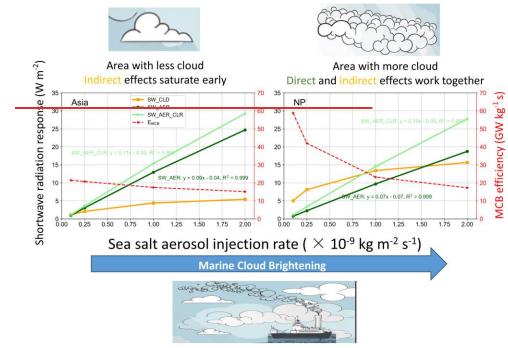
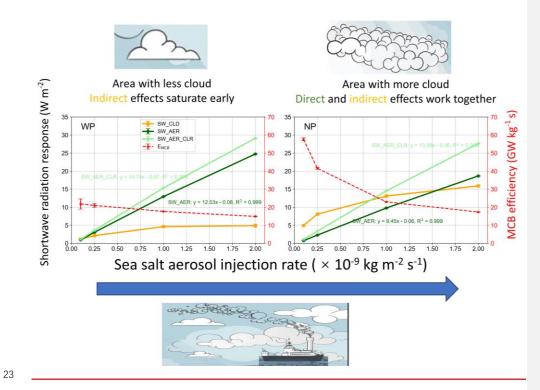
1	The effectiveness of solar radiation management for marine cloud brightening
2	geoengineering by fine sea spray in worldwide different climatic regions
3	
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# 21 Graphic Abstract





## 25 Abstract

Marine Cloud Brightening (MCB) geoengineering aims to inject aerosols over oceans to brighten 26 clouds and reflect more sunlight to offset the impacts of global warming or to achieve localized climate 27 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 conducted 30 experimentsdesigned model simulations with injected sea-salt aerosols in the same framework for five open oceans around the globe. Our results show that a uniform injection strategy that diddoes not depend 31 on wind speed captured the sensitive areas of the regions that produced the largest radiative perturbations 32 33 during the implementation of MCB. When the injection amounts were are low, the sea-salt aerosols 34 dominated dominate the shortwave radiation mainly through the indirect effects of brightening clouds, 35 showing obvious spatial heterogeneity. As the indirect effectseffect of aerosols saturatedsaturates with increasing injection rates, the direct effects still increased effect increases linearly and exceeded exceeds 36 37 the indirect effects, producing a consistent increase in the spatial distributions of top-of-atmosphere 38 upward shortwave radiation. Our research emphasizes that MCB was best implemented in areas with 39 extensive cloud cover, while the aerosol direct scattering effects remained dominant when clouds were 40 searceThis study provides quantifiable radiation and cloud variability data for multiple regional MCB implementations and suggests that injection strategies can be optimized by adjusting injection amounts 41 and selecting sensitive areas in the simulations of regional models. 42

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

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## 47 1. Introduction

As global temperatures continue to rise, the international community is facing an unprecedented challenge 48 to achieve the ambitious goal set in the Paris Agreement of limiting global warming to within 1.5 °C 49 50 (Mengel et al., 2018). Even the recent One of the key outcomes of the recently concluded 28th Conference 51 of the Parties (COP28) proposing to phase out all fossil fuels from the current energy system to 52 achievewas the global goal on adaptation and its framework, it is also recognized completion of the first 53 Global Stocktake (GST), a mid-term assessment of the progress made by countries toward achieving the climate goals of the Paris Agreement. However, the report highlighted that our-current efforts are still 54 insufficient to meet the COP28 goals (https://www.cop28.com/).to reduce emissions had fallen short of 55 the intended targets (https://www.cop28.com/). Against this backdrop, scientists are turning their 56 57 attentionsattention to more radical and innovative geoengineering methods, by attempting to reduce or offset the impacts of climate change through artificial interventions in the climate (Visioni et al., 2023). 58 59 Some geoengineering methods seek to capture or remove CO<sub>2</sub> from the atmosphere to increase carbon sinks, while others focus on modifying solar radiation, reducing incoming solar shortwave radiation, or 60 61 reflecting more sunlight to cool the earth, known as solar radiation management (SRM) (Lenton and 62 Vaughan, 2009). Among these, marine cloud brightening (MCB) has a certain realistic basis and is considered the most likely geoengineeringSRM method for regional applications (Latham et al., 2014). It 63 has been observed that exhaust emissions from ocean-going vessels can lead to brighter clouds, with clear 64 ship tracks also visible from satellites, and MCB aims to replicate this effect by spraying sea-salt aerosols 65 66 (Chen et al., 2012).

67 Aerosol-cloud interactions and their impacts on climate are complex (Rosenfeld et al., 2014, 2019). 68 Injected sea-salt aerosols affect clouds through indirect effects (Paulot et al., 2020). In the case of a constant liquid water content, an increase in cloud droplet number concentration (CDNC) decreases the 69 cloud droplet size, increases the total surface area of cloud droplets, thereby enhancing the cloud albedo, 70 71 forming brighter clouds, and reflecting more sunlight back to space (the first indirect effect or Twomey 72 effect) (Twomey, 1974). At the same time, the decrease in cloud droplet size suppresses precipitation, thereby increasing the cloud's lifespan and optical thickness (the second indirect effect of aerosols) 73 (Albrecht, 1989). Reducing the cloud drop size induces a faster evaporation and loss of cloud water. 74 75 However, the effect of the coarse part of the sea spray aerosols has an opposite effect that offsets the loss of liquid water passpath (Liu et al., 2022). In addition, those aerosols that are not injected aerosols into the 76 77 clouds scatter more sunlight back into space through the direct scattering effect (Ahlm et al., 2017; **设置了格式:**字体颜色:黑色,英语(美国)

Partanen et al., 2012; Zhao et al., 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.-MCB has unique advantages compared to other geoengineering schemes. For example, the sprayed aerosols are non-polluting, and can be applied locally to change the regional climate.

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. Therefore, most of the current literatures examine the environmental and climate impacts of MCB implementation through modeling.

90 Table S1 summarized summarizes the results of current modeling experiments simulations on MCB 91 and similar spraying of with sea-salt aerosols, as well as their implementation strategies. Most MCB 92 studies useduse Earth-System Models to assess the impacts of the implementation of MCB on climate. 93 Early MCB studies assumed the effects of MCB implementation by setting a fixed CDNC or directly modifying the cloud effective radius (re), ignoring the processes such as generation, transport, dry and 94 wet deposition, and activation of injected sea-salt aerosols, and not including the direct radiative effect of 95 aerosols. With the development of models, researchers started to conduct more detailed studies by 96 injecting aerosols or increasing sea-salt aerosol emissions, taking into account the post-treatmentinjection 97 98 processes of aerosols mentioned above. The implementation region of MCB is crucial, and existing 99 research. Existing studies have focused on the impacts of implementing-MCB implementation in three 100 key areas: open oceans globally, the equatorial region (between 30°S and 30°N), and coastal areas with 101 widespread marine stratocumulus clouds. Alterskjær et al. (2012)Alterskjær et al. (2012) used the cloudweighted susceptibility function to find the most sensitive regions to the injection of sea-salt aerosols. 102 103 Similarly, Jones and Haywood (2012)Similarly, Jones and Haywood (2012) determined the 10% of the marine regions globally most suitable for implementing MCB through an iterative method. The 104 contributions of direct and indirect effects of aerosols during the implementation of MCB are still 105 106 controversial and quantitative assessment of both is lacking.

Here, we <u>useduse</u> the two-way coupled <u>Weather Research and Forecasting - Community Multi-scale</u>
 <u>Air Quality model (WRF-CMAQ-model,)</u>, combined with previous studies on the region and injection

strategies, to implement MCB in five open oceans worldwide. This study <u>simulatedsimulates</u> 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.

113

## 114 2. Experiments and methods

## 115 2.1 Model configuration

The two-way coupled Weather Research and Forecasting (WRF v3.4) - Community Multi-scale Air 116 Quality (CMAQ v5.0.2) model that considered both direct and indirect effects of aerosols was used in 117 this study43. In the two-way coupled model, acrosols predicted by CMAQ wereWRF (v3.4) - CMAQ .18 (v5.0.2) model that considers both direct and indirect effects of aerosols was used in this study (Yu et al., 119 120 2014). In the two-way coupled model, aerosols predicted by CMAQ are able to affect clouds, radiation, 121 and precipitation simulated by WRF in a consistent online coupled manner (Wong et al., 2012). Yu et al., 122 (2014)Yu et al. (2014) further extended the two-way coupled WRF-CMAQ model by incorporating the 123 aerosol indirect effects (including the first, second, and glaciation aerosol indirect effects), improving the 124 ability of the WRF-CMAQ model to predict clouds and radiation. Wang et al. (2021)Wang et al. (2021) 125 validated this model.

