1 The effectiveness of solar radiation management using fine sea spray across multiple

2 climatic regions

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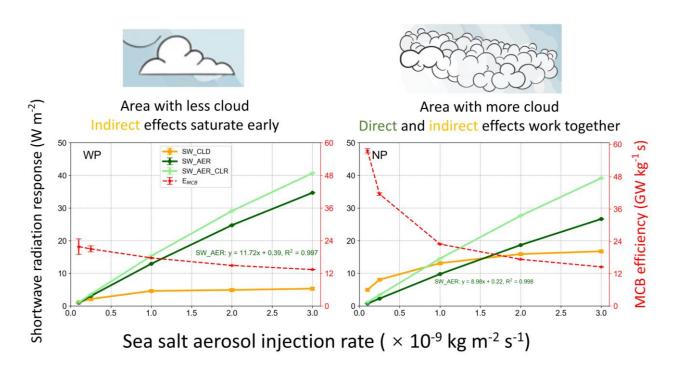
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23 Graphic Abstract



26 Abstract

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

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

44 **1. Introduction**

As global temperatures continue to rise, the international community is facing an unprecedented challenge 45 to achieve the ambitious goal set in the Paris Agreement of limiting global warming to within 1.5 °C 46 (Mengel et al., 2018). One of the key outcomes of the recently concluded 28th Conference of the Parties 47 (COP28) was the completion of the first Global Stocktake (GST), a mid-term assessment of the progress 48 made by countries toward achieving the climate goals of the Paris Agreement. However, the report 49 highlighted that current efforts to reduce emissions had fallen short of the intended targets 50 (https://www.cop28.com/). Against this backdrop, scientists are turning their attention to geoengineering 51 methods to reduce or offset the impacts of climate change through artificial interventions in the climate 52 (Visioni et al., 2023). Some geoengineering methods seek to capture or remove CO₂ from the atmosphere 53 to increase carbon sinks, while others focus on modifying solar radiation, reducing incoming solar 54 shortwave radiation, or reflecting more sunlight to cool the earth, known as solar radiation management 55 (SRM) (Lenton and Vaughan, 2009). Among these, marine cloud brightening (MCB) has a realistic basis 56 57 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 can lead to brighter clouds, with clear ship 58 tracks also visible from satellites, and MCB aims to replicate this effect by spraying sea-salt aerosols 59 (Chen et al., 2012). 60

Aerosol-cloud interactions and their impacts on climate are complex (Rosenfeld et al., 2014, 2019). 61 Injected sea-salt aerosols affect clouds through indirect effects (Paulot et al., 2020). In the case of a 62 constant liquid water content, an increase in cloud droplet number concentration (CDNC) decreases the 63 cloud droplet size, increases the total surface area of cloud droplets, thereby enhancing the cloud albedo, 64 65 forming brighter clouds, and reflecting more sunlight back to space (the first indirect effect or Twomey effect) (Twomey, 1974). At the same time, the decrease in cloud droplet size suppresses precipitation, 66 thereby increasing the cloud's lifespan and optical thickness (the second indirect effect of aerosols) 67 (Albrecht, 1989). In addition, those aerosols that are not injected into the clouds scatter more sunlight 68 back into space through the direct scattering effect (Ahlm et al., 2017; Partanen et al., 2012; Zhao et al., 69 70 2021). Therefore, this method is also called marine sky brightening (MSB), which can work even when there are no clouds. Here, we collectively refer to the practice of injecting sea-salt aerosols as MCB. 71

Compared to other geoengineering schemes, such as stratospheric aerosol injection (SAI), MCB has 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 (Carlisle et al., 2020; Feingold et al.,
2024). Therefore, most of the current literature examine the environmental and climate impacts of MCB
implementation through modeling.

80 Table S1 summarizes the results of current modeling simulations on MCB with sea-salt aerosols, as well as their implementation strategies. Most MCB studies use Earth-System Models to assess the impacts 81 of the implementation of MCB on climate. Early MCB studies assumed the effects of MCB 82 implementation by setting a fixed CDNC or directly modifying the cloud effective radius (r_e), ignoring 83 the processes such as generation, transport, dry and wet deposition, and activation of injected sea-salt 84 aerosols, and not including the direct radiative effect of aerosols. With the development of models, 85 researchers started to conduct more detailed studies by injecting aerosols or increasing sea-salt aerosol 86 emissions, taking into account the post-injection processes of aerosols mentioned above. 87

The implementation region of MCB is crucial. Existing studies have focused on the impacts of MCB 88 implementation in three key areas: open oceans globally, the equatorial region (between 30°S and 30°N), 89 and coastal areas with widespread marine stratocumulus clouds. Alterskjær et al. (2012) used the cloud-90 weighted susceptibility function to find the most sensitive regions to the injection of sea-salt aerosols. 91 Similarly, Jones and Haywood (2012) determined the 10% of the marine regions globally most suitable 92 for implementing MCB through an iterative method. The contributions of direct and indirect effects of 93 aerosols during the implementation of MCB are still uncertain and quantitative assessment of both is 94 lacking (Haywood et al., 2023; Partanen et al., 2012). 95

96 Here, we use the two-way coupled Weather Research and Forecasting - Community Multi-scale Air 97 Quality model (WRF-CMAQ), combined with previous studies on the region and injection strategies, to 98 implement MCB in five open ocean regions. This study simulates the regional radiation and cloud 99 responses caused by injecting sea-salt aerosols. This aims to explore the commonalities and differences 100 in MCB implementation in different regions and to seek the optimal strategy for MCB injection.

101

102 2. Experiments and methods

103 2.1 Model configuration

The two-way coupled WRF (v3.4) - CMAQ (v5.0.2) model that considers both direct and indirect effects
of aerosols was used in this study (Yu et al., 2014). In the two-way coupled model, aerosols predicted by

106 CMAQ are able to affect clouds, radiation, and precipitation simulated by WRF in a consistent online 107 coupled manner (Wong et al., 2012). Yu et al. (2014) further extended the two-way coupled WRF-CMAQ 108 model by incorporating the aerosol indirect effects (including the first, second, and glaciation aerosol 109 indirect effects), improving the ability of the WRF-CMAQ model to predict clouds and radiation. Wang 110 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 111 asymmetric convective model (ACM2) for a planetary boundary layer (PBL) scheme (Pleim, 2007), the 112 Morrison 2-moment cloud microphysics scheme (Morrison et al., 2009), the Kain-Fritsch (KF2) cumulus 113 cloud parameterization, the Rapid Radiative Transfer Model for General Circulation Models (RRTMG) 114 longwave and shortwave radiation schemes, and the Pleim-Xiu (PX) land-surface scheme. The 115 meteorological initial and boundary conditions were provided by the National Center for Environmental 116 Prediction (NCEP) final analysis dataset (FNL) with a spatial resolution of 1°×1° and temporal resolution 117 of 6 h. The carbon bond gas-phase chemical mechanism (CB05) and aerosol module of AERO6 were 118 used in the CMAQ model. The anthropogenic emissions were taken from the Hemispheric Transport of 119 Air Pollution (HTAP V2) projects (Janssens-Maenhout et al., 2015). The biogenic emissions were 120 121 estimated by the Biogenic Emissions Inventory System version 3.14 (BEISv3.14) model (Carlton and Baker, 2011). 122

