liu, X., Easter, R. C., Zaveri, R., Rasch, P. J., Yoon, J.-H., and Eaton, B.: Toward a Minimal Representation
 of Aerosols in Climate Models: Comparative Decomposition of Aerosol Direct, Semidirect, and Indirect Radiative
 Forcing, Journal of Climate, 25, 6461–6476, https://doi.org/10.1175/JCLI-D-11-00650.1, 2012.
- Glotfelty, T., Alapaty, K., He, J., Hawbecker, P., Song, X., and Zhang, G.: The Weather Research and Forecasting Model
 with Aerosol–Cloud Interactions (WRF-ACI): Development, Evaluation, and Initial Application, Mon Weather Rev, 147,
 1491–1511, https://doi.org/10.1175/MWR-D-18-0267.1, 2019.
- Goddard, P. B., Kravitz, B., MacMartin, D. G., and Wang, H.: The Shortwave Radiative Flux Response to an Injection
 of Sea Salt Aerosols in the Gulf of Mexico, Journal of Geophysical Research: Atmospheres, 127, e2022JD037067,
 https://doi.org/10.1029/2022JD037067, 2022.
- Gong, S. L.: A parameterization of sea-salt aerosol source function for sub- and super-micron particles, Global
 Biogeochemical Cycles, 17, https://doi.org/10.1029/2003GB002079, 2003.
- Grythe, H., Ström, J., Krejci, R., Quinn, P., and Stohl, A.: A review of sea-spray aerosol source functions using a large
 global set of sea salt aerosol concentration measurements, Atmospheric Chemistry and Physics, 14, 1277–1297,
 https://doi.org/10.5194/acp-14-1277-2014, 2014.
- Haywood, J. M., Jones, A., Jones, A. C., and Rasch, P. J.: Climate Intervention using marine cloud brightening (MCB)
 compared with stratospheric aerosol injection (SAI) in the UKESM1 climate model, EGUsphere, 1–38,
 https://doi.org/10.5194/egusphere-2023-1611, 2023.
- Hill, S. and Ming, Y.: Nonlinear climate response to regional brightening of tropical marine stratocumulus, Geophysical
 Research Letters, 39, https://doi.org/10.1029/2012GL052064, 2012.

Hoffmann, F. and Feingold, G.: Cloud Microphysical Implications for Marine Cloud Brightening: The Importance of the
Seeded Particle Size Distribution, Journal of the Atmospheric Sciences, 78, 3247–3262, https://doi.org/10.1175/JAS-D21-0077.1, 2021.

695 Horowitz, H. M., Holmes, C., Wright, A., Sherwen, T., Wang, X., Evans, M., Huang, J., Jaeglé, L., Chen, Q., Zhai, S., and Alexander, B.: Effects of Sea Salt Aerosol Emissions for Marine Cloud Brightening on Atmospheric Chemistry: 696 Forcing, 697 Geophysical Research Letters, Implications for Radiative 47, e2019GL085838, https://doi.org/10.1029/2019GL085838, 2020. 698

Janssens-Maenhout, G., Crippa, M., Guizzardi, D., Dentener, F., Muntean, M., Pouliot, G., Keating, T., Zhang, Q.,
Kurokawa, J., Wankmüller, R., Denier van der Gon, H., Kuenen, J. J. P., Klimont, Z., Frost, G., Darras, S., Koffi, B., and
Li, M.: HTAP_v2.2: a mosaic of regional and global emission grid maps for 2008 and 2010 to study hemispheric transport
of air pollution, Atmospheric Chemistry and Physics, 15, 11411–11432, https://doi.org/10.5194/acp-15-11411-2015,
2015.

- Jones, A. and Haywood, J. M.: Sea-spray geoengineering in the HadGEM2-ES earth-system model: radiative impact and
 climate response, Atmospheric Chemistry and Physics, 12, 10887–10898, https://doi.org/10.5194/acp-12-10887-2012,
 2012.
- Jones, A., Haywood, J., and Boucher, O.: Climate impacts of geoengineering marine stratocumulus clouds, Journal of
 Geophysical Research: Atmospheres, 114, https://doi.org/10.1029/2008JD011450, 2009.

Kelly, J. T., Bhave, P. V., Nolte, C. G., Shankar, U., and Foley, K. M.: Simulating emission and chemical evolution of
coarse sea-salt particles in the Community Multiscale Air Quality (CMAQ) model, Geoscientific Model Development,
3, 257–273, https://doi.org/10.5194/gmd-3-257-2010, 2010.