126 The physical schemes of the WRF model are the same as those set in Yu et al. (2014), Yu et al. (2014), including the asymmetric convective model (ACM2) for a planetary boundary layer (PBL) scheme (Pleim, 127 128 2007), the Morrison 2-moment cloud microphysics scheme (Morrison et al., 2009)(Morrison et al., 2009), 129 the Kain-Fritsch (KF2) cumulus cloud parameterization, the Rapid Radiative Transfer Model for General 130 Circulation Models (RRTMG) longwave and shortwave radiation schemes, and the Pleim-Xiu (PX) land-131 surface scheme. The meteorological initial and boundary conditions were provided by the National Center for Environmental Prediction (NCEP) final analysis dataset (FNL) with a spatial resolution of 1°×1° and 132 temporal resolution of 6 h. The carbon bond gas-phase chemical mechanism (CB05) and aerosol module 133 of AERO6 were used in the CMAQ model. The anthropogenic emissions were taken from the 134 Hemispheric Transport of Air Pollution (HTAP\_V2) projects (Janssens-Maenhout et al., 2015). The 135 biogenic emissions were estimated by the Biogenic Emissions Inventory System version 3.14 (BEISv3.14) 136 model (Carlton and Baker, 2011). Sea salt emissions were calculated online in CMAQ and were divided 137 138 into open-ocean and surf-zone emissions. In the open ocean, Gong (2003) In the open ocean, Gong (2003) extended the sea-salt aerosol parameterization of Monahan et al. (1986)Monahan et al. (1986) to 139

	140	submicron sizes, with the emission flux being linearly proportional to the ocean area covered by whitecaps,
	141	The geometric mean diameter of accumulation mode sea salt aerosols in the CMAQ ranged from 0.2651
	142	to 0.8187 µm. The particle size distributions of the emitted sea-salt aerosols were CMAQ represents the
	143	atmospheric particle distribution as the superposition of three log-normal modes, the Aitken,
	144	Accumulation, and Coarse modes (Binkowski and Roselle, 2003). The particle size distribution and the
	145	geometric standard deviation of the emitted sea-salt aerosols are adjusted to the local relative humidity
	146	before mixing with the ambient particle modes (Zhang et al., 2005). Surf zone emissions were calculated
	147	using the open ocean-source function of Gong (2003) The geometric mean diameter of accumulation mode
	148	sea-salt aerosols in the CMAQ ranged from 0.2651 to 0.8187 µm, with the geometric standard deviation
	149	constrained between 1.76 and 1.83. Surf-zone emissions were calculated using the open ocean-source
	150	function of Gong (2003), with a fixed whitecap coverage of 100% and a surf-zone width of 50 m. Kelly
	151	et al. (2010)Kelly et al. (2010) provided a detailed description of these processes. In the CMAQ model,
	152	the number concentration emission rate was calculated from the mass emissions rate as follows;
1	192	the number concentration emission rate was calculated from the mass emissions rate as follows.

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

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

154

where *En* was the mass emissions rate for species *n* and  $\rho_n$  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%.

#### 162 2.2 Experimental setup

As summarized in Table S1, the MCB geoengineering implementation areas included the globe, the equator (30°S–30°N), regions with extensive coverages of marine stratocumulus clouds, and so on. Therefore, based on previous experimental designs, we <u>useduse</u> the WRF-CMAQ model to simulate the injections of sea-salt aerosols in the five open ocean regions (Fig. 1c). These regions <u>were Asiaare WP</u> and NP, located in <u>East Asia on</u> the western <u>side of theand northern</u> Pacific Ocean; Equa, <u>situatedlocated</u> in the Philippine Sea along the equator; and NP, SP, and SA, <u>which referred tolocated in</u> the <u>Northsouth</u> Pacific, South Pacific, and Southsouth Atlantic, respectively. These The three regions, NP, SP and SA, are
located along the western coast of continents, were considered to have extensive coverage of marine
stratocumulus clouds and were the most suitable areas for implementing MCB (Alterskjær et al., 2012;
Hill and Ming, 2012; Jones et al., 2009; Partanen et al., 2012; Stuart et al., 2013).

The grid numbers of WRF and CMAQ wereare 190×190 and 173×173, respectively, and both hadhave a horizontal resolution of 12 km, with 29 vertical layers from the surface to about 21 km altitude. The simulation period for the AsiaWP, Equa, and NP regions in the northern hemisphere wasis from July 24, 2018, to September 1, 2018, while for the SP and SA regions in the southern hemisphere, the simulation period wasis from February 24, 2023, to April 1, 2023. The first 8 days of the model simulations wereare considered as the spin-up period to minimize the impacts of initial chemical conditions. 带格式的: 缩进: 首行缩进: 2 字符

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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 tested<u>test</u> four different sea-salt aerosol injection methods<u>strategies</u>, wind-speed-dependent Natural×5, Wind-adjusted, Fixed-wind-adjusted at 10<sup>-9</sup> kg m<sup>-2</sup> s<sup>-1</sup> and Fixed-wind-adjusted uniform injections of sea-salt aerosols at a fixed rate of  $10^{-9}$  kg m<sup>-2</sup> s<sup>-1</sup> All additional injected sea-salt aerosols wereare in the accumulation mode. In this study, the geometrical mean dry diameter of sea-salt aerosols injected into the five regions wasis about 0.11–0.15 µm, and wasis similar for all emission scenarios.

Natural×5: IncreasedIncrease the emission rates of accumulation mode sea-salt aerosols by a factor of 5 (Hill and Ming, 2012). This wasis a simple wind-speed-dependent increase. The injection rates in the five regions wereare equivalent to  $0.03031-0.09085\times10^{-9}$  kg m<sup>-2</sup> s<sup>-1</sup> (Table S2).

Wind-adjusted: Salter et al. (2008) designed a spray vessel for injecting sea salt acrosols with a196spray efficiency that was dependent on wind speed and was expected to achieve maximum spray outputs197at wind speeds between 6 - 8 m s<sup>-1</sup>. The threshold wind speed was set to 7 m s<sup>-1</sup> and the spray efficiency198at lower wind speeds was 1.5 times the wind speed. We used the source function of Partanen et al. (2012)199as follows, where u was the 10 m wind speed. For example, at wind of 7 m s<sup>-1</sup> the injection rate would be

200  $\frac{0.26 \times 10^{-9} \text{ kg m}^{-2} \text{ s}^{-1}}{\text{ kg m}^{-2} \text{ s}^{-1}}$ 

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<sup>-1</sup>. The threshold wind speed was set to 7 m s<sup>-1</sup> 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<sup>-1</sup> the injection rate will be  $0.26 \times 10^{-9}$  kg m<sup>-2</sup> s<sup>-1</sup>.

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

Fixed at 10<sup>-9</sup> kg m<sup>-2</sup> s<sup>-1</sup>: Unlike the previous two injection methods, the injections of sea-salt aerosols 208 at a fixed rate of 10<sup>-9</sup> kg m<sup>-2</sup> s<sup>-1</sup> wereare not dependent on wind speed and increased uniformly over all 209 210 ocean grids. Injecting sea-salt aerosols at a fixed rate identified the geographic areas that were most 211 sensitive to increased sea-salt aerosols and produced the largest top-of-atmosphere (TOA) radiative 212 perturbations (Alterskjær et al., 2012). Many other studies have used this method (Goddard et al., 2022; 213 Horowitz et al., 2020; Mahfouz et al., 2023), Uniform injections of sea-salt aerosols throughout the region 214 ignored aerosol transports and dispersion at the boundary. Therefore, based on the results of a fixed 10-9 215 kg m<sup>-2</sup> s<sup>-1</sup> injection rate, we identified the geographical regions ( $30 \times 50$  grid points, approximately 360 km × 600 km, away from the domain boundary) in five ocean areas where the TOA radiative perturbations 216 217 caused by uniform injection were the largest, and the most sensitive. Table S3 shows the locations of these sensitive regions. The injection amount in the sensitive region at a fixed 10<sup>-9</sup> kg m<sup>-2</sup> s<sup>-1</sup> injection rate is 218 219 found to be about 1/20 of those in the full domain. Fixed-wind-adjusted: To rule out differences in radiative and cloud response due to wind 220

variabilities on spray rates, we <u>performedperform</u> an additional adjustment. Similar to **Natural**×5, the injections of sea-salt aerosols were also dependent on the wind speed but the integrated amounts in the region <u>wereare</u> set to be equal to the case that all area had a fixed rate of  $10^{-9}$  kg m<sup>-2</sup> s<sup>-1</sup> (Fixed).