Sea salt emissions were calculated online in CMAQ and were divided into open-ocean and surf-zone 123 emissions. In the open ocean, Gong (2003) extended the sea-salt aerosol parameterization of Monahan et 124 al. (1986) to submicron sizes, with the emission flux being linearly proportional to the ocean area covered 125 by whitecaps. CMAQ represents the atmospheric particle distribution as the superposition of three log-126 127 normal modes, the Aitken, Accumulation, and Coarse modes (Binkowski and Roselle, 2003). The particle 128 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). The geometric 129 mean diameter of accumulation mode sea-salt aerosols in the CMAQ ranged from 0.2651 to 0.8187 µm, 130 with the geometric standard deviation constrained between 1.76 and 1.83. Surf-zone emissions were 131 calculated using the open ocean-source function of Gong (2003), with a fixed whitecap coverage of 100% 132 and a surf-zone width of 50 m. Kelly et al. (2010) provided a detailed description of these processes. In 133 the CMAQ model, the number concentration emission rate was calculated from the mass emissions rate 134 as follows: 135

136
$$E_{3n} = \left(\frac{6}{\pi}\right) \left(\frac{E_n}{\rho_n}\right) \tag{1}$$

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

145 **2.2 Experimental setup**

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As summarized in Table S1, the MCB geoengineering implementation areas include globally, the 146 147 equator (30°S-30°N) and 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 148 injections of sea-salt aerosols in the five open ocean regions (Fig. 1c). These regions are WP and NP, 149 located in the western and northern Pacific Ocean; Equa, located in the Philippine Sea along the equator; 150 151 and SP and SA, located in the south Pacific and south Atlantic, respectively. The three regions, NP, SP and SA, are located along the western coast of continents, were considered to have extensive coverage of 152 marine stratocumulus clouds and were the most suitable areas for implementing MCB (Alterskjær et al., 153 2012; Hill and Ming, 2012; Jones et al., 2009; Partanen et al., 2012; Stuart et al., 2013). 154

The grid 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 spinup period to minimize the impacts of initial chemical conditions.

161 The results of the Base simulations with the model settings described above and default sea salt 162 emissions (no aerosol injection) were obtained. As can been seen, there are significant differences in the 163 cloud distributions for the five ocean regions in the Base simulations during the study period, with wider 164 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.

172 **Natural**×5: Increase the emission rates of accumulation mode sea-salt aerosols by a factor of 5 (Hill 173 and Ming, 2012). This is a simple wind-speed-dependent increase. The injection rates in the five regions 174 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.

181
$$F_{\rm m, \, baseline} = \begin{cases} 5 \times 2.8 \times 10^{-12} \times \left(\frac{u}{1 \,{\rm m \, s^{-1}}}\right)^{1.5} \,{\rm kg \, m^{-2} s^{-1}}, & u < 7 \,{\rm m \, s^{-1}} \\ 5 \times 2.8 \times 10^{-12} \times 7^{1.5} \,{\rm kg \, m^{-2} s^{-1}}, & u \ge 7 \,{\rm m \, s^{-1}} \end{cases}$$
(3)

Fixed at 10⁻⁹ kg m⁻² s⁻¹: Unlike the previous two injection methods, the injections of sea-salt aerosols 182 at a fixed rate of 10⁻⁹ kg m⁻² s⁻¹ are not dependent on wind speed and increased uniformly over all ocean 183 grids. Injecting sea-salt aerosols at a fixed rate identified the geographic areas that were most sensitive to 184 increased sea-salt aerosols and produced the largest top-of-atmosphere (TOA) radiative perturbations 185 186 (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 187 aerosol transports and dispersion at the boundary. Therefore, based on the results of a fixed 10⁻⁹ kg m⁻² s⁻ 188 ¹ injection rate, we identified the geographical regions (30×50 grid points, approximately 360 km $\times 600$ 189 190 km, away from the domain boundary) in five ocean areas where the TOA radiative perturbations caused by uniform injection were the largest, and the most sensitive. Table S3 shows the locations of these 191 sensitive regions. The injection amount in the sensitive region at a fixed 10⁻⁹ kg m⁻² s⁻¹ injection rate is 192 193 found to be about 1/20 of those in the full domain.

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

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

198 2.3 Calculations

The calculation method related to radiation, cloud properties, and cloud radiation forcing is based on 199 200 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 201 of effective radiation forcing (ERF) (Forster et al., 2007). The sea surface temperature in the model is 202 preset by NCEP-FNL, so the model's surface temperature and upward longwave radiation would not 203 respond to the increased sea-salt aerosols. The total upward shortwave radiation flux (SW TOT) at the 204 TOA is under the all-sky conditions. The responses of SW TOT to the injections of sea-salt aerosols 205 could be divided into the cloud radiative effects (SW CLD, excluding the direct effect of the aerosols) 206 and direct scattering effects when clouds are present (SW AER). 207

208

$$SW_TOT = SW_CLD + SW_AER$$
 (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). For this flux, only the direct scattering effect of aerosols exist as clouds are ignored, which are 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 by the four different injection strategies, we propose the concept of MCB efficiency (E_{MCB}) to measure the relationships between the amount of seasalt aerosol injections and the resulting radiation flux responses (Table S2).

219
$$E_{MCB} = \frac{\text{SW}_{\text{TOT}} \text{ response due to injection of sea-salt aerosol (W m-2)}}{\text{Sea-salt aerosol injections (kg m-2 s-1)}}$$
(5)

This is a measure of the mass efficiency of MCB implementing in different regions, that is, how much the 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 is expected to produce a 1 GW (10⁹ 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

225 flux to different injection methods and injection amounts.

This study focuses on the changes in liquid clouds and evaluated 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):

 $CRF_{param} = \alpha_c f \tag{6}$

232 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):

235

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

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 mean of the Base, and the prime (') defines the monthly mean differences between the sensitivity experiments and Base. The fourth column of Fig. S17 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:

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

Additional statistics are obtained by generating three ensemble members for each experiment in each region using a stochastic kinetic-energy backscatter scheme to add stochastical perturbations (Berner et al., 2011). A two-tailed t-test was applied to assess whether the difference between the Base simulation
and the experiment was statistically significant at the 95% confidence level. Unless otherwise specified,
all results in this study are shown as overall regional monthly averages of the ensemble.

257 **3. Results**

258 **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 259 conclusions, and these variations may be one of the reasons for differences in the assessments of MCB 260 potentials in the previous studies. In this study, sea-salt aerosols injected in different strategies (with dry 261 diameters of about 0.11–0.15 µm, Fig. 1a) increase the SW TOT at the TOA by 0.07–25 W m⁻² in the 262 263 five ocean regions compared with the Base experiment (Fig. 3a). The Natural×5 and Wind-adjusted strategies, which rely on wind speeds, inject sea-salt aerosols of 0.031-0.085 and $0.18-0.21 \times 10^{-9}$ kg m⁻ 264 2 s⁻¹ into the five regions, respectively, and result in SW TOT variations of 0.07–2.1 and 1.4–8.4 W m⁻², 265 respectively (Fig. 3a and Table 2). Uniform injections of sea-salt aerosols at a fixed rate of 10⁻⁹ kg m⁻² s⁻ 266 ¹ results in SW TOT changes of 11–25 W m⁻² in the five regions. The three stratocumulus regions of NP, 267 SP, and SA have the most significant SW TOT responses, all exceeding 20 W m⁻², while the SW TOT 268 responses in the WP and Equa regions are 18 and 11 W m⁻², respectively. 269

Injecting the same amount of sea-salt aerosols results in substantial variations in SW TOT responses 270 271 across the different regions (Fig. S2). The sea-salt aerosols spraved in the Fixed-wind-adjusted experiments are also dependent on wind speed, but the amount of emission rate integrated in the full 272 domain is consistent with the fixed rate of 10^{-9} kg m⁻² s⁻¹, ruling out the differences caused by the amount 273 of injected sea-salt aerosols. Although both strategies inject the same amounts of sea-salt aerosols, the 274 275 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 276 flux changes caused by wind-speed-dependent injections are smaller than those caused by uniform 277 injections, and showed regional differences. 278

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 showed higher MCB number efficiency with less aerosol number flux injected (Fig. S3c). In the same quality injected, the aerosol number varied greatly (Fig. S3d) and the MCB number efficiency is higher for Fixedwind-adjusted than for uniform injection (Fig. S3c).