- Korhonen, H., Carslaw, K. S., and Romakkaniemi, S.: Enhancement of marine cloud albedo via controlled sea spray
 injections: a global model study of the influence of emission rates, microphysics and transport, Atmospheric Chemistry
 and Physics, 10, 4133–4143, https://doi.org/10.5194/acp-10-4133-2010, 2010.
- Kravitz, B., Forster, P. M., Jones, A., Robock, A., Alterskjær, K., Boucher, O., Jenkins, A. K. L., Korhonen, H., 715 716 Kristjánsson, J. E., Muri, H., Niemeier, U., Partanen, A.-I., Rasch, P. J., Wang, H., and Watanabe, S.: Sea spray 717 geoengineering experiments in the geoengineering model intercomparison project (GeoMIP): Experimental design and 718 Research: preliminary results. Journal of Geophysical Atmospheres, 118, 11,175-11,186, 719 https://doi.org/10.1002/jgrd.50856, 2013.
- Kravitz, B., Wang, H., Rasch, P. J., Morrison, H., and Solomon, A. B.: Process-model simulations of cloud albedo
 enhancement by aerosols in the Arctic, Phil. Trans. R. Soc. A., 372, 20140052, https://doi.org/10.1098/rsta.2014.0052,
 2014.
- Latham, J., Rasch, P., Chen, C.-C., Kettles, L., Gadian, A., Gettelman, A., Morrison, H., Bower, K., and Choularton, T.:
 Global temperature stabilization via controlled albedo enhancement of low-level maritime clouds, Philosophical
 Transactions of the Royal Society A: Mathematical, Physical and Engineering Sciences, 366, 3969–3987,
 https://doi.org/10.1098/rsta.2008.0137, 2008.
- 727 Latham, J., Bower, K., Choularton, T., Coe, H., Connolly, P., Cooper, G., Craft, T., Foster, J., Gadian, A., Galbraith, L., 728 Iacovides, H., Johnston, D., Launder, B., Leslie, B., Meyer, J., Neukermans, A., Ormond, B., Parkes, B., Rasch, P., Rush, 729 J., Salter, S., Stevenson, T., Wang, H., Wang, Q., and Wood, R.: Marine cloud brightening, Philosophical Transactions 730 of the Roval Society A: Mathematical, Physical and Engineering Sciences, 370, 4217-4262,

- 731 https://doi.org/10.1098/rsta.2012.0086, 2012.
- Latham, J., Gadian, A., Fournier, J., Parkes, B., Wadhams, P., and Chen, J.: Marine cloud brightening: regional applications, Philosophical Transactions of the Royal Society A: Mathematical, Physical and Engineering Sciences, 372, 20140053, https://doi.org/10.1098/rsta.2014.0053, 2014.
- Lenton, T. M. and Vaughan, N. E.: The radiative forcing potential of different climate geoengineering options,
 Atmospheric Chemistry and Physics, 9, 5539–5561, https://doi.org/10.5194/acp-9-5539-2009, 2009.
- Liu, F., Mao, F., Rosenfeld, D., Pan, Z., Zang, L., Zhu, Y., Yin, J., and Gong, W.: Opposing comparable large effects of
 fine aerosols and coarse sea spray on marine warm clouds, Commun Earth Environ, 3, 1–9,
 https://doi.org/10.1038/s43247-022-00562-y, 2022.
- Liu, Z., Wang, M., Rosenfeld, D., Zhu, Y., Bai, H., Cao, Y., and Liang, Y.: Evaluation of Cloud and Precipitation
 Response to Aerosols in WRF-Chem With Satellite Observations, Journal of Geophysical Research: Atmospheres, 125,
 e2020JD033108, https://doi.org/10.1029/2020JD033108, 2020.
- Mahfouz, N. G. A., Hill, S. A., Guo, H., and Ming, Y.: The Radiative and Cloud Responses to Sea Salt Aerosol
 Engineering in GFDL Models, Geophysical Research Letters, 50, e2022GL102340,
 https://doi.org/10.1029/2022GL102340, 2023.
- Mengel, M., Nauels, A., Rogelj, J., and Schleussner, C.-F.: Committed sea-level rise under the Paris Agreement and the
 legacy of delayed mitigation action, Nat Commun, 9, 601, https://doi.org/10.1038/s41467-018-02985-8, 2018.
- Monahan, E. C., Spiel, D. E., and Davidson, K. L.: A Model of Marine Aerosol Generation Via Whitecaps and Wave
 Disruption, in: Oceanic Whitecaps: And Their Role in Air-Sea Exchange Processes, edited by: Monahan, E. C. and
 Niocaill, G. M., Springer Netherlands, Dordrecht, 167–174, https://doi.org/10.1007/978-94-009-4668-2 16, 1986.
- Morrison, H., Thompson, G., and Tatarskii, V.: Impact of Cloud Microphysics on the Development of Trailing Stratiform
 Precipitation in a Simulated Squall Line: Comparison of One- and Two-Moment Schemes, Monthly Weather Review,
 137, 991–1007, https://doi.org/10.1175/2008mwr2556.1, 2009.
- Niemeier, U., Schmidt, H., Alterskjær, K., and Kristjánsson, J. E.: Solar irradiance reduction via climate engineering:
 Impact of different techniques on the energy balance and the hydrological cycle, Journal of Geophysical Research:
 Atmospheres, 118, 11,905-11,917, https://doi.org/10.1002/2013JD020445, 2013.
- Partanen, A.-I., Kokkola, H., Romakkaniemi, S., Kerminen, V.-M., Lehtinen, K. E. J., Bergman, T., Arola, A., and
 Korhonen, H.: Direct and indirect effects of sea spray geoengineering and the role of injected particle size, Journal of
 Geophysical Research: Atmospheres, 117, https://doi.org/10.1029/2011JD016428, 2012.
- Paulot, F., Paynter, D., Winton, M., Ginoux, P., Zhao, M., and Horowitz, L. W.: Revisiting the Impact of Sea Salt on
 Climate Sensitivity, Geophysical Research Letters, 47, e2019GL085601, https://doi.org/10.1029/2019GL085601, 2020.
- 762 Pleim, J. E.: A Combined Local and Nonlocal Closure Model for the Atmospheric Boundary Layer. Part I: Model 763 Description and Testing. Journal of Applied Meteorology and Climatology, 46. 1383-1395. 764 https://doi.org/10.1175/jam2539.1, 2007.
- Pringle, K. J., Carslaw, K. S., Fan, T., Mann, G. W., Hill, A., Stier, P., Zhang, K., and Tost, H.: A multi-model assessment
 of the impact of sea spray geoengineering on cloud droplet number, Atmospheric Chemistry and Physics, 12, 11647–
 11663, https://doi.org/10.5194/acp-12-11647-2012, 2012.