224 2.3 Calculations

The calculation method related to radiation, cloud properties, and cloud radiation forcing was based on Goddard et al. (2022), briefly described here as follows. This study focused on the shortwave radiative flux responses at the TOA due to the injections of sea salt aerosols, which was consistent with the definition of effective radiation forcing (ERF) (Forster et al., 2007). The sea surface temperature in the · 设置了格式:字体: Times New Roman,字体颜色: 红色

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229	model was preset by NCEP-FNL, so the model's surface temperature and upward longwave radiation
230	would not respond to the increased sea-salt aerosols. The total upward shortwave radiation flux (SW_TOT)
231	at the TOA was under the all-sky conditions. The responses of SW_TOT to the injections of sea-salt
232	aerosols could be divided into the cloud radiation effects (SW_CLD, excluding the influence of aerosols)
233	and the direct scattering effects when clouds were present (SW_AER).
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236	flux responses at the TOA due to the injections of sea-salt aerosols, which is consistent with the definition
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238	preset by NCEP-FNL, so the model's surface temperature and upward longwave radiation would not
239	respond to the increased sea-salt aerosols. The total upward shortwave radiation flux (SW_TOT) at the
240	TOA is under the all-sky conditions. The responses of SW_TOT to the injections of sea-salt aerosols
241	could be divided into the cloud radiation effects (SW_CLD, excluding the direct effect of the aerosols)
242	and direct scattering effects when clouds are present (SW_AER)
243	$SW_TOT = SW_CLD + SW_AER$ (4)
244	The diagnosis of CLEAN-SKY (no aerosols) wasis not considered in the previous WRF-CMAQ
245	model. So in this study, we extended extend this feature in the WRF-CMAQ model using the methodology
246	of Ghan et al. (2012)Ghan et al. (2012) by performing a double radiative call at each time step to calculate
246 247	
247	of Ghan et al. (2012)Ghan et al. (2012) by performing a double radiative call at each time step to calculate
247 248	of Ghan et al. (2012)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 <u>have</u> also <u>studiedstudy</u> the impacts of
247 248 249	of Ghan et al. (2012)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-have also studiedstudy the impacts of injecting sea-salt aerosols on the upward shortwave radiation flux at the TOA under the clear-sky
247 248 249 250	of Ghan et al. (2012)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-have also studiedstudy 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
247 248 249 250 251	of Ghan et al. (2012)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-have also studiedstudy 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 wasjs considered to be the maximum MSB potential generated by injecting sea-salt aerosols when there
247 248 249 250 251 252	of Ghan et al. (2012)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-have also studiedstudy 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 wasis considered to be the maximum MSB potential generated by injecting sea-salt aerosols when there wereis no clouds. Unless otherwise specified, all results in this study were monthly averages, cloud.
247 248 249 250 251 252 253	of Ghan et al. (2012)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-have also studiedstudy 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 wasjs considered to be the maximum MSB potential generated by injecting sea-salt aerosols when there wereis no clouds. Unless otherwise specified, all results in this study were monthly averages.cloud. Due to the different amounts of sea-salt aerosols injected in different ways, it resultedresults in-
247 248 249 250 251	of Ghan et al. (2012)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 <u>have</u> also <u>studiedstudy</u> 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 <u>wasis</u> considered to be the maximum MSB potential generated by injecting sea-salt aerosols when there <u>wereis</u> no <u>clouds</u> . Unless otherwise specified, all results in this study were monthly averages. <u>cloud</u> . Due to the different amounts of sea-salt aerosols injected in different ways, it <u>resultedresults</u> interfore, we <u>proposedpropose</u> the concept of MCB efficiency (E <sub>MCB</sub> ) to
247 248 249 250 251 252 253 254	of Ghan et al. (2012)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-have also studiedstudy 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 wasis considered to be the maximum MSB potential generated by injecting sea-salt aerosols when there wereis no clouds. Unless otherwise specified, all results in this study were monthly averages, cloud. Due to the different amounts of sea-salt aerosols injected in different ways, it resultedresults in the different SW_TOT responses. Therefore, we proposed propose the concept of MCB efficiency (E <sub>MCB</sub> ) to measure the relationships between the amount of sea-salt aerosol injections and the resulting radiation
247 248 249 250 251 252 253 254 255	of Ghan et al. (2012) 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-have also studiedstudy 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 wasis considered to be the maximum MSB potential generated by injecting sea-salt aerosols when there wereis no clouds. Unless otherwise specified, all results in this study were monthly averages.cloud. Due to the different amounts of sea-salt aerosols injected in different ways, it resultedresults in different SW_TOT responses. Therefore, we proposed propose the concept of MCB efficiency (E <sub>MCB</sub> ) to measure the relationships between the amount of sea-salt aerosol injections and the resulting radiation flux responses: <u>(Table S2)</u> .

at a rate of 1 kg m<sup>-2</sup> s<sup>-1</sup>.  $E_{MCB} = 1$  means that injecting 1 kg of sea-salt aerosols per unit time in the current study area wasis expected to produce a 1 GW (10<sup>9</sup> W) SW\_TOT response. Note that this value ( $E_{MCB}$ ) wasis based on model calculations under specific atmospheric conditions within the study region and wasis only used to analyze the sensitivities of the radiative flux to different injection methods and injection amounts.

This study <u>focused focuses</u> on the changes in liquid clouds and evaluated the responses in cloud condensation nuclei (CCN), cloud fraction, CDNC,  $r_e$ , <u>liquid water path (LWP)</u><sub>52</sub> cloud optical thickness (COT), and cloud albedo due to the injections of sea-salt aerosols. These calculations <u>wereare</u> shown in Supplementary Text S1.

268 Cloud radiation forcing (CRF) parameters can be used to quantify the responses of SW\_CLD to 269 changes in cloud cover or cloud albedo, defined as follows (Goddard et al., 2022):

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

where  $\alpha_c \frac{\text{wasis}}{\text{max}}$  mean cloud albedo and  $f \frac{\text{wasis}}{\text{wasis}}$  mean cloud fraction.

272 The CRF parameters <u>couldcan</u> be approximated using the perturbation method as follows (Goddard
273 et al., 2022):

274

270

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

where the first term on the right-hand side indicated indicates the changes in  $CRF_{param}$  driven by the perturbation of cloud albedo, the second term indicated indicates the changes driven by the perturbation of cloud fraction, and the third term denoted denotes the changes driven by the interactions of the two. The horizontal bars on  $\alpha_c$  and *f* wereare defined as the monthly means of the Base, and the prime (') defined defines the monthly mean differences between the sensitivity experiments and Base. The fourth column of Fig. <u>\$24\$23</u> shows that the differences between  $CRF_{param}$  and  $CRF'_{param}$  wereare small enough that the perturbation method could can be used to approximate the  $CRF'_{param}$ .

The changes in cloud albedo were 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:

286	$\frac{\Delta \alpha}{\Delta m} = f \Delta \alpha (1 - \alpha) \left( \frac{1 \Delta \ln \text{CDNG}}{1 + \Delta \ln \text{CLWP}} + \frac{\Delta \ln f}{2} \right) $
200	$\frac{\Delta \ln AOD}{\Delta \ln AOD} = \frac{\Delta \ln AOD}{\Delta \ln AOD} = \frac{\Delta \ln AOD}{\Delta \ln AOD} = \frac{\Delta \ln AOD}{\Delta \ln AOD}$
287	The changes in cloud albedo are driven by multiple processes. Based on Quaas et al. (2008) and
288	Christensen et al. (2020), Goddard et al. (2022) established the following equation to assess the relative

289 effects of CDNC, LWP, and mean cloud fraction on the responses of SW CLD due to the injections of 290 sea-salt aerosols:  $\frac{\Delta \alpha}{\Delta \ln \text{AOD}} = f \Delta \alpha_c (1 - \alpha_c) \left( \frac{1}{3} \frac{\Delta \ln \text{CDNC}}{\Delta \ln \text{AOD}} + \frac{5}{6} \frac{\Delta \ln \text{CLWP}}{\Delta \ln \text{AOD}} + \frac{\Delta \ln f}{\Delta \ln \text{AOD}} \right)$ 291 (8) 292 where  $\alpha$  wasis the planetary albedo,  $\Delta$  represented represents the difference in monthly average results 293 between sensitivity experiments and Base simulations, and  $\alpha_c$  wasis the cloud albedo. The three terms inside the right parenthesis represented represent the relative contributions of Twomey effect, LWP effect, 294 and cloud fraction effect, respectively, with the latter two related to the second aerosol indirect effect 295 (Albrecht, 1989). 296

The perturbations by generating three ensemble members for each experiment in each region were 297 added. The results of all sensitivity experiments are compared to those of Base simulations. Unless 298 otherwise specified, all results in this study are shown as overall regional monthly averages of the 299 800 ensemble.

801

#### 302 3. Results

#### 303 3.1 The impacts of different injection strategies on shortwave radiation at the TOA.

304 In modeling studies, using different variations in methods used to increase sea-salt aerosols may lead to 805 different conclusions, and this discrepancy 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 806 807 different strategies (with dry diameters of about 0.11-0.15 µm, Fig. 1a) increased increase the SW TOT at the TOA by 0.06-24.5007-25 W m<sup>-2</sup> in the five ocean regions (Fig. 23a). The Natural×5 and Wind-808 809 adjusted methodsstrategies, which reliesrely on wind speeds, injectedinject sea-salt aerosols of 0.03031-0.09085 and 0.18-0.21 × 10<sup>-9</sup> kg m<sup>-2</sup> s<sup>-1</sup> into the five regions, respectively, resulting and result in SW TOT 810 811 variations of 0.0607-2.08 and 1.35 and 1.4-8.474 W m<sup>-2</sup> (Table S2), respectively. Except for the Equa region, the other four regions can initially achieve the radiation flux responses required to offset the 812 radiative forcing of 3.7 W m<sup>2</sup> due to doubling of atmospheric CO<sub>2</sub> concentration since industrialization 813 814 (Latham et al., 2008; Ramaswamy et al., 2001). (Fig. 3a and Table 2). Uniformly injections of sea-salt 815 aerosols at a fixed rate of 10<sup>-9</sup> kg m<sup>-2</sup> s<sup>-1</sup> resultedresults in SW\_TOT changes of 10.96 24.5011-25 W m<sup>-</sup> 816 <sup>2</sup> in the five regions. This value far exceeded the radiation flux response envisioned for offsetting 817 geoengineering, so we only used it to explore the sensitivities of different injection methods and regional responses, and subsequent studies on the impacts of injecting sea salt aerosols in sensitive areas on the 818 13

entire region. The three continental west coast stratocumulus regions of NP, SP, and SA hadhave the most
 significant SW\_TOT responses, all exceeding 20 W m<sup>-2</sup>, while the SW\_TOT responses in the AsiaWP
 and Equa regions are 18 and Equa regions were 17.34 and 10.9611 W m<sup>-2</sup>, respectively,