The productions of sea-salt aerosols in nature are strongly correlated with wind speed, and most 291 models associated sea-salt aerosol emissions with wind speed (Ahlm et al., 2017; Grythe et al., 2014). 292 Injection strategies depending on wind speed make the distributions of added sea-salt aerosols closer to 293 294 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, 295 injection strategies depending on wind speed concentrate the added sea-salt aerosols in strong-wind areas 296 and surf zones, while the weak-wind regions increase relatively little sea-salt aerosols (Fig. S4). Injecting 297 uniformly at a fixed rate in the model will result in a large increase of sea-salt aerosols in places with 298 299 originally low aerosol concentrations (e.g., weak-wind regions). This strategy may not truly reflect the 300 distribution characteristics in the natural environment. However, the uniform increase injection strategy also has its advantages: it can not only avoid the situation of a smaller increase in sea-salt emissions in 301 302 regions with lower wind speeds, but can also identify the geographical areas most sensitive to the increased sea-salt aerosols and producing the largest TOA radiation perturbations (Alterskjær et al., 2012). 303 Therefore, when using models to simulate the injections of sea-salt aerosols by increasing the emission 304 305 rate, it is necessary to fully consider the impact of different injection strategies on the distribution of sea 306 salt emissions and to choose a suitable strategy with the purpose of the study.

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 amount is about 1/20 of the full domain injection) results in changes of 0.49-3.4 W m⁻² in 308 SW TOT in the five ocean regions (Table S2). The SW TOT responses are the largest in the SP region, 309 at 3.4 W m⁻², and 2.7 and 1.7 W m⁻² in the NP and SA regions, respectively, while they were only 0.49 310 and 0.83 W m⁻² in the WP and Equa regions, respectively. The injected sea-salt aerosols produced 311 SW TOT changes of 5.11–14.3 W m⁻² in the sensitive areas (Fig. 1b). Similarly, the increases in SW TOT 312 in the SP, SA, and NP regions all exceeded 9 W m⁻², with the highest in the SP region at 14.3 W m⁻². In 313 the WP and Equa regions, the increases in SW TOT are 5.11 and 5.26 W m⁻², respectively. Considering 314

that the original intents of MCB or MSB design are regional application (hurricane mitigation, coral reef protection and polar sea ice recovery) (Latham et al., 2014), choosing to inject sea-salt aerosols in the 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×5 324 325 and Wind-adjusted strategies is caused by the SW CLD response (Fig. 4a). In the NP, SP, and SA regions, the contributions of SW CLD exceed 70%, suggesting that sea-salt aerosols injected at these locations 326 327 increase the SW TOT mainly by affecting clouds through indirect effects. In the Equa, the responses of SW TOT are entirely caused by SW AER. This is due to the low cloud cover in Equa (Fig. 2i), so the 328 SW CLD caused by aerosol injection is small here. The proportion of SW AER produced by the uniform 329 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, 330 Equa, and SP regions, the proportion of SW AER exceeded that of SW CLD. In the SA region, SW CLD 331 and SW AER are almost equal, while in the NP region, the SW CLD response is 13 W m⁻², still greater 332 than SW AER (9.8 W m⁻²). This is because there is a saturation phenomenon in the cloud response to 333 aerosols injections (discussed below), and the NP, SP, and SA regions provide more SW CLD response, 334 while the cloud responses in the WP and Equa regions saturate and no longer increase. The results of 335 Fixed-wind-adjusted case show that, at the same injection amount, the SW AER responses caused by the 336 337 injection strategy relying on wind speed is significantly smaller than those of the method with fixed-rate uniform injection, while the disparity in SW CLD responses is minimal. This is mainly because the fixed-338 rate uniform injection leads to a larger aerosol number flux (Fig. S3d). In addition, the injection strategy 339 relying on wind speed distributed most of the increased sea-salt aerosols to areas with already high 340 341 emissions, such as strong-wind areas and surf zones, where the excess marine aerosols have already 342 saturated the cloud responses, resulting in minor changes in SW CLD. In areas with weak winds, the potentials for direct aerosol scattering are not fully exploited due to the relatively small amounts of sea-343 344 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 345 different injection methods in the five ocean regions. The SW CLD responses are stronger in the three 346 regions of NP, SP, and SA, while they are weaker in the regions of WP and Equa, and in some locations 347 they even led to a reduction of the upward shortwave radiation (Fig. S5). The spatial distributions of the 348 SW CLD responses exhibit noticeable differences, reflecting significant regional differences in the non-349 350 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 351 on cloud formations and distributions, simulation results related to clouds show significant spatial 352 353 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. 354

355 In contrast, the spatial distributions of the SW AER response are smoother, leading to consistent increases in upward shortwave radiation at the TOA in all ocean regions (Fig. S6). This indicates smaller 356 spatial limitations in the distributions of aerosol particles, allowing direct scattering effects to take place 357 everywhere. The direct scattering effect of aerosols is primarily related to the concentrations and physical 358 properties of the particles (discussed below), unlike clouds, which are influenced by multiple variables. 359 360 These results suggest that when implementing geoengineering measures, it is essential to comprehensively consider the interactions between aerosols and clouds, as well as their different response 361 patterns in various regions. 362

The SW CLD response resulting from the injection of sea-salt aerosols in the sensitive areas of five 363 ocean regions exhibits significant spatial differences. The SW CLD response is larger than the SW AER 364 response in the sensitive areas of NP, SP, and SA, indicating that the changes in SW TOT are mainly 365 driven by the cloud radiative response (Fig. 5). In contrast, the SW CLD response is smaller in the WP 366 367 and Equa regions. This is because of the low cloud cover in the Equa, and it is also worth noting that the cloud in the WP is centrally distributed in the northern part of the region, and its SW CLD response is 368 larger in the north. This regional difference is similar to that observed with uniform injection across the 369 entire region. The SW AER response shows consistent results in all areas, resulting in a radiation 370 response change of 3.58–5.44 W m⁻² within the injection areas. In the WP and Equa, the variations in 371 372 SW TOT are primarily driven by the direct scattering effects of aerosols. Aerosols can have a greater impact on radiation responses outside the sensitive areas through transports and diffusions, reaching up 373 374 to three times the total radiation within the sensitive areas (Fig. 6). In all regions except WP, the total SW CLD response outside the sensitive region was about 270%-408% higher than inside. In WP, the 375