- Quaas, J., Boucher, O., Bellouin, N., and Kinne, S.: Satellite-based estimate of the direct and indirect aerosol climate
 forcing, Journal of Geophysical Research: Atmospheres, 113, https://doi.org/10.1029/2007JD008962, 2008.
- Rasch, P., Hirasawa, H., Wu, M., Doherty, S., Wood, R., Wang, H., Jones, A., Haywood, J., and Singh, H.: A protocol for
 model intercomparison of impacts of Marine Cloud Brightening Climate Intervention, EGUsphere, 1–43,
 https://doi.org/10.5194/egusphere-2024-1031, 2024.
- Rasch, P. J., Latham, J., and Chen, C.-C. (Jack): Geoengineering by cloud seeding: influence on sea ice and climate
 system, Environ. Res. Lett., 4, 045112, https://doi.org/10.1088/1748-9326/4/4/045112, 2009.
- Rosenfeld, Daniel, Sherwood, Steven, Wood, Robert, Donner, and Leo: Climate Effects of Aerosol-Cloud Interactions.,
 Science, https://doi.org/10.1126/science.1247490, 2014.
- Rosenfeld, D., Zhu, Y., Wang, M., Zheng, Y., Goren, T., and Yu, S.: Aerosol-driven droplet concentrations dominate
 coverage and water of oceanic low-level clouds, Science, 363, eaav0566, https://doi.org/10.1126/science.aav0566, 2019.
- 779 Salter, S., Sortino, G., and Latham, J.: Sea-going hardware for the cloud albedo method of reversing global warming,
- 780 Philosophical Transactions of the Royal Society A: Mathematical, Physical and Engineering Sciences, 366, 3989–4006,
- 781 https://doi.org/10.1098/rsta.2008.0136, 2008.
- Stjern, C. W., Muri, H., Ahlm, L., Boucher, O., Cole, J. N. S., Ji, D., Jones, A., Haywood, J., Kravitz, B., Lenton, A.,
 Moore, J. C., Niemeier, U., Phipps, S. J., Schmidt, H., Watanabe, S., and Kristjánsson, J. E.: Response to marine cloud
 brightening in a multi-model ensemble, Atmospheric Chemistry and Physics, 18, 621–634, https://doi.org/10.5194/acp18-621-2018, 2018.
- Stuart, G. S., Stevens, R. G., Partanen, A.-I., Jenkins, A. K. L., Korhonen, H., Forster, P. M., Spracklen, D. V., and Pierce,
 J. R.: Reduced efficacy of marine cloud brightening geoengineering due to in-plume aerosol coagulation:
 parameterization and global implications, Atmospheric Chemistry and Physics, 13, 10385–10396,
 https://doi.org/10.5194/acp-13-10385-2013, 2013.
- Twomey, S.: Pollution and the planetary albedo, Atmospheric Environment (1967), 8, 1251–1256,
 https://doi.org/10.1016/0004-6981(74)90004-3, 1974.
- Visioni, D., Kravitz, B., Robock, A., Tilmes, S., Haywood, J., Boucher, O., Lawrence, M., Irvine, P., Niemeier, U., Xia,
 L., Chiodo, G., Lennard, C., Watanabe, S., Moore, J. C., and Muri, H.: Opinion: The scientific and community-building
 roles of the Geoengineering Model Intercomparison Project (GeoMIP) past, present, and future, Atmospheric
 Chemistry and Physics, 23, 5149–5176, https://doi.org/10.5194/acp-23-5149-2023, 2023.
- Wang, K., Zhang, Y., Yu, S., Wong, D. C., Pleim, J., Mathur, R., Kelly, J. T., and Bell, M.: A comparative study of twoway and offline coupled WRF v3.4 and CMAQ v5.0.2 over the contiguous US: performance evaluation and impacts of
 chemistry–meteorology feedbacks on air quality, Geoscientific Model Development, 14, 7189–7221,
 https://doi.org/10.5194/gmd-14-7189-2021, 2021.
- Wong, D. C., Pleim, J., Mathur, R., Binkowski, F., Otte, T., Gilliam, R., Pouliot, G., Xiu, A., Young, J. O., and Kang, D.:
 WRF-CMAQ two-way coupled system with aerosol feedback: software development and preliminary results,
 Geoscientific Model Development, 5, 299–312, https://doi.org/10.5194/gmd-5-299-2012, 2012.
- Wood, R.: Assessing the potential efficacy of marine cloud brightening for cooling Earth using a simple heuristic model,
 Atmospheric Chemistry and Physics, 21, 14507–14533, https://doi.org/10.5194/acp-21-14507-2021, 2021.