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822 Injecting the same amount of sea-salt aerosols resultedresults in substantial variations in SW TOT 823 responses across the different regions (Fig. S1S2). The sea-salt aerosols sprayed in the Fixed-wind-824 adjusted experiments wereare also dependent on wind speed, but the amount of emission rate integrated 325 in the full domain wasis consistent with the fixed rate of  $10^{-9}$  kg m<sup>-2</sup> s<sup>-1</sup>, ruling out the differences caused by the amount of injected sea-salt aerosols. Although both methods injectedstrategies inject the same 826 amounts of sea-salt aerosols, the SW TOT responses they produced wereproduce are significantly 827 different. The Fixed-wind-adjusted method resultedstrategy results in SW TOT changes of 5.00-19.780-828 20 W m<sup>-2</sup> in the five regions, (Fig. 3a), indicating that the shortwave radiation flux changes caused by 829 wind-speed-dependent injections wereare smaller than those caused by uniformly injections, and showed 830 831 regional differences. Due to the different amounts of sea-salt acrosol injected in different ways, it resulted 832 in different SW TOT responses. Therefore, we proposed the concept of MCB efficiency (E<sub>MCB</sub>) to measure the relationships between the amounts of sea-salt aerosol injections and the resulting radiation 833 834 flux responses.  $E_{MCB} = 1$  GW kg<sup>-1</sup> s means that injecting 1 kg of sea salt aerosols per unit time in the current study area was expected to produce a 1 GW (10<sup>9</sup> W) SW TOT response. 835

Figure 33b shows the EMCB values of different sea-salt injection strategies in the five regions. Overall, 836 MCB implementation wasis more efficient in the NP, SP, and SA regions, while it wasis less efficient in 837 the AsiaWP and Equa, which wasis similar to the previous SW TOT response results. E<sub>MCB</sub> also 838 839 variedvaries for different injection methodsstrategies. In the NP, SP, and SA regions, the E<sub>MCB</sub> values of 840 the Natural×5 and Wind-adjusted methodsstrategies with relatively small injection amounts were are 841 higher. With the increases in sea salt aerosols injections, EMCB decreased (discussed below), than the other two strategies with large injection amounts. At the same injection amount, injecting at a fixed rate shows 842 higher EMCB compared relative to injections dependent depending on wind speed, as consistently shown in 843 all five regions- (Fig. 3b). Since the number flux of aerosols increased with the decreases of the injected 844 845 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 846 injection rate of aerosol number (Fig. S3c). In the same quality injected, the aerosol number varies greatly 847 848 (Fig. S3d).

## The productions of sea-salt aerosols in nature wereare strongly correlated with wind speed, and most

models associated sea-salt aerosol emissions with wind speed (Ahlm et al., 2017; Grythe et al., 2014). 350 851 Injection strategies depending on wind speed mademake the distributions of added sea-salt aerosols closer to the natural distributions. In natural environments, sea-salt aerosol emissions in strong-wind areas (e.g., 352 353 storm or typhoon areas) and surf zones are usually much larger than in weak-wind areas. Therefore, 854 injection methodsstrategies depending on wind speed concentrated concentrate the added sea-salt aerosols 355 in strong-wind areas and surf zones, while the weak-wind regions increasedincrease relatively little sea-856 salt aerosols (Fig. <u>\$254</u>). Injecting uniformly at a fixed rate in the model wouldwill result in a large 357 increase of sea-salt aerosols in places with 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 358 859 rate, it wasis necessary to consider the impacts of different injection methods on the distributions of sea-360 salt emissions. Using a uniformly increasing method independent of wind speed can not only avoid the 361 situation of a smaller increase in sea-salt emissions in regions with lower wind speeds, but can also 362 identify the geographical areas most sensitive to the increased sea-salt aerosols and producing the largest 363 TOA radiation perturbations (Alterskjær et al., 2012).

Uniform injections of sea-salt aerosols throughout the region ignored the transport and diffusion of 864 865 aerosols. Therefore, we captured the geographical areas with the most sensitive and largest TOA radiation 366 disturbances due to uniform injections in various ocean regions (30 × 50 grids, approximately 360 km × 600 km, and away from the domain boundaries). Injecting sea-salt aerosols in the sensitive areas with the 867 same uniform injections ( $10^{-9}$  kg m<sup>-2</sup> s<sup>-1</sup>, the injection rate is about 1/20 of the full domain injection) 368 resultedresults in changes of 0.6549 - 3.274 W m<sup>-2</sup> in SW TOT in the five ocean regions (Table S2). The 869 870 SW TOT responses wereare the largest in the SP region, at 3.274 W m<sup>-2</sup>, and 2.697 and 1.817 W m<sup>-2</sup> in the NP and SA regions, respectively, while they were only 0.6549 and 0.7483 W m<sup>-2</sup> in the AsiaWP and 871 872 Equa regions, respectively. The injected sea-salt aerosols produced SW TOT changes of 5.11-14-15.22.3 W m<sup>-2</sup> in the sensitive areas (Fig. 1b). Similarly, the increases in SW TOT in the SP, SA, and NP regions 373 874 all exceeded 109 W m<sup>-2</sup>, with the highest in the SP region at 15.2214.3 W m<sup>-2</sup>. In the Asia WP and Equa 875 regions, although the increases in SW TOT wereare only 6.225.11 and 5.1426 W m<sup>-2</sup>, respectively, it can still achieve the goal of offsetting the overall effective radiative foreing produced by anthropogenic 876 activities in the sensitive areas. Also, when injecting at a uniform rate of 10-9 kg m<sup>-2</sup> s<sup>-1</sup> within the sensitive 877 areas, for the entire region, the MCB efficiencies were greatly improved for all sea areas except Asia (Fig. 878 879 S3). Considering that the original intents of MCB or MSB design wereare regional application (hurricane mitigation, coral reef protection and polar sea ice recovery); (Latham et al., 2014), choosing to inject sea-380

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.

## 383 **3.2 Characterization of the radiation responses.**

384 SW TOT responses wereare defined as the sum of the upward shortwave radiation flux response at the 385 TOA 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 386 and SW\_CLD responses in the SW\_TOT produced by different injection methodsstrategies in the five 887 888 ocean regions. The majority of the SW TOT radiative flux response due to the less injected lower mass injection Natural×5 and Wind-adjusted strategies wasis caused by the SW CLD response (Fig. 4 and 889 390 Table 14a). In the NP, SP, and SA regions, the contributioncontributions of SW\_CLD exceededexceed 70%, suggesting that sea-salt aerosols injected at these locations increased increase the SW TOT mainly 891 392 by affecting clouds through indirect effects. In the Equa, the responses of SW TOT wereare entirely caused by SW AER. The proportion of SW AER produced by the uniform injection of sea-salt aerosols 393 at a fixed rate of 10<sup>-9</sup> kg m<sup>-2</sup> s<sup>-1</sup> continued to increase (Fig. 4c). In the Asia WP, Equa, and SP regions, the 894 proportion of SW AER exceeded that of SW CLD. In the SA region, SW CLD and SW AER wereare 895 896 almost equal, while in the NP region, the SW CLD response wasis 13.41 W m<sup>-2</sup>, still greater than SW AER (9.708 W m<sup>-2</sup>). This is because there wasis a saturation phenomenon in the cloud response to 897 aerosols injections (discussed below), and the NP, SP, and SA regions providedprovide more SW CLD 898 responses, while the cloud responses in the Asia WP and Equa regions saturated saturate and no longer 899 400 increasedincrease. The results of Fixed-wind-adjusted case show that, at the same injection amount, the 401 SW AER responses caused by the injection methodstrategy relying on wind speed wasis significantly smaller than those of the method with fixed-rate uniform injection, while the disparity in SW CLD 402 403 responses wasis minimal. This is because the injection methodstrategy relying on wind speed distributed 404 most of the increased sea-salt aerosols to areas with already high emissions, such as strong-wind areas 405 and surf zones, where the excess marine aerosols hadhave already saturated the cloud responses, resulting 406 in minor changes in SW CLD. In areas with weak winds, the potentials for direct aerosol scattering 407 wereare not fully exploited due to the relatively small amounts of sea-salt aerosols injected, leading to a lower SW AER response. 408

Figures <u>S4S5</u> and <u>S5S6</u> 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 <u>wereare</u> stronger in