SW CLD response outside the sensitive area has a negative effect. The SW CLD responses in NP, SP, 376 and SA extend to the west and northwest of the injection due to the prevailing winds, indicating that 377 clouds in these areas are affected by the injection of sea-salt aerosols (Fig. 5). Changes in cloud 378 microphysical properties will be presented later. The SW CLD variations in other directions are not 379 uniform, and there is negative SW CLD responses in some grids, which again reflected the spatial 380 complexities of cloud radiation effects. The direct scattering effects of aerosols on areas outside the 381 sensitive region is reflected in a widespread increase in upward shortwave radiation at the TOA. The total 382 SW AER responses outside the sensitive areas in the five ocean regions are approximately 160%–281% 383 higher than inside, but lower than the impacts of SW CLD responses outside the sensitive areas. The 384 SW AER and SW CLD responses have similar spatial distributions due to the transport of the aerosols. 385

386 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 387 388 driven by SW CLD responses. As the injected sea-salt aerosols increased, the SW CLD responses gradually reach saturation. After reaching a certain injection level, the increases of SW CLD responses 389 stabilize at its maximum value and no longer increases with further injections. The SW CLD responses 390 show large differences in the five ocean regions, and the different shapes and slopes of the curves indicate 391 that the cloud radiative forcing responses to the sea-salt aerosol injections are different in each region. 392 This might be due to variations in cloud types, cloud amounts, and atmospheric conditions in the different 393 regions. In the NP, SP, and SA, the SW_CLD responses exceed 10 W m⁻², while in WP, it saturates at 5 394 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 0.5 395 W m⁻², and even when the injection doubled, the SW CLD response remained at 0.5 W m⁻². This implies 396 that the SW TOT at Equa is almost exclusively from the contributions of the direct scattering effects of 397 398 aerosols.

In contrast to SW_CLD, the SW_AER responses increase linearly with the injections of sea-salt aerosols ($R^2 > 0.99$). As the injection increases, the contributions of SW_AER to SW_TOT gradually increase, surpassing the SW_CLD responses, and show the same trends across the five regions. This implies that at higher injection levels, the contributions of SW_CLD to total radiation change saturate, and cloud properties no longer significantly change. At this point, sea-salt aerosols primarily affect radiation through direct scattering effects, and the aerosol particles' ability to scatter solar radiation continued to increase with the increases in aerosol quantities. In some cloud-free regions or weather 406 conditions, injected sea-salt aerosols are still able to cool through direct scattering.

There exists a specific injection level at which the SW CLD and SW AER responses are equal. In 407 the NP region, when the injection level is approximately 1.55×10^{-9} kg m⁻² s⁻¹, both SW CLD and 408 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} 409 ⁹ kg m⁻² s⁻¹, respectively. While in WP, the responses were already equal when the injection amount was 410 0.15×10⁻⁹ kg m⁻² s⁻¹. Since there is a saturation of the cloud radiation effects, E_{MCB} decreases with the 411 increases in sea-salt aerosol injection amounts (Fig. 7, red dashed line). This can also explain the higher 412 E_{MCB} of the Natural×5 and Wind-adjusted strategies with relatively low injection amounts (Fig. 3b). The 413 lower E_{MCB} of the Fixed-wind-adjusted injection relative to the fixed uniform injection therefore indicates 414 that wind-dependent injection strategies led to the injection of large amounts of sea-salt aerosols in certain 415 416 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. 417

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 depended on the increases in SW_AER responses, leading to a decrease in E_{MCB} (Fig. 7) Therefore, implementing geoengineering with sea-salt aerosol injections required considering local atmospheric conditions and balancing the relationships between cooling goals and sea-salt injection efficiencies.

424 Under clear and cloudless conditions, injecting sea-salt aerosols could still increase the SW TOT through direct scattering, and this effect exceeds those of aerosol direct scattering when clouds are present. 425 The variation of the upward shortwave radiation flux at the TOA under the clear-sky conditions 426 (SW AER CLR) does not exhibit significant regional heterogeneity across the ocean areas (Figs. 5 and 427 428 S7), suggesting that the contribution of direct aerosol scattering is more uniform globally when considering the effects of sea-salt injections on the Earth's radiation budget. The SW AER CLR 429 responses are also linearly correlated with the injection of sea-salt aerosols ($R^2 > 0.99$), and it exceeds the 430 SW AER responses (Fig. 7). This is because cloud layers also scatter and absorb solar radiation, so this 431 432 scattering effect is more significant under clear sky conditions. This is reflected in the fact that in regions with higher cloud fractions, such as the NP, SP, and SA regions, the differences between the SW AER 433 and SW AER CLR responses are also larger (Fig.7). When injecting sea-salt aerosols in sensitive areas, 434 435 the spatial distributions of SW AER CLR and SW AER responses are highly consistent (Fig. 5). 436 Therefore, injecting sea-salt aerosol under conditions of low cloud covers or clear skies also increases the

437 upward shortwave radiation flux at the TOA.

438 **3.4 Factors affecting the radiation effects.**

439 Uniform injections of 10^{-9} kg m⁻² s⁻¹ sea-salt aerosols led to an increase in aerosol optical depth (AOD) of 440 0.20–0.37 in all regions (Fig. 8). The distributions of AOD within the regions are not uniform due to 441 aerosol transports and diffusions, with some areas showing an increase in AOD of over 0.6. Injecting sea-442 salt aerosols in sensitive areas lead to an AOD increase of 0.077–0.12, while outside the injection areas, 443 AOD gradually decreases as the aerosols transport and disperse. With the increases in sea-salt aerosol 444 injections, AOD shows a linear increase within a certain range in all five ocean regions (R² > 0.998, Fig. 445 9a).

In the regions with more cloud cover, such as NP, SP, and SA, injected sea-salt aerosols significantly 446 increases cloud fraction (Fig. 2, third column and Table 1), leading to the formations of more clouds or 447 expanding the coverage, vertical thickness and lifetime of existing clouds (Goddard et al., 2022). Taking 448 the SP region as an example, Fig. 10 demonstrates that uniform injections of 10⁻⁹ kg m⁻² s⁻¹ sea-salt 449 aerosols significantly increases the CDNC. More cloud droplets capture more water vapor, leading to an 450 451 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%, 452 and ultimately increases the mean cloud albedo over the region by 0.19 (~64%). Similarly, injecting sea-453 salt aerosols in the NP and SA regions led to average cloud albedo increases of 0.17 and 0.20, respectively, 454 while in the WP and Equa, the increases are 0.15 and 0.13, respectively (Figs. S8–S11). The injection of 455 456 sea-salt aerosols uniformly within the sensitive areas results in smaller effects on cloud microphysical 457 properties compared to uniform injections across the entire region, even though the total injection amount 458 within the sensitive areas is the same in both scenarios. This is because when sea-salt aerosols are injected 459 across the entire region, the surrounding sea-salt aerosols affect the sensitive areas through transports, 460 resulting in an enhanced 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 461 transports, increases the average cloud albedo across the entire area by 0.032 over the entire region and 462 463 by 0.12 within the sensitive regions, which is less than the effects of injection across the entire area (Fig. S12). Similarly, injecting sea-salt aerosols in the sensitive areas of other ocean regions lead to average 464 cloud albedo increases of 0.015–0.024 across the entire area, with increases of 0.11 in the sensitive areas 465 of the SP and SA regions, and increases of 0.090 and 0.10 in the WP and Equa, respectively (Figs. S12-466