Yu, S., Mathur, R., Pleim, J., Wong, D., Gilliam, R., Alapaty, K., Zhao, C., and Liu, X.: Aerosol indirect effect on the
grid-scale clouds in the two-way coupled WRF–CMAQ: model description, development, evaluation and regional
analysis, Atmospheric Chemistry and Physics, 14, 11247–11285, https://doi.org/10.5194/acp-14-11247-2014, 2014.

Zhang, K. M., Knipping, E. M., Wexler, A. S., Bhave, P. V., and Tonnesen, G. S.: Size distribution of sea-salt emissions
as a function of relative humidity, Atmospheric Environment, 39, 3373–3379,
https://doi.org/10.1016/j.atmosenv.2005.02.032, 2005.

Zhao, D., Lin, Y., Dong, W., Qin, Y., Chu, W., Yang, K., Letu, H., and Huang, L.: Alleviated WRF Summer Wet Bias
Over the Tibetan Plateau Using a New Cloud Macrophysics Scheme, Journal of Advances in Modeling Earth Systems,
15, e2023MS003616, https://doi.org/10.1029/2023MS003616, 2023.

Zhao, M., Cao, L., Duan, L., Bala, G., and Caldeira, K.: Climate More Responsive to Marine Cloud Brightening Than
Ocean Albedo Modification: A Model Study, Journal of Geophysical Research: Atmospheres, 126, e2020JD033256,
https://doi.org/10.1029/2020JD033256, 2021.

Areas	Cloud Fraction		CDNC (# cm ⁻³)		LWP (g m ⁻²)		Regional sea-salt aerosols (µg m ⁻³)	
	Base	Exp	Base	Exp	Base	Exp	Base	Exp
WP	0.0445	0.0488	19.3	100.0	12.8	19.8	8.91	143
NP	0.0678	0.0760	9.7	60.2	24.6	43.9	7.18	126
Equa	0.0051	0.0059	17.5	83.4	0.85	1.4	7.32	102
SP	0.0547	0.0617	11.5	89.4	21.6	38.9	6.79	176
SA	0.0519	0.0575	12.3	92.2	23.5	41.6	7.00	149

Table 1. The cloud fraction, CDNC, LWP, and regional sea-salt aerosol concentrations at Base and after injection of sea-salt aerosols at 10^{-9} kg m⁻² s⁻¹ (Exp) for five ocean regions.