411 the three regions of NP, SP, and SA, while they wereare weaker in the regions of AsiaWP and Equa, and 412 in some grids they even led to a reduction of the upward shortwave radiation- (Fig. S5). The spatial 413 distributions of the SW CLD responses exhibitedexhibit noticeable discontinuity, reflecting significant 414 regional differences in the non-uniform distributions of clouds and their impacts on shortwave radiation 415 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 416 417 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. 418 419 In contrast, the spatial distributions of the SW AER response wereare smoother, leading to consistent 420 increases in upward shortwave radiation at the TOA in all ocean regions- (Fig. S6). This indicates smaller 421 spatial limitations in the distributions of aerosol particles, allowing direct scattering effects to take place 422 everywhere. The direct scattering effect of aerosols wasis primarily related to the concentrations and 423 physical properties of the particles (discussed below), unlike clouds, which wereare influenced by 424 multiple variables. These results suggest that when implementing geoengineering measures, it is essential 425 to comprehensively consider the interactions between aerosols and clouds, as well as their different 426 response patterns in various regions. Furthermore, the high spatial variabilities of cloud radiation effects 427 emphasizedemphasize the need for improved resolution in future model studies of cloud-aerosol interactions. 428

The SW CLD response resulting from the injection of sea-salt aerosols in the sensitive areas of five 429 ocean regions exhibits significant spatial differences. The SW CLD response is larger than the SW AER 430 431 response in the sensitive areas of NP, SP, and SA, indicating that the changes in SW TOT are mainly driven by the cloud radiation response (Fig. 5). In contrast, the SW\_CLD response is smaller in the 432 433 AsiaWP and Equa regions. This regional difference is similar to that observed with uniform injection 434 across the entire region. The SW AER response shows consistent results in all areas, resulting in a radiation response change of 3.5558-5.4244 W m<sup>-2</sup> within the injection areas. In the Asia WP and Equa, 435 436 the variations in SW TOT were 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 to three times the total radiation within the sensitive areas (Fig. 6).
In all regions except <u>AsiaWP</u>, the total SW\_CLD response outside the sensitive region was about 270%–
408% higher than inside. In <u>AsiaWP</u>, the SW\_CLD response outside the sensitive area was only 29% of
the response inside.<u>has a negative effect</u>. The SW\_CLD responses in NP, SP, and SA <u>extendedextend</u> to

442 the west and northwest of the injection areasby the prevailing winds, indicating that clouds in these areas 443 wereare affected by the injection of sea-salt aerosols (Fig. 5). Changes in cloud microphysical properties wouldwill be presented later. The SW CLD variations in other directions wereare not uniform, and there 444 wasis negative SW CLD responses in some grids, which again reflected the spatial complexities of cloud 445 446 radiation effects. The direct scattering effects of aerosols on areas outside the sensitive region is reflected 447 in a widespread increase in upward shortwave radiation at the TOA. The total SW AER responses outside 448 the sensitive areas in the five ocean regions wereare approximately 160%-281% higher than inside, but lower than the impacts of SW\_CLD responses outside the sensitive areas. There wereare consistencies in 449 the spatial distributions of SW AER and SW CLD responses. 450

## 451 **3.3 Saturation of the cloud radiative responses.**

452 Figure 7 shows that under low levels of sea-salt aerosol injections, radiation response changes wereare 453 mainly driven by SW CLD responses. As the injected sea-salt aerosols increased, the SW CLD responses 454 gradually reachedreach saturation. After reaching a certain injection level, the increases of SW CLD 455 responses stabilized stabilized at its maximum value and no longer increases with further injections. The 456 SW CLD responses show large differences in the five ocean regions, and the different shapes and slopes 457 of the curves indicated indicate that the cloud radiative forcing responses to the sea-salt aerosol injections wereare different in each region. This might be due to variations in cloud types, cloud amounts, and 458 atmospheric conditions in the different regions. In the NP, SP, and SA, the SW CLD responses 459 460 exceededexceed 10 W m<sup>-2</sup>, while in Asia WP, it saturated saturates at 5 W m<sup>-2</sup>. In Equa, when the sea-salt aerosol injection rate wasis 10-9 kg m<sup>-2</sup> s<sup>-1</sup>, the SW\_CLD response wasis 0.545 W m<sup>-2</sup>, and even when the 461 injection doubled, the SW CLD response remained at 0.545 W m<sup>-2</sup>. This implies that the SW TOT at 462 Equa was almost exclusively from the contributions of the direct scattering effects of aerosols. 463

464 In contrast to SW CLD, the SW AER responses increased increase linearly with the injections of 465 sea-salt aerosols ( $R^2 > 0.99$ ). As the injection increased increases, the contributions of SW\_AER to SW TOT gradually increasedincrease, surpassing the SW CLD responses, and showedshow the same 466 467 trends across the five regions. This implies that at higher injection levels, the contributions of SW CLD 468 to total radiation change saturated, and cloud properties no longer significantly changed change. At this 469 point, sea-salt aerosols primarily affected affect radiation through direct scattering effects, and the aerosol 470 particles' ability to scatter solar radiation continued to increase with the increases in aerosol quantities. In 471 some cloud-free regions or weather conditions, injected sea-salt aerosols wereare still able to function

472 through direct scattering.

473 There existed exists a specific injection level at which the SW CLD and SW AER responses were are 474 equal. In the NP region, when the injection level wasis approximately 1.55×10<sup>-9</sup> kg m<sup>-2</sup> s<sup>-1</sup>, both SW CLD and SW AER responses wereare 15 W m<sup>-2</sup>. In the SP and SA, these levels wereare about 0.67×10<sup>-9</sup> kg m<sup>-2</sup> 475  $^{2}$  s<sup>-1</sup> and 1×10<sup>-9</sup> kg m<sup>-2</sup> s<sup>-1</sup>, respectively. While in Asia<u>WP</u>, the responses were already equal when the 476 477 injection amount was 0.15×10<sup>-9</sup> kg m<sup>-2</sup> s<sup>-1</sup>. Since there wasis a saturation of the cloud radiation effects, 478 E<sub>MCB</sub> decreaseddecreases with the increases in sea-salt aerosol injection amounts (Fig. 7, red dashed line). 479 This can also explain the higher EMCB of the Natural×5 and Wind-adjusted methodsstrategies with relatively low injection amounts (Fig. 3).-3b). Therefore, wind-dependent injection strategies led to the 480 injection of large amounts of sea-salt aerosols in certain areas with high wind speeds, leading to saturation 481 of cloud radiation effects, which might affect the performances of MCB in the simulations of regional 482 483 and global models.

When less sea-salt aerosols wereare injected, both SW\_CLD and SW\_AER responses contributed<u>contribute</u> to the changes of SW\_TOT. As the injection amounts increased<u>increase</u>, the SW\_CLD responses saturated<u>saturate</u>, and the increases in SW\_TOT depended on the increases in SW\_AER responses, leading to a decrease in  $E_{MCB_{\tau}}$  (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.

Under clear and cloudless conditions, injecting sea-salt aerosols could still increase the SW\_TOT 490 through direct scattering, and this effect exceeded exceeds those of aerosol direct scattering when clouds 491 492 wereare present. The variations variation of the upward shortwave radiation flux at the TOA under the 493 clear-sky conditions (SW\_AER\_CLR) diddoes not exhibit significant regional heterogeneity across the 494 ocean areas (Figs. 5 and <u>\$6\$7</u>), suggesting that the contribution of direct aerosol scattering wasis more 495 uniform globally when considering the effects of sea-salt injections on the Earth's radiation budget. The SW AER CLR responses were are also linearly correlated with the injection of sea-salt aerosols ( $R^2 >$ 496 0.99), and it exceededexceeds the SW AER responses (Fig. 7). This is because cloud layers also 497 498 seatteredscatter and absorbedabsorb solar radiation, so this scattering effect wasis more significant under 499 clear sky conditions. It wasis reflected that in regions with strong cloud radiation effects, such as the NP, SP, and SA regions, the differences between the SW\_AER and SW\_AER\_CLR responses wereare also 500 larger (Fig .7). When injecting sea-salt aerosol in sensitive areas, the spatial distributions of 501 SW\_AER\_CLR and SW\_AER responses wereare highly consistent (Fig. 5). Therefore, injecting sea-salt 502

aerosol under conditions of low cloud covers or clear skies also increased increases the upward shortwave
 radiation flux at the TOA.