467 S16).

468 **3.5 Drivers of SW_CLD responses.**

The cloud radiation forcing (CRF) parameters are used to calculate the effects of changes in cloud cover 469 and cloud albedo on the SW CLD responses due to the injections of sea-salt aerosols. Figure S17 470 illustrates the increase in the CRF parameter coinciding with the increases in the SW CLD responses 471 after uniform injection of sea-salt aerosols in the five regions (Fig. S5, third row). The CRF'param 472 calculated using the perturbation method indicates that in the five ocean regions, CRF'param is primarily 473 driven by perturbations in cloud albedo (Fig. S18 first column), and it surpasses the changes in cloud 474 fractions and their interactions. Cloud albedo changes explain over 70% of the CRF'param in all five regions 475 except the Equa. The contribution of cloud fraction changes ranges from 13.9% to 23.7%, while the 476 interactions between the two factors account for only about 10% (Fig. S18, second and third columns). 477

Figure 11 evaluates the relative effects of Twomey, LWP, and cloud fractions on the SW CLD 478 responses after uniform injecting sea-salt aerosols in five ocean regions. The results indicate that changes 479 in CDNC (Twomey effect) and LWP are the main drivers of SW CLD responses, while changes in cloud 480 fraction contribute minimally to the SW CLD responses. Except for the Equa region, changes in CDNC 481 and LWP accounted for 48.4%-52.5% and 39.0%-41.7% of the SW CLD changes, respectively, with 482 cloud fraction changes contributing to less than 10.0% (Fig. 11). The results are similar for injections in 483 sensitive areas, with changes in CDNC and LWP contributing similarly and more than changes in cloud 484 fractions to SW CLD (Fig. S19). The changes in SW CLD responses after aerosol injections in the 485 sensitive areas of Equa are mainly contributed by LWP effects (~70%). 486

Uniform injections of sea-salt aerosols at a rate of 10⁻⁹ kg m⁻² s⁻¹ produced susceptibilities ($\frac{\Delta \alpha}{\Delta \ln AOD}$) 487 ranging from 0.00030 to 0.0035 in the five regions, with corresponding spatial distributions shown in Fig. 488 11. NP, SP, and SA regions exhibit cloud responses that are more sensitive to aerosol injections in most 489 of the region, with susceptibilities ranging from 0.0028 to 0.0035. The Equa shows the lowest 490 susceptibility, indicating that the system is less responsive to variations in aerosol injections. It is 491 noteworthy that although the average susceptibility in the WP region is 0.0013, the higher susceptibility 492 values are concentrated in the north of 35°N, where the average susceptibility is 0.0026, similar to those 493 of the SP region, suggesting that clouds here are more susceptible to aerosol injections. Injecting sea-salt 494 495 aerosols in sensitive areas mostly results in cloud responses that are located outside the sensitive areas (Fig. S19). Injecting sea-salt aerosols in the sensitive areas of SP and SA have a greater impact on the 496

497 northwest. In the sensitive areas of NP, injecting sea-salt aerosols have a larger impact on the west. In the 498 WP, the injection of sea-salt aerosols into the sensitive area does not fully reflect its susceptibility because 499 we choose to calculate the sensitive areas away from the boundary, and the greatest susceptibilities in the 500 WP region happens to be in the northern part of the region near the boundary.

501 **4. Discussions and conclusions**

Many studies have discussed the contributions of both the direct and indirect effects of MCB. Some 502 503 studies suggest that MCB primarily relies on the indirect effects, as originally conceived, i.e., injecting aerosols to brighten clouds (Jones and Haywood, 2012; Latham et al., 2012). Other studies proposed that 504 the direct scattering effects of aerosols may be more important (Ahlm et al., 2017; Kravitz et al., 2013; 505 Mahfouz et al., 2023; Niemeier et al., 2013; Partanen et al., 2012). Our results indicate that the 506 507 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 508 509 early stage of MCB implementations, changes in radiative response are mainly driven by indirect effects, causing clouds to brighten easily. As the injection of sea-salt aerosol increases, the radiative effect on 510 511 clouds saturates, and the clouds are difficult to brighten. In contrast, the direct effect continued to increase linearly, leading to a subsequent decrease in the efficiencies of MCB. Partanen et al. (2012) first 512 considered the relative importance of aerosol direct and indirect effects in MCB and preliminarily found 513 the saturated non-linear phenomenon of indirect effects at high CDNC, as well as the linear relationships 514 between direct effects and injection amounts. Haywood et al. (2023) also found a decrease in MCB 515 efficiency with increasing aerosol injections. Regions initially susceptible to cloud brightening gradually 516 became less susceptible, and aerosol direct radiation effects dominated. Other General Circulation Model 517 (GCM) studies also found similar results (Alterskjær and Kristjánsson, 2013; Rasch et al., 2024; Stjern et 518 519 al., 2018).

This study highlights and quantifies these findings in a regional model for the first time, showing the changing trends of direct and indirect effects with injection amounts in the different ocean regions. This study provides more detailed cloud composition changes due to sea-salt aerosols injection. The model achieves higher droplet nucleation rates at higher resolution due to increased subgrid vertical velocity and higher aerosol concentrations (Ma et al., 2015). The best results are obtained in regions with persistent stratocumulus clouds (e.g., the oceans along the west coast of the continent), where the injected 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 scattering effect of aerosols. Considering the uncertainty in the model's resolution of clouds and the fact that, in reality, the cloud distributions are also greatly influenced by the local meteorological conditions, the direct scattering effects of sea-salt aerosols on MCB contributions are relatively certain. Therefore, in cloud-free or less cloudy regions, the direct effect of aerosols becomes more important.

In the early stages of Earth-System modeling studies, the MCB processes were often simulated by presetting CDNC = 375 or 1000 cm⁻³ in the lower regions of the ocean (Jones et al., 2009; Latham et al., 2008; Rasch et al., 2009). However, many follow-up studies have suggested that injections of sea-salt aerosols have difficulty to produce a uniform CDNC field due to aerosol dilutions, depositions, and the dependences of the spray rate on wind speed. The CDNC is highly variable spatially, and studies have even reported reductions in CCN and CDNC caused by the injections of sea-salt aerosols (Alterskjær et 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 539 average CDNC concentrations for five ocean regions range from 60.2 to 100 cm⁻³, and the spatial 540 distributions are uneven (Fig. 10 and Figs. S8-S11). Figure 9b indicates that the CCNs in the five regions 541 increase linearly $(R^2 = 1)$ with increasing sea-salt aerosol injections, but not all of the CCNs are converted 542 to cloud droplets. After doubling the injection amounts, the regional average CDNC is 84.8–130 cm⁻³, 543 with only some grid points exceeding 200 cm⁻³ within the regions. When the injection amounts are 544 increased to 3×10^{-9} kg m⁻² s⁻¹, the regional average CDNC is 98.8–140 cm⁻³. This implies that injecting 545 more sea-salt aerosols at this point does not result in more cloud droplets, and the conversion of CCN into 546 cloud droplets is less efficient, which slows the CDNC growths and tends to saturation (Fig. 9c). 547