Strategies	Areas	SW_TOT (W m ⁻²)	SW_CLD (W m ⁻²)	SW_AER (W m ⁻²)	SW_AER_CLR (W m ⁻²)
	WP	0.5	0.4	0.11	0.16
	NP	2.1	2.0	0.11	0.19
Natural×5	Equa	0.07	0.01	0.06	0.07
	SP	1.7	1.6	0.08	0.14
	SA	1.4	1.3	0.11	0.16
	WP	3.8	1.9	1.9	2.3
	NP	8.4	6.8	1.6	2.4
Wind-adjusted	Equa	1.4	0.3	1.2	1.2
	SP	7.6	5.8	1.8	2.6
	SA	8.0	5.9	2.1	2.8
	WP	18.0	4.6	13.0	15.0
	NP	23.0	13.0	9.8	15.0
10 ⁻⁹ kg m ⁻² s ⁻¹	Equa	11.0	0.6	10.0	11.0
	SP	25.0	11.0	14.0	19.0
	SA	22.0	11.0	11.0	15.0
	WP	6.9	2.9	4.0	5.1
F' 1 ' 1	NP	16.0	11.0	5.1	7.8
Fixed-wind-	Equa	5.0	0.5	4.5	4.7
adjusted	SP	17.0	9.9	6.6	9.8
	SA	20.0	11.0	9.1	13.0

Table 2. Differences (Exp - Base) in SW_TOT, SW_CLD, SW_AER and SW_AER_CLR at the TOA due to the injection of sea-salt aerosols in different strategies in five ocean regions.

Note: SW_TOT is upward shortwave radiative flux at the TOA for all-sky conditions. The response of SW_TOT to the sea-salt aerosols injection can be separated into the influence of the cloud radiative effect (SW_CLD, where the influence of the aerosol is excluded) and the influence of the aerosol direct scattering effect (SW_AER) in the presence of clouds. That is, $SW_TOT = SW_CLD + SW_AER$. The SW_AER_CLR is the response of aerosol direct scattering to the upward shortwave radiative flux at the TOA under clear skies.

Table 3. Relative effects of cloud fraction and albedo changes on CRF'_{param} and Twomey, LWP, and cloud fraction effects to SW_CLD responses after uniform fixed injection of 10^{-9} kg m⁻² s⁻¹ sea-salt aerosols over five ocean regions.

		CRF'param		$\frac{\Delta \alpha}{\Delta \ln AOD}$			
Areas	$\alpha_c' \bar{f}$	$\bar{\alpha}_c f'$	$\alpha'_c f'$	Twomey Effect	LWP Effect	Cloud Fraction Effect	
WP	71.5%	20.7%	7.82%	48.4%	41.6%	10.1%	
NP	72.7%	16.9%	10.4%	48.5%	41.7%	9.71%	
Equa	60.2%	27.3%	12.4%	36.4%	58.5%	5.1%	
SP	73.8%	15.9%	10.3%	51.8%	39.0%	9.2%	
SA	77.3%	13.9%	8.81%	52.5%	39.7%	7.8%	

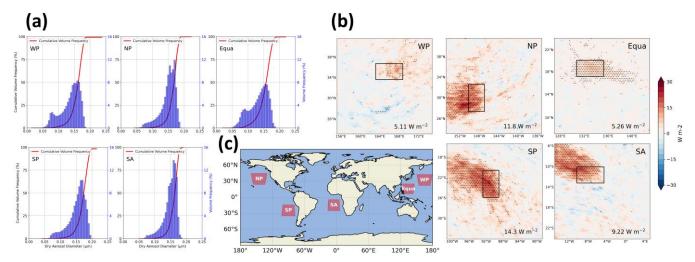


Figure 1. Injecting sea-salt aerosols into five open sea regions to simulate the implementation of MCB geoengineering. (a) The cumulative volume frequency of increased aerosol dry particle size (uniform injection of 10^{-9} kg m⁻² s⁻¹ sea-salt aerosols over the entire region). (b) Differences (Exp - Base) in the spatial distribution of the TOA upward shortwave radiative flux response (SW_TOT) resulting from uniform injection of 10^{-9} kg m⁻² s⁻¹ sea-salt aerosol in sensitive areas in five ocean regions, with SW_TOT response values resulting only in sensitive areas labeled in the lower right corner. Areas labeled with dots indicate mean differences that are significant at the 95% confidence level. Black rectangles are sensitive areas. (c) Location of the five ocean modeling domains.

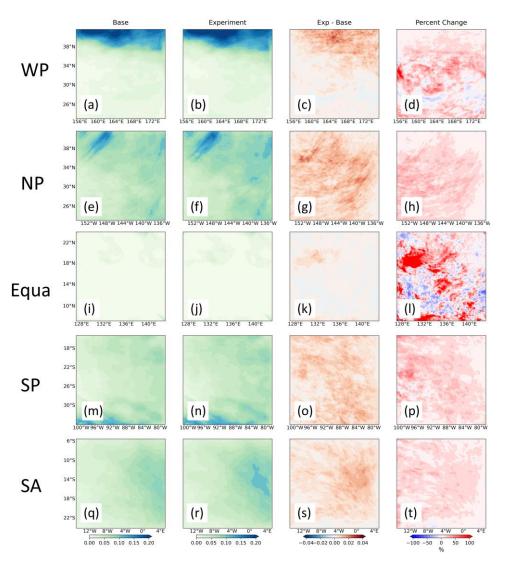


Figure 2. Column mean liquid cloud fraction from the surface to 3000 m altitude for five regions. The first to fourth columns are Base, the sensitivity experiment with a uniform injection of 10^{-9} kg m⁻² s⁻¹ seasalt aerosols over the entire region, Exp - Base, and the percent change of Exp - Base, respectively.