## 505 **3.4 Factors affecting the radiation effects.**

506 The direct radiative effect of aerosols is mainly determined by their own optical properties. In WRF-507 CMAQ, the emitted sea-salt acrosol particle size distributions were adjusted to the local relative humidity (Kelly et al., 2010; Zhang et al., 2005). The dry diameter of sea salt aerosols injected into the five regions 508 509 was about 0.11 0.15 um (Figs. 1a and S7), and the wet diameter was about 0.22 0.3 um (Fig. S8). The 510 single scattering albedo (SSA) of aerosols describes the ratio of aerosol particles' ability to absorb and scatter solar radiations. After the injection of sea salt aerosols, the SSA of the accumulation mode aerosols 511 512 in the five regions generally increased by about 0.003 0.005, and in some regions within the area, the SSA increased by over 0.007, with an average increase of 0.001 0.003 in sensitive areas (Fig. S9). This 513 514 indicates that the injected sea salt acrosol particles could seatter sunlight more effectively than absorb it, causing solar radiation to be reflected back into space. The asymmetry factor of aerosols is a parameter 515 516 describing the directionality of aerosol particle scattering of sunlight, and an important factor for 517 evaluating direct aerosol radiative forcing (Zhao et al., 2018). The injection of sea-salt aerosols in the five 518 regions reduced the asymmetry factor by 0.007 0.029, with an average reduction of 0.01 0.027 in 519 sensitive areas (Fig. S10). This indicates that the injected sea-salt aerosols tended to seatter more 520 uniformly or backward rather than in a forward direction. Uniform injections of 10<sup>-9</sup> kg m<sup>-2</sup> s<sup>-1</sup> sea-salt aerosols led to an increase in aerosol optical depth (AOD) of 521 0.220-0.37 in all regions (Fig. 8). The distributions of AOD within the regions wereare not uniform due 522 523 to aerosol transports and diffusions, with some areas showing an increase in AOD of over 0.6. Injecting 524 sea-salt aerosols in sensitive areas ledlead to an AOD increase of 0.08077-0.12, while outside the 525 injection areas, AOD gradually decreased decreases as the aerosols were transported transport and 526 disperseddisperse. With the increases in sea-salt aerosol injections, AOD showed shows a linear increase 527 within a certain range in all five ocean regions ( $R^2 > 0.997$ , Fig. 9a). There wasis a strong correlation 528 between the AOD changes caused by sea-salt injection and the SW AER responses. When sea-salt 529 aerosols wereare uniformly injected across the entire region, the correlation coefficients between AOD 530 and SW AER responses in the five ocean areas wereare greater than 0.94, and when injected in sensitive 531 areas, the correlation coefficients wereare greater than 0.99 (Fig. S8), S11). There was also a strong spatial 532 consistency between The optical properties of injected aerosols are described in Supplementary Text S2.

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In general, the spatial distribution of AOD injected sea-salt aerosols scatter sunlight more efficiently than absorb it, causing solar radiation to be reflected back into space and SW\_AER response (Fig. S5, third row, and Fig. 8, second column; Fig. 5, second column, and Fig. 8, third column).tend to scatter more uniformly or backward rather than forward.

537 There were significant differences in the distributions of clouds in the five ocean regions during the study period, with wide distributions of liquid clouds in the NP, SP and SA regions, and less clouds in 538 539 Asia and Equa. The cloud heights were distributed between 500 2000 meters, centered around 1000 540 meters (Figs. S12 and S14). In the regions with higher cloud cover, such as NP, SP, and SA, injected seasalt aerosols significantly increased increases cloud fraction, (Fig. 2, third column and Table 1), leading to 541 542 the formations of more clouds or expanding the coverage, vertical thickness and lifetime of existing 543 clouds (Goddard et al., 2022). The injection of sea-salt aerosols in sensitive areas hadhave similar results, 544 where cloud fractions increased increase both inside the injection areas and in the regions affected by 545 aerosol transports and diffusions (Fig. S13).

546 The injected sea salt aerosols affected the cloud microphysical properties through indirect effects, thereby influencing cloud radiative responses. Taking the SP region as an example, Fig. 10 demonstrates 547 548 that uniformly injections of 10<sup>-9</sup> kg m<sup>-2</sup> s<sup>-1</sup> sea-salt aerosols significantly increased increases the CDNC. 549 More cloud droplets eaptured capture more water vapor, leading to an increase in liquid water path (LWP). Additionally, the increases in cloud thickness also contributed contribute to the increase in LWP. The 550 551 increases increase in CDNC decreased decreases the mean  $r_e$  by 8.9  $\mu$ m (~ -37%), increased increases the eloud optical thickness (COT) by more than 220%, and ultimately increased increases the mean cloud 552 553 albedo over the region by 0.19 (~64%). Similarly, injecting sea-salt aerosols in the NP and SA regions led 554 to average cloud albedo increases of 0.17 and 0.20, respectively, while in the AsiaWP and Equa, the 555 increases wereare 0.15 and 0.13, respectively (Figs. S15 S18S14-S17). The injectionsinjection of sea-556 salt aerosols within the sensitive areas hadhas less effect on cloud microphysical properties than the whole region injections. This is because when sea-salt aerosols were are injected across the entire region, the 557 surrounding sea-salt aerosols affected affect the sensitive areas through transports, resulting in an 558 enhanced cumulative effect on cloud microphysical properties in the sensitive areas. Injecting sea-salt 559 aerosol in the sensitive area of the SP affected clouds in the surrounding region through transports, 560 561 increasedincreases the average cloud albedo across the entire area by 0.03032 over the entire region and by 0.12 within the sensitive regions, which wasis less than the effects of injection across the entire area 562 (Fig. S19S18). Similarly, injecting sea-salt aerosols in the sensitive areas of other seaocean regions ledlead 563

to average cloud albedo increases of 0.01015-0.02024 across the entire area, with increases of 0.11 in the sensitive areas of the SP and SA regions, and increases of 0.09090 and 0.10 in the AsiaWP and Equa, respectively (Figs.  $\frac{820-823819-822}{2}$ ).

#### 567 **3.5 Drivers of SW\_CLD responses.**

568 The cloud radiation forcing (CRF) parameters wereare used to calculate the effects of changes in cloud cover and cloud albedo on the SW CLD responses due to the injections of sea-salt aerosols. Figure 569 570 \$24\$23 illustrates the increase in the CRF parameter coinciding with the increases in the SW CLD 571 responses after uniform injection of sea-salt aerosols in the five regions (Fig. \$485, third row). The results 572 wereare similar for injections in the sensitive areas (Fig. 525S24, third column, and Fig. 5, first column). The CRF' param calculated using the perturbation method indicates that in the five ocean regions, CRF' param 573 wasis primarily driven by perturbations in cloud albedo (Fig. <u>\$26\$25</u>, first column), and it significantly 574 575 surpassedsurpasses the changes in cloud fractions and their interactions. Cloud albedo changes explainedexplain over 70% of the CRF'param in all fourfive regions except the Equa. The 576 577 contributionscontribution of cloud fraction changes rangedranges from 13.9% to 23.87%, while the interactions between the two factors accounted account for only about 10% (Fig. S26S25, second and third 578 579 columns). The results wereare similar for injections in sensitive regions, where changes in cloud albedo accounted for 68.9% 79.658.8% 99.4% of the CRF'param, followed by changes in cloud fractions, with 580 581 the smallest contributions from their interactions (Fig. <u>\$27\$26</u>).

582 Figure 11 evaluates the relative effects of Twomey, LWP, and cloud fractions on the SW CLD responses after uniformly injecting sea-salt aerosols in five ocean regions. The results indicate that 583 584 changes in CDNC (Twomey effect) and LWP wereare the main drivers of SW CLD responses, while changes in cloud fraction contributedcontribute minimally to the SW CLD responses. Except for the 585 586 Equa region, changes in CDNC and LWP accounted for 48.24%-52.45% and 38.939.0%-41.97% of the SW CLD changes, respectively, with cloud fraction changes contributing to less than 10%...0% (Fig. 11). 587 588 The results wereare similar for injections in sensitive areas, with changes in CDNC and LWP contributing 589 similarly and more than changes in cloud fractions to SW CLD (Fig. S28S27). The changes in SW CLD 590 responses after aerosol injections in the sensitive areas of Equa wereare mainly contributed by LWP effects (~70%). 591

592 Uniform injections of sea-salt aerosols at a rate of  $10^{-9}$  kg m<sup>-2</sup> s<sup>-1</sup> produced susceptibilities ( $\frac{\Delta \alpha}{\Delta \ln AOD}$ ) 593 ranging from 0.000300030 to 0.0035 in the five regions, with corresponding spatial distributions shown 22 594 in Fig. 11. NP, SP, and SA regions exhibitedexhibit cloud responses that wereare more sensitive to aerosol injections in most of the region, with susceptibilities ranging from 0.0028 to 0.0035. The Equa 595 showedshows the lowest susceptibility, indicating that the system wasis less responsive to variations in 596 597 aerosol injections. It is noteworthy that although the average susceptibility in the AsiaWP region wasis 598 0.0013, the higher susceptibility values wereare concentrated in the north of 35°N, where the average 599 susceptibility wasis 0.0026, similar to those of the SP region, suggesting that clouds here wereare more 600 susceptible to aerosol injections. Injecting sea-salt aerosols in sensitive areas mostly resultedresults in cloud that wereare located outside the sensitive areas (Fig. <u>\$28\$27</u>). Injecting sea-salt aerosols in the 601 sensitive areas of SP and SA hadhave a greater impact on the northwest. In the sensitive areas of NP, 602 603 injecting sea-salt aerosols hadhave a larger impact on the west. In the AsiaWP, the injection of sea-salt 604 aerosols into the sensitive area diddoes not fully reflect its susceptibility because we ehosechoose to 605 calculate the sensitive areas away from the boundary, and the greatest susceptibilities in the AsiaWP 606 region happened happens to be in the northern part of the region near the boundary.