Our findings align with Alterskjær et al. (2012), who injected sea-salt aerosols at the same rate (10-548 ⁹ kg m⁻² s⁻¹) and observed the average CDNC below 375 cm⁻³ due to competitive effects and reduced 549 aerosol activation. Notably, however, Wood (2021) found that decreased activation due to competition 550 may be overestimated in the Abdul-Razzak and Ghan activation parameterization used in many GCMs 551 552 relative to a parcel model. Partanen et al. (2012) used wind-adjusted injections and reported CDNC values of 596-784 cm⁻³, with even higher values (>1000 cm⁻³) for smaller-sized aerosols, attributing this to 553 554 overestimations of particle solubility and size. Hill and Ming (2012) increased sea-salt aerosol concentrations by a factor of five, raising CDNC from 68 to 148 cm⁻³ at 850–925 hPa. It is noteworthy 555 556 that Hill and Ming (2012) increased all modes of sea-salt aerosols. Many studies have reported that selecting the appropriate injection particle size is crucial for MCB (Andrejczuk et al., 2014; Hoffmann 557

and Feingold, 2021; Partanen et al., 2012), and injecting Aitken and coarse modes may even lead to a 558 positive forcing with CDNC decreasing (Alterskjær and Kristjánsson, 2013). However, Wood (2021) 559 560 argued that particles with a geometric mean dry diameter of 30-60 nm were most effective in brightening cloud layers, and Goddard et al. (2022) similarly found that injecting Aitken mode sea-salt aerosols 561 generated larger radiative flux changes compared to accumulation mode. There are still considerable 562 563 discussions about choosing the appropriate aerosol particle sizes during the implementation of MCB, with different models and parameterization schemes providing different recommendations. The sensitivity of 564 MCB to particle size is not considered in this paper and was left for future research. 565

In this study, the injection of 10⁻⁹ kg m⁻² s⁻¹ accumulation mode sea-salt aerosols increases cloud 566 albedo in the five ocean regions by 0.13–0.20, with a local maximum of more than 0.3. After doubling 567 the injection amounts, the regional average cloud albedo reaches 0.45-0.55, representing a cloud albedo 568 change of 0.15-0.24 (Fig. 9d). Bower et al. (2006) suggested that to compensate for the warming 569 associated with doubling atmospheric CO₂ concentrations, a cloud albedo change of 0.16 was needed in 570 three stratocumulus cloud regions (off the west coast of Africa and North and South America, representing 571 3% of global cloud cover). The cloud albedo changes produced by the injected aerosols in this study 572 573 achieved the targets envisioned in previous studies. Wood (2021) proposed seeding Aitken mode particles in approximately 9% of the ocean to achieve a corresponding cloud albedo increase of 0.16. It was also 574 suggested that injecting sea-salt aerosols in a clean, undisturbed state would produce more brightening 575 (Wood, 2021). Fig. 9d confirms this finding, indicating that clouds are more likely to brighten in the early 576 stages of sea-salt aerosol injection, and the efficiency of cloud brightening decreases with increasing 577 injection amounts. Goddard et al. (2022), simulating injecting accumulation mode sea-salt aerosols in the 578 579 central Gulf of Mexico, achieved a simulated cloud albedo change of approximately 0.1 in the main 580 impact region, while switching to Aitken 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), 0.062 581 582 (Latham et al., 2008), or 0.074 (Lenton and Vaughan, 2009) were estimated.

The contributions of the change in cloud fractions to the SW_CLD responses in this study are small, 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 those of the CDNC and LWP (Chen et al., 2014; Rosenfeld et al., 2019). Goddard et al. (2022) believe that this was due to the fact that the regional atmosphere was wetter during the simulation periods and that the relative contributions of changes in cloud fraction to the SW_CLD response would be expected

to increase in drier months. Three of the five ocean regions in this study, SA, SP, and NP are much drier 589 and more stable than the Gulf of Mexico simulated by Goddard et al. (2022) (Fig. S20). Furthermore, 590 when we switched to conducting the experiments again in the dry months of the same year, the 591 contribution of cloud fraction to SW CLD did not change much, remaining at ~10%. We believe that this 592 might be a difference due to the parameterization scheme or resolution of the model. Liu et al. (2020) 593 594 simulated with WRF-Chem model and found that the cloud fraction susceptibilities to aerosols in Morrison scheme and the Lin scheme were only about half of those observed by Moderate Resolution 595 Imaging Spectroradiometer (MODIS). The neglected subgridded clouds in the 12-km resolution 596 simulations might lead to an underestimation of the radiative effects of clouds (Yu et al., 2014). In addition, 597 cloud fractions are more commonly underestimated in the model (Glotfelty et al., 2019), and using an 598 updated parameterization scheme that accounts for subgrid condensation might improve the model's 599 ability to resolve clouds (Zhao et al., 2023). The high spatial variabilities of cloud radiation effects 600 emphasize the need for improved resolution in future model studies of cloud-aerosol interactions. The 601 effects of finer resolution and more parameterization schemes on aerosol-cloud interactions still need to 602 be verified. Considering the difficulties of modeling to accurately capture the effects of cloud fractions 603 604 on radiation, the actual effects of MCB may be underestimated.

This study provides quantifiable data on cloud and radiation changes for the implementation of MCB 605 over five ocean regions, and an optimization scheme on the injection strategy by adjusting the injection 606 amounts and selecting sensitive areas. It is noteworthy that different parameterization schemes, models, 607 and resolutions can influence results, especially the cloud feedback on the injected sea-salt aerosols, 608 which is a major reason for discrepancies between models (Stjern et al., 2018). In Earth-system model 609 610 studies, there has been a rich discussion of the climate and ecological impacts of the MCB with the same framework under the Geoengineering Model Intercomparison Project (GeoMIP) (Rasch et al., 2024). 611 However, there is still a lack of a unified framework for mid-scale MCB research. 612

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615 Data and code availability

- 616 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.

618 *Author contributions*

- 619 SY, DR and ZS conceived and designed the research. ZS performed the model simulations. SY and ZS
- 620 conducted data analysis. SY, ZS, PL, NY, LC, YS, BJ and DR contributed to the scientific discussions.
- 621 SY and ZS wrote and revised the manuscript.
- 622 Supplemental information.
- 623 The supplementary information related to this article is available online.
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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	12.8	19.8	8.91	143
NP	0.0678	0.0760	9.67	60.2	24.6	43.9	7.18	126
Equa	0.0051	0.0059	17.5	83.4	0.85	1.39	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	0.46	0.35	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	1.59	0.08	0.14
	SA	1.4	1.26	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	0.27	1.2	1.2
	SP	7.6	5.8	1.8	2.6
	SA	8.0	5.9	2.1	2.8
	WP	18	4.6	13	15
	NP	23	13	9.8	15
10 ⁻⁹ kg m ⁻² s ⁻¹	Equa	11	0.55	10	11
	SP	25	11	14	19
	SA	22	11	11	15
	WP	6.9	2.9	4.0	5.1
T ' 1 ' 1	NP	16	11	5.1	7.8
Fixed-wind-	Equa	5.0	0.50	4.5	4.7
adjusted	SP	17	9.9	6.6	9.8
	SA	20	11	9.1	13

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	$\alpha_c' \bar{f}$	$\bar{lpha}_c f'$	$\alpha'_c f'$	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%	5.09%	
SP	73.8%	15.9%	10.3%	51.8%	39.0%	9.19%	
SA	77.3%	13.9%	8.81%	52.5%	39.7%	7.78%	

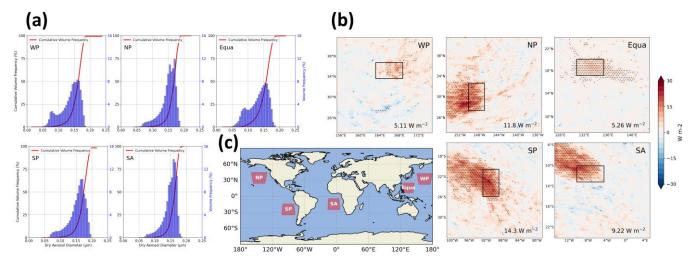


Figure 1. Injecting sea-salt aerosols into five open sea regions to simulate the implementation of MCB geoengineering. (a) The cumulative volume frequency of increased aerosol dry particle size (uniform injection of 10^{-9} kg m⁻² s⁻¹ sea-salt aerosols over the entire region). (b) Differences (Exp - 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.