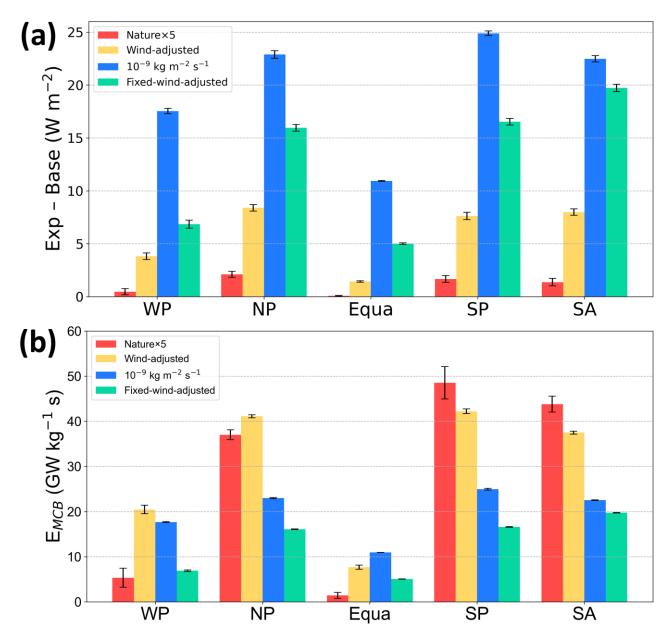


Figure 3. (a) The differences in SW_TOT and (b) the MCB efficiency (E_{MCB}) due to the injection of seasalt aerosols in different strategies in five ocean regions.

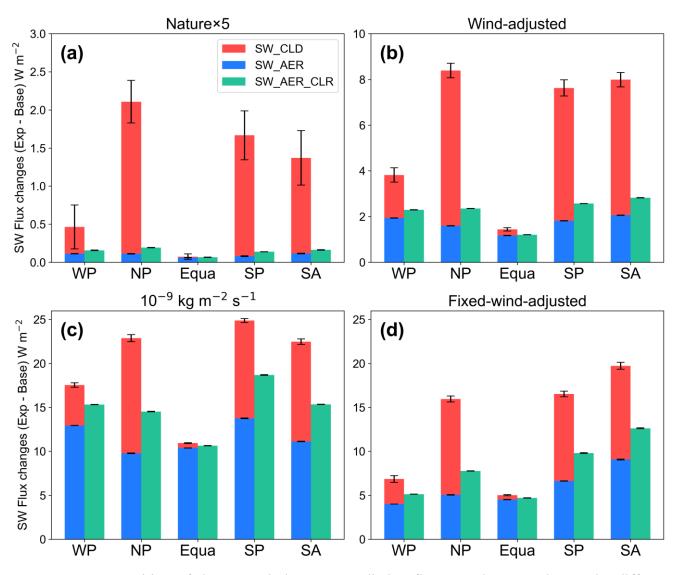


Figure 4. Decomposition of the upward shortwave radiative fluxes at the TOA due to the different strategies of injecting sea-salt aerosols in the five regions. Note that the y-axis ranges are not consistent.

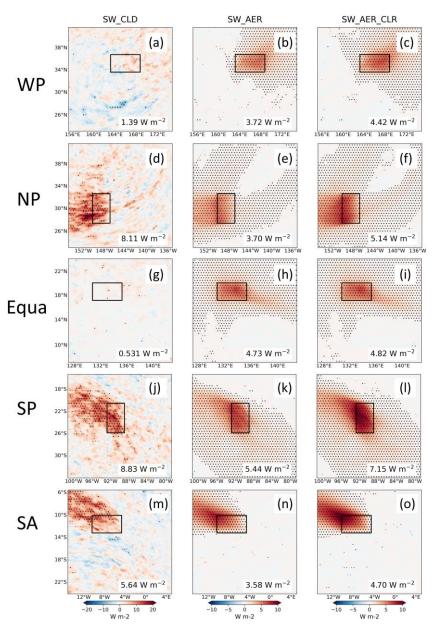


Figure 5. Spatial distribution of SW_CLD (first column), SW_AER (second column), and SW_AER_CLR (third column) responses resulting from the injection of 10⁻⁹ kg m⁻² s⁻¹ sea-salt aerosols in the sensitive areas over five ocean regions. The values of the radiative flux responses generated only in the sensitive area are labeled in the lower right corner. Areas labeled with dots indicate mean differences that are significant at the 95% confidence level. The black rectangles are sensitive areas.