## 607 4. Discussions and conclusions

Many studies have discussed the contributions of both the direct and indirect effects of MCB. Some 608 studies suggest that MCB primarily relies on the indirect effects, as originally conceived, i.e., injecting 609 610 aerosols to brighten clouds (Jones and Haywood, 2012; Latham et al., 2012). On the other hand, 611 otherOther studies proposed that the direct scattering effects of aerosols may be more important (Ahlm et 612 al., 2017; Kravitz et al., 2013; Mahfouz et al., 2023; Niemeier et al., 2013; Partanen et al., 2012). Our 613 results indicate that the importance importances of both aerosol direct and indirect effects during MCB 614 implementation dependeddepend on the injection amountsstrategies and the choice of injection regions. 615 In cases of low sea-salt aerosol injections or the early stage of MCB implementations, changes in radiative 616 response wereare mainly driven by indirect effects, causing clouds to brighten easily. As the 617 injectionsinjection of sea-salt aerosol increased increases, the radiative effects effect on clouds saturatedsaturates, and the clouds wereare difficult to brighten. In contrast, the direct effect continued to 618 619 increase linearly, leading to a subsequent decrease in the efficiencies of MCB. Partanen et al. 620 (2012)Partanen et al. (2012) first considered the relative importance of aerosol direct and indirect effects in MCB and preliminarily found the saturated non-linear phenomenon of indirect effects at high CDNC, 621 as well as the linear relationships between direct effects and injection amounts. Haywood et al. 622 623 (2023)Haywood et al. (2023) also found a decrease in MCB efficiency with increasing aerosol injections.

Regions initially susceptible to modification gradually became less susceptible, and aerosol direct 624 625 radiation effects dominated, This study emphasized and quantified these findingsOther use General Circulation Model (GCM) studies also found similar results (Alterskiær and Kristjánsson, 2013; Rasch et 626 627 al., 2024; Stjern et al., 2018). This study highlights and quantifies these findings in a regional model for 628 the first time, showing the changing trends of direct and indirect effects with injection amounts in the 629 different ocean regions. The best results were Also due to the higher resolution of the regional model, this 630 study provides more detailed cloud component changes due to sea-salt aerosol injection. The best results 631 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 workedwork together through both direct and indirect 632 633 effects. However, in cloud-free or less cloudy regions, MCB implementation can achieve the goal of 634 reflecting more sunlight through the direct scattering effect of aerosols. Considering the uncertainty in 635 the model's resolution of clouds and the fact that, in reality, the cloud distributions wereare also greatly 636 influenced by the local meteorological conditions, the direct scattering effects of sea-salt aerosols on MCB 637 contributions wereare relatively certain. Therefore, in cloud-free or less cloudy regions, the direct 638 effects effect of aerosols become becomes more important.

639 In the early stages of Earth-System modeling studies, the MCB processes were often simulated by presetting CDNC = 375 or 1000 cm<sup>-3</sup> in the lower regions of the ocean (Jones et al., 2009; Latham et al., 640 2008; Rasch et al., 2009). However, many follow-up studies have suggested that injections of sea-salt 641 aerosols were difficult to produce a uniform CDNC field due to aerosol dilutions, depositions, and the 642 643 dependences of the spray rate on wind speed. The CDNC wesis highly variable spatially, and studies have 644 even reported reductions in CCN and CDNC caused by the injections of sea-salt aerosols (Alterskjær et 645 al., 2012; Korhonen et al., 2010; Pringle et al., 2012). In this study, after injecting accumulation mode 646 sea-salt aerosols at a rate of 10-9 kg m<sup>-2</sup> s<sup>-1</sup>, the average CDNC concentrations for five ocean regions 647 rangedrange from 60.2 to 103100 cm<sup>-3</sup>, and the spatial distributions wereare uneven (Fig. 10 and Figs. <del>\$15 \$18</del>\$14-\$17). Figure 9b indicates that the <del>CCN</del>CCNs in the five regions <del>increased</del>increase linearly 648  $(R^2 = 1)$  with increasing sea-salt aerosol injections, but not all of the <u>CCN was</u>CCNs are converted to 649 650 cloud droplets. After doubling the injection amounts, the regional average CDNC was 85 134 is 84.8-130 cm<sup>-3</sup>, with only some grid points exceeding 200 cm<sup>-3</sup> within the regions. This implies that injecting more 651 sea-salt aerosols at this point diddoes not result in more cloud droplets, and the conversion of CCN into 652 cloud droplets wasis less efficient, which slowedslows the CDNC growths and tendedtends to saturation 653 (Fig. 9c). Alterskjær et al. (2012)Alterskjær et al. (2012) similarly injected sea-salt aerosols at a rate of 654

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10<sup>-9</sup> kg m<sup>-2</sup> s<sup>-1</sup> and found that despite emitting sea-salt mass 70 times larger than suggested by Latham et 655 al. (2008), the average CDNC over the ocean was below their assumed value of 375 cm<sup>-3</sup>. This is mainly 656 due to increased competitive effects, decreased maximum supersaturations, inhibitions of aerosol 657 activations, and closures of SO4 nucleation, resulting in reduced effectiveness of sea salt 658 injections.Latham et al. (2008), the average CDNC over the ocean was below their assumed value of 375 659 660 cm<sup>-3</sup>. This is mainly due to increased competitive effects, decreased maximum supersaturations, 661 inhibitions of aerosol 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 662 may be overestimated in the Abdul-Razzak and Ghan activation parameterization used in many GCMs 663 relative to a parcel model. When Partanen et al. (2012) injected sea-salt aerosols in a Wind-adjusted way 664 (injection amount different from this study), they found the CDNC values of 596, 650, and 784 cm<sup>-3</sup> in 665 666 the NP, SP, and SA regions, respectively. Injecting smaller-sized sea-salt aerosols even yielded CDNC 667 values exceeding 1000 cm<sup>-3</sup>. They concluded that such high values were mainly due to the model's 668 overestimation of the sizes and solubilities of accumulated mode particles, with some non-activated 669 particles forming cloud droplets. Hill and Ming (2012) increased the concentrations of sea-salt aerosols 670 by a factor of five, resulting in an average CDNC increasing from 68 to 148 cm<sup>-3</sup> between 850–925 hPa. 671 It is noteworthy that Hill and Ming (2012) It is noteworthy that Hill and Ming (2012) increased all modes of sea-salt aerosols, while this study only injected accumulation mode sea-salt aerosols, Many studies 672 have reported that selecting the appropriate injection particle size wasis crucial for MCB (Andrejczuk et 673 al., 2014; Hoffmann and Feingold, 2021; Partanen et al., 2012), and injecting Aitken and coarse modes 674 675 may even lead to a positive forcing with CDNC decreasing (Alterskjær and Kristjánsson, 2013). However, 676 Wood (2021) 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 677 aerosols generated larger radiative flux changes compared to accumulation mode (8.4 W m<sup>-2</sup> versus 3.1 678 679 W m<sup>-2</sup>). There wereare still considerable discussions about choosing the appropriate aerosol particle sizes during the implementation of MCB, with different models and parameterization schemes providing 680 681 different recommendations. The sensitivity of MCB to particle size wasis not considered in this paper and was left for future research. 682

In this study, the injection of  $10^{-9}$  kg m<sup>-2</sup> s<sup>-1</sup> accumulation mode sea-salt aerosols increased increases cloud albedo in the five ocean regions by 0.13–0.20, with a maximum of more than 0.3. After doubling the injection amounts, the regional average cloud albedo could reach 0.45–0.55, representing a cloud

686 albedo change of 0.15-0.24 (Fig. 9d). These values achievedachieve the targeted cloud albedo change as envisioned in previous studies. Bower et al. (2006) suggested that to compensate for the warming 687 associated with doubling atmospheric CO<sub>2</sub> concentrations, a cloud albedo change of 0.16 was needed in 688 three stratocumulus cloud regions (off the west coast of Africa and North and South America, representing 689 3% of global cloud cover). Wood (2021) proposed seeding Aitken mode particles in approximately 9% of 690 the ocean to achieve a corresponding cloud albedo increase of 0.16. It was also suggested that injecting 691 sea-salt aerosols in a clean, undisturbed state would produce more brightening. Fig. 9d confirms this 692 finding, indicating that clouds are more likely to brighten in the early stages of sea-salt aerosol injection, 693 and the efficiency of cloud brightening decreases with increasing injection amounts. Kravitz et al. (2014) 694 695 achieved a maximum cloud albedo change of 0.23 by injecting CCN in the Arctic region. Goddard et al. (2022), simulating injecting accumulation mode sea-salt aerosols in the central Gulf of Mexico, achieved 696 697 a simulated cloud albedo change of approximately 0.1 in the main impact region, while switching to 698 Aitken mode injection resulted in a cloud albedo change of up to 0.35. For the global implementation of 699 MCB, global cloud albedo increases of 0.02 (Bower et al., 2006), 0.062 (Latham et al., 2008), or 0.074 700 (Lenton and Vaughan, 2009) were estimated. The change in cloud albedo is influenced by the properties 701 of injected particles and the injection strategies. Jenkins et al. (2013) proposed that the optimal injection '02 time should be in the early morning over weakly precipitating cloud regions, achieving a cloud albedo increase of 0.28. Goddard et al. (2022), simulating injecting accumulation mode sea salt aerosols in the 703 central Gulf of Mexico, and achieved a simulated cloud albedo change of approximately 0.1 in the main 04 impact region, while switching to Aitken mode injection resulted in a cloud albedo change of up to 0.35. 705 706 The contributions of the change in cloud fractions to the SW CLD responses in this study wereare 707 small, which wasis consistent with the results of Goddard et al. (2022)Goddard et al. (2022). However, 708 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). 709 710 Goddard et al. (2022)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 711 712 cloud fraction to the SW CLD response would be expected to increase in drier months (Fig. S29). Three 713 of the five ocean regions in this study-(, SA, SP, and NP)-were are much drier and more stable than the Gulf of Mexico simulated by Goddard et al. (2022). Goddard et al. (2022) (Fig. S28). Furthermore, when 714 715 we switched to conducting the experiments again in the dry months of the same year, the contribution of cloud fraction to SW\_CLD did not change much, remaining at ~10%. (Fig. S28). We believe that this 716

717 might be a difference due to the parameterization scheme or resolution of the model. Liu et al. (2020)Liu 718 et al. (2020) 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 719 720 Resolution Imaging Spectroradiometer (MODIS). The neglected sub-gridded clouds in the 12-km 721 resolution simulations might lead to an underestimation of the radiative effects of clouds (fu et al., 2014). 722 In addition, cloud fractions wereare more commonly underestimated in the model (Glotfelty et al., 2019), 723 and using an updated parameterization scheme that accounts for sub-grid condensation might improve the model's ability to resolve clouds (Zhao et al., 2023). The effects of finer resolution and more 724 parameterization schemes on aerosol-cloud interactions still need to be verified. Considering the 725 difficulties of modeling to accurately capture the effects of cloud fractions on radiation, the actual effects 726 of MCB may be underestimated. The radiative results obtained in this study may represent a lower limit 727 728 to cooling.

729 This study provided provides quantifiable data on cloud and radiation changes for the implementation 730 of MCB over the regional oceans, and an optimization scheme on the injection strategy by adjusting the injection amounts and selecting sensitive areas. It is noteworthy that different parameterization schemes, 731 732 models, and resolutions can influence results, especially the cloud feedback on the injected sea-salt 733 aerosols, 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 734 the Geoengineering Model Intercomparison Project (GeoMIP) under the same framework. In Earth-735 system model studies, there has been a rich discussion of the climate and ecological impacts of the MCB 736 737 with the same framework under the Geoengineering Model Intercomparison Project (GeoMIP) (Rasch et al., 2024), However, there is still a lack of a unified framework for mid-scale MCB research. 738 739

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## 741 Data and code availability

- T42 The computational code for cloud and radiation can be found in the code publicly available from Goddard
- 43 et al. (2022)Goddard et al. (2022). The model results are available upon request.
- 744 Supplemental information.
- 745 The supplementary information related to this article is available online.
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Areas	Cloud	Fraction		<u>NC</u> <u>m<sup>-3</sup>)</u>	<u>LV</u> ( <u>g 1</u>	<u>VP</u> <u>n<sup>-2</sup>)</u>	<u>Regional sea</u> (µg	
	Base	<u>Exp</u>	Base	Exp	Base	Exp	Base	Exp
WP	<u>0.0445</u>	<u>0.0488</u>	<u>19.3</u>	<u>100</u>	<u>12.8</u>	<u>19.8</u>	<u>8.91</u>	<u>143</u>
<u>NP</u>	<u>0.0678</u>	<u>0.0760</u>	<u>9.67</u>	<u>60.2</u>	<u>24.6</u>	<u>43.9</u>	<u>7.18</u>	<u>126</u>
<u>Equa</u>	<u>0.0051</u>	<u>0.0059</u>	<u>17.5</u>	<u>83.4</u>	<u>0.85</u>	<u>1.39</u>	<u>7.32</u>	<u>102</u>
<u>SP</u>	0.0547	<u>0.0617</u>	<u>11.5</u>	<u>89.4</u>	21.6	<u>38.9</u>	<u>6.79</u>	<u>176</u>
<u>SA</u>	<u>0.0519</u>	<u>0.0575</u>	<u>12.3</u>	<u>92.2</u>	<u>23.5</u>	<u>41.6</u>	<u>7.00</u>	<u>149</u>

**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<sup>-2</sup> s<sup>-1</sup> (Exp) for five ocean regions.

	<u> </u>	<u> </u>		<u> </u>	_ <u>_</u>
Strategies	Areas	SW_TOT (W m <sup>-2</sup> )	SW_CLD (W m <sup>-2</sup> )	SW_AER (W m <sup>-2</sup> )	SW_AER_CLR (W m <sup>-2</sup> )
	Asia <u>WP</u>	0. <u><del>60</del>46</u>	0.4 <u>935</u>	0.11	0.16
	NP	2. <del>08</del> 1	<u>1.962.0</u>	0.11	0.19
Natural×5	Equa	0.0607	-0.01	0. <del>07<u>06</u></del>	0.07
	SP	1. <del>55<u>7</u></del>	1. <u>4659</u>	0.08	0.14
	SA	1. <mark>43<u>4</u></mark>	1. <del>32<u>26</u></del>	0.11	0.16
	Asia <u>WP</u>	4.02 <u>3.8</u>	<del>2.09<u>1.9</u></del>	1. <del>93</del> 9	2. <del>30<u>3</u></del>
	NP	8. <del>47<u>4</u></del>	6. <del>89<u>8</u></del>	1. <del>59<u>6</u></del>	2. <del>35<u>4</u></del>
Wind-adjustee	ed Equa	1. <del>35<u>4</u></del>	0. <del>17<u>27</u></del>	1. <u><del>18</del>2</u>	1. <del>21</del> 2
	SP	7. <del>75</del> 6	5. <mark>94<u>8</u></mark>	1. <u>818</u>	2. <del>57<u>6</u></del>
	SA	<del>7.91<u>8.0</u></del>	5. <del>86</del> 9	2. <del>05</del> 1	2. <del>83<u>8</u></del>
	Asia <u>WP</u>	<del>17.34<u>18</u></del>	4.40 <u>6</u>	<u>12.9413</u>	15 <del>.35</del>
	NP	23.11	13 <mark>.41</mark>	9. <del>70</del> 8	<u> 14.5115</u>
10 <sup>-9</sup> kg m <sup>-2</sup> s <sup>-1</sup>	<sup>1</sup> Equa	<del>10.96<u>11</u></del>	0. <del>54<u>55</u></del>	10 <del>.42</del>	<u> 10.6811</u>
	SP	<del>24.50<u>25</u></del>	<u>10.7311</u>	<u>13.7714</u>	<del>18.67<u>19</u></del>
	SA	22 <del>.36</del>	11 <del>.27</del>	11 <del>.08</del>	15 <del>.33</del>
	Asia	0.65	0.16	<del>0.49</del>	0.60
<del>10<sup>-9</sup> kg m<sup>-2</sup> s<sup>-1</sup></del>	+ <u>NP</u>	<del>2.69</del>	<del>2.01</del>	0.68	<del>0.97</del>
in the sensitiv		<del>0.74</del>	<del>0.10</del>	<del>0.64</del>	<del>0.65</del>
area	SP	3.27	<del>2.24</del>	1.04	1.35
	<del>SA</del>	<del>1.81</del>	<del>1.20</del>	<del>0.61</del>	<del>0.79</del>
	AsiaWP	<del>7.21<u>6.9</u></del>	<u>3.212.9</u>	<u>3.994.0</u>	5. <u>141</u>
	NP	16 <mark>.07</mark>	11 <del>.07</del>	5. <del>001</del>	7. <del>73<u>8</u></del>
Fixed-wind-	Equa	5. <del>00</del> 0	0. <u>4850</u>	4. <del>53</del> 5	4. <del>72</del> 7
adjusted	SP	<u></u> 	9. <del>81</del> 9	6. <del>59</del> 6	9. <del>76</del> 8
	SA	<del>19.78</del> 20	<del>10.79</del> 11	<del>8.98</del> 9.1	<del>12.54</del> 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.

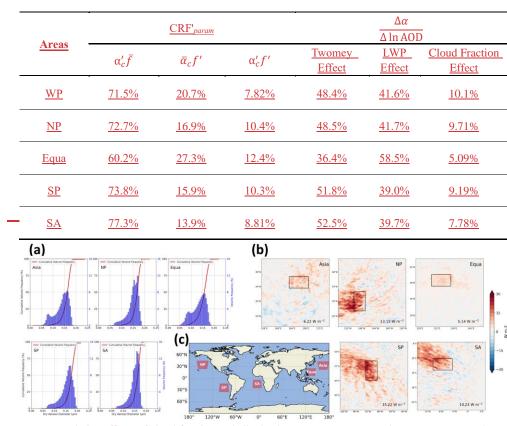
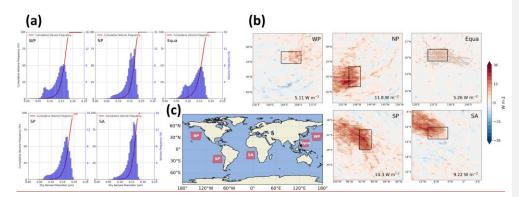


 Table 3. Relative effects of