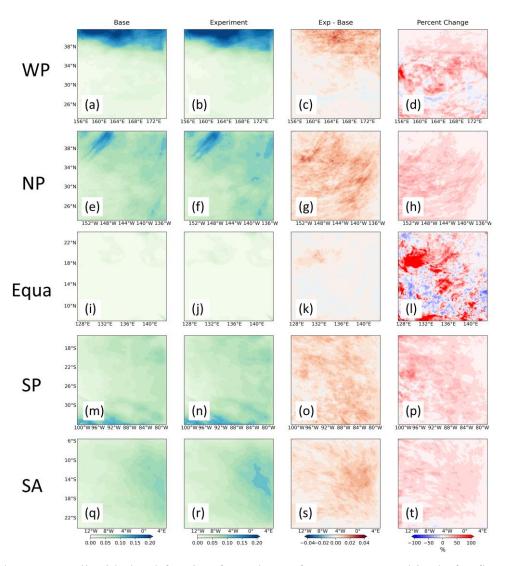


Figure 2. Column mean liquid cloud fraction from the surface to 3000 m altitude for five regions. The first to fourth columns are Base, the sensitivity experiment with a uniform injection of 10^{-9} kg m⁻² s⁻¹ seasalt aerosols over the entire region, Exp - Base, and the percent change of Exp - Base,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

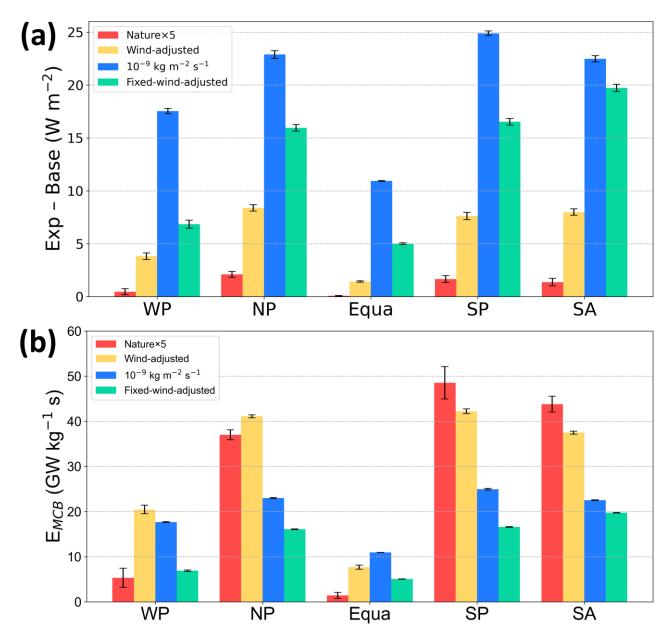


Figure 3. (a) The differences in SW_TOT and (b) the MCB efficiency (E_{MCB}) due to the injection of seasalt aerosols in different strategies in five ocean regions.

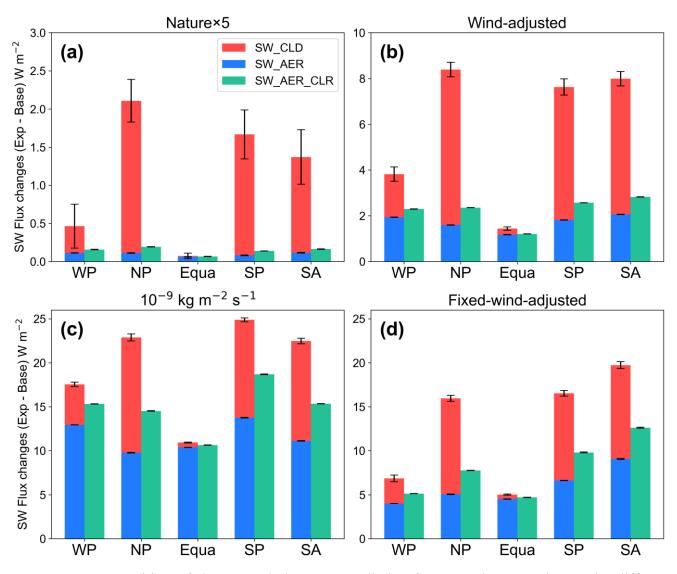


Figure 4. Decomposition of the upward shortwave radiative fluxes at the TOA due to the different strategies of injecting sea-salt aerosols in the five regions. Note that the y-axis ranges are not consistent.

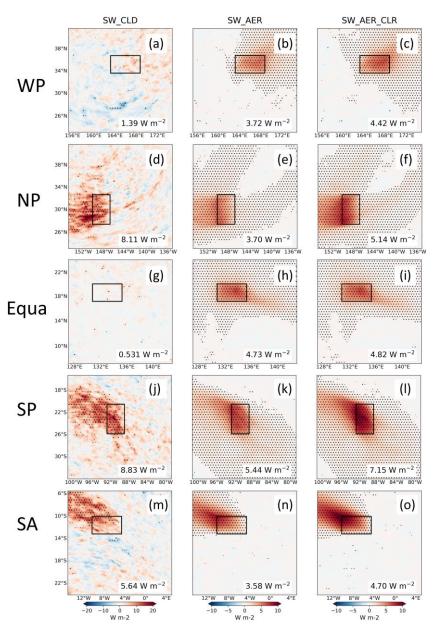


Figure 5. Spatial distribution of SW_CLD (first column), SW_AER (second column), and SW_AER_CLR (third column) responses resulting from the injection of 10⁻⁹ kg m⁻² s⁻¹ sea-salt aerosols in the sensitive areas over five ocean regions. The values of the radiative flux responses generated only in the sensitive 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.