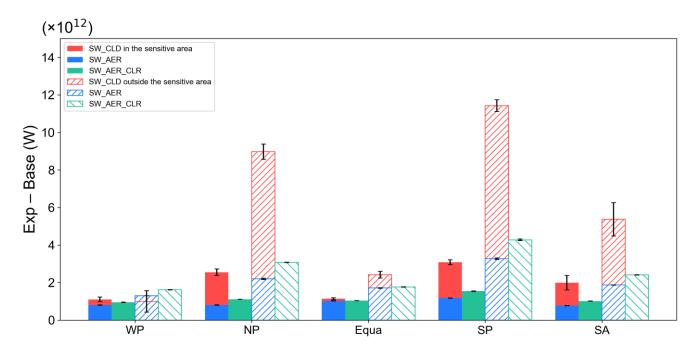


Figure 6. Total SW_CLD, SW_AER, and SW_AER_CLR responses resulting from the injection of 10⁻⁹ kg m⁻² s⁻¹ sea-salt aerosols within the sensitive areas of the five regions. The solid columns indicate the total radiative response calculated for aerosol injection within the sensitive areas. Columns filled with hatching indicate the total radiative response outside the sensitive areas.

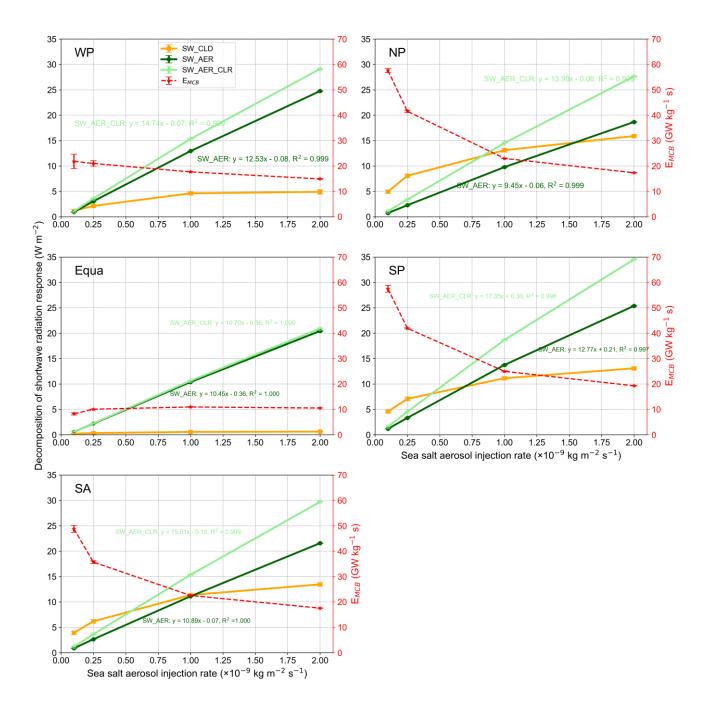


Figure 7. Changes in SW_CLD, SW_AER, and SW_AER_CLR radiative responses due to sea-salt aerosols uniformly injected in varying amounts in five ocean regions, and corresponding changes in E_{MCB} . SW_AER and SW_AER_CLR are labeled with the results of the corresponding linear regression analysis.

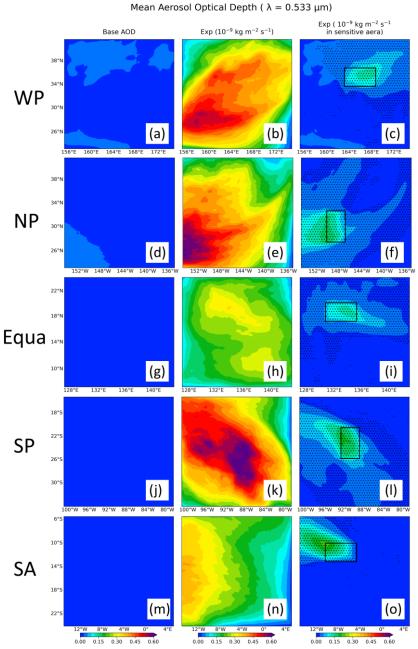


Figure 8. Spatial distribution of mean AOD ($\lambda = 0.533 \ \mu m$) for five ocean regions. The first column is the AOD for Base, the second column is the AOD after uniform injection at 10⁻⁹ kg m⁻² s⁻¹, and the third column is the AOD after uniform injection in sensitive areas. Areas labeled with dots indicate mean differences that are significant at the 95% confidence level. The black rectangles are sensitive areas.

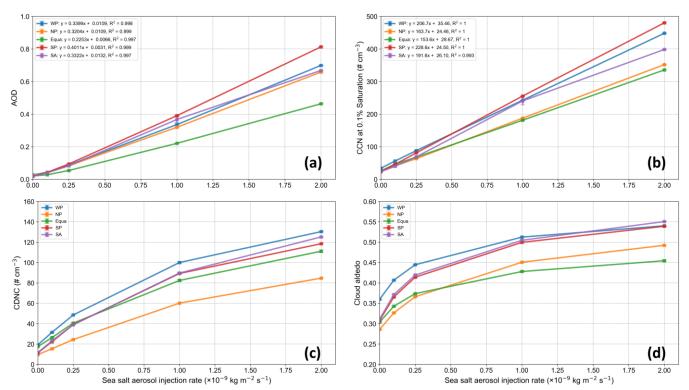


Figure 9. Relationship between changes in regional mean (a) AOD, (b) CCN, (c) CDNC, and (d) cloud albedo due to uniform injection of sea-salt aerosols across the region and the amounts of sea-salt aerosols injected. The results of the linear regression of (a) AOD and (b) CCN on the sea-salt aerosols injection amount are given at the legends.

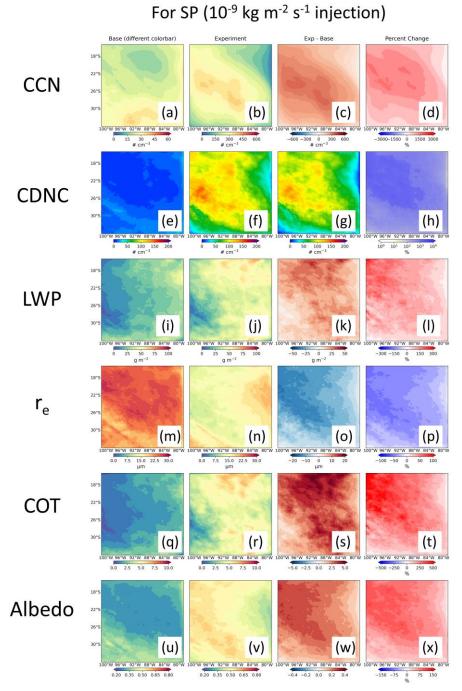


Figure 10. Spatial distribution of liquid cloud property responses after uniform injection of sea-salt aerosols with 10^{-9} kg m⁻² s⁻¹ in the SP region. Results are shown for cloud condensation nuclei (CCN, S = 0.1%, # cm⁻³), cloud droplet number concentration (# cm⁻³), liquid water path (LWP, g m⁻²), cloud effective radius (r_{e} , µm), cloud optical thickness (COT), and cloud albedo for Base (first column), Exp (second column), Exp - Base (third column), and the percentage change in Exp - Base (fourth column), respectively.

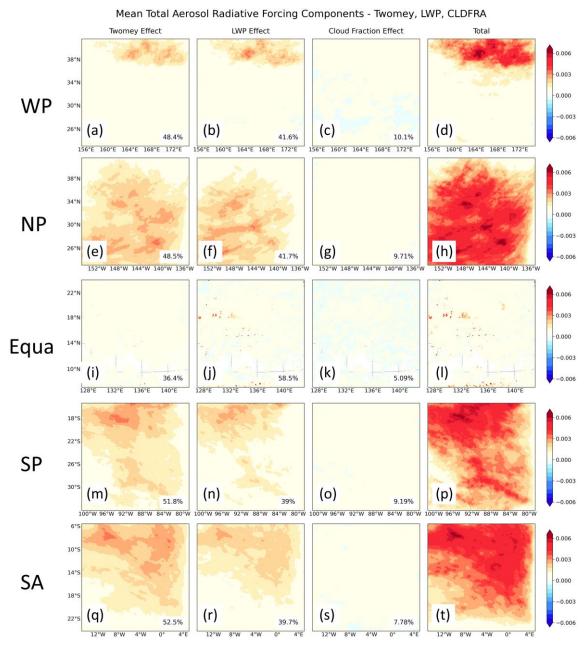


Figure 11. Spatial distribution of cloud property changes in response to SW_CLD radiation after uniform injection of sea-salt aerosols in five regions. The first column is the Twomey effect, the second column is the LWP effect, the third column is the cloud fraction effect, and the fourth column is the cloud susceptibility $(\frac{\Delta \alpha}{\Delta \ln AOD})$ to aerosol injection for the sum of the three effects. The percentage contribution of each to the total SW_CLD response over the entire region is labeled in the lower right corner.