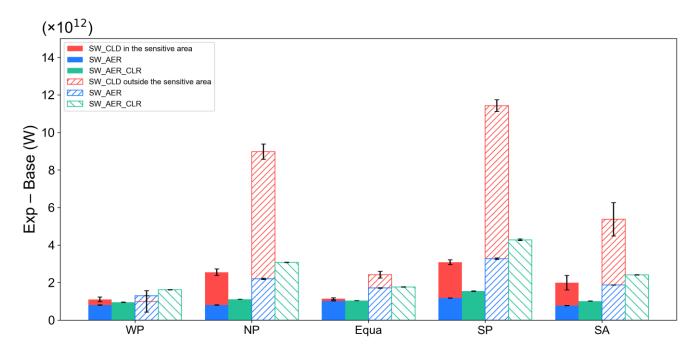


Figure 6. Total SW_CLD, SW_AER, and SW_AER_CLR responses resulting from the injection of 10⁻⁹ kg m⁻² s⁻¹ sea-salt aerosols within the sensitive areas of the five regions. The solid columns indicate the total radiative response calculated for aerosol injection within the sensitive areas. Columns filled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

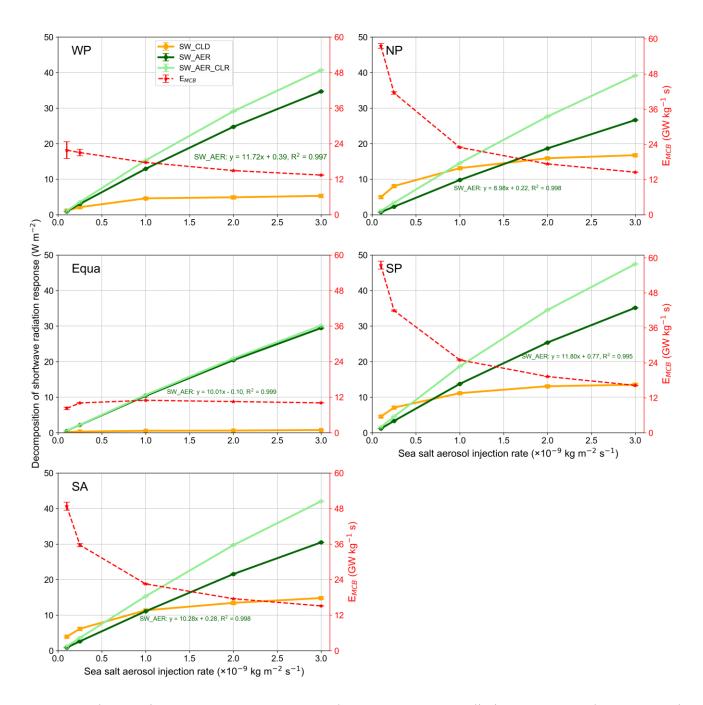


Figure 7. Changes in SW_CLD, SW_AER, and SW_AER_CLR radiative responses due to sea-salt aerosols uniform injected in varying amounts in five ocean regions, and corresponding changes in E_{MCB}. SW_AER and SW_AER_CLR are labeled with the results of the corresponding linear regression analysis. Error bars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

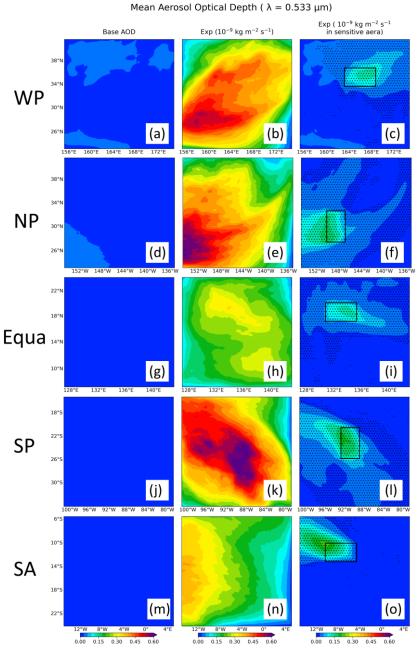


Figure 8. Spatial distribution of mean AOD ($\lambda = 0.533 \ \mu m$) for five ocean regions. The first column is the AOD for Base, the second column is the AOD after uniform injection at $10^{-9} \text{ kg m}^{-2} \text{ s}^{-1}$, and the third column is the AOD after uniform injection in sensitive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

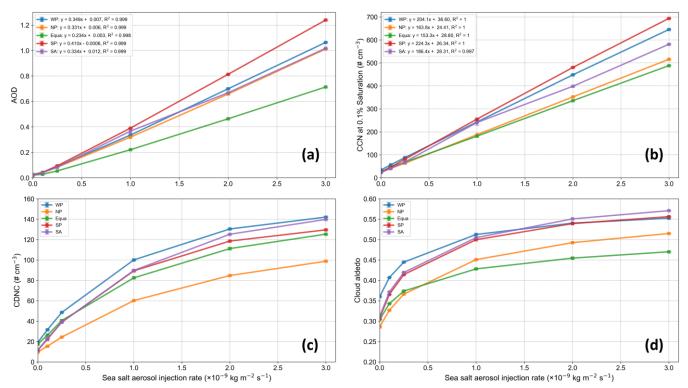


Figure 9. Relationship between changes in regional mean (a) AOD, (b) CCN, (c) CDNC, and (d) cloud albedo due to uniform injection of sea-salt aerosols across the region and the amounts of sea-salt aerosols injected. The results of the linear regression of (a) AOD and (b) CCN on the sea-salt aerosols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

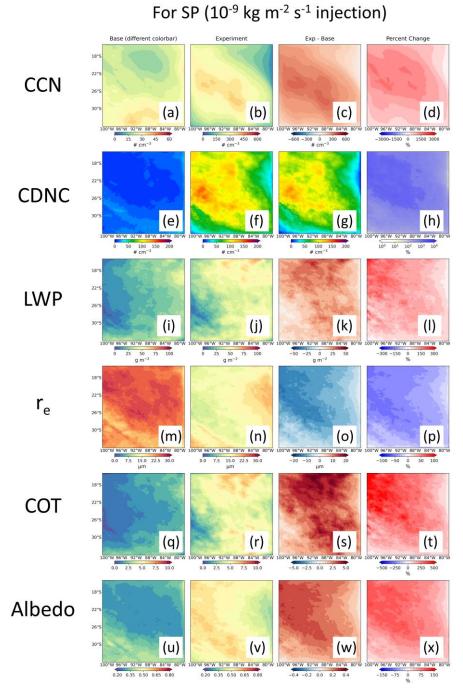


Figure 10. Spatial distribution of liquid cloud property responses after uniform injection of sea-salt aerosols with 10^{-9} kg m⁻² s⁻¹ in the SP region. Results are shown for cloud condensation nuclei (CCN, S = 0.1%, # cm⁻³), cloud droplet number concentration (# cm⁻³), liquid water path (LWP, g m⁻²), 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.

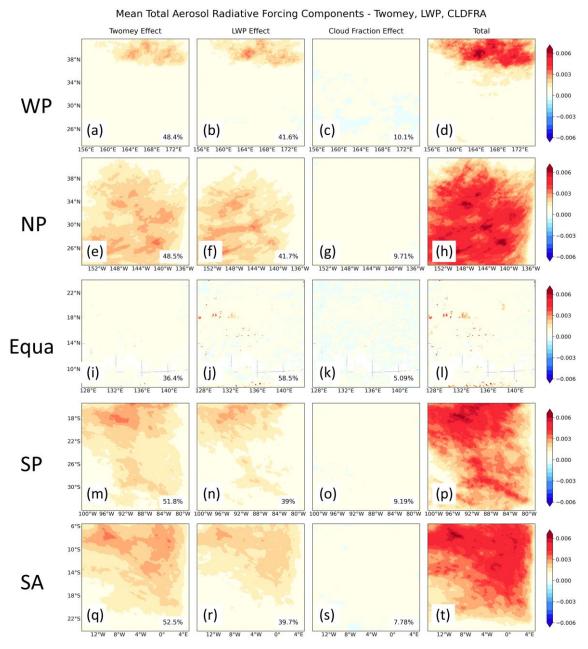


Figure 11. Spatial distribution of cloud property changes in response to SW_CLD radiation after uniform injection of sea-salt aerosols in five regions. The first column is the Twomey effect, the second column is the LWP effect, the third column is the cloud fraction effect, and the fourth column is the 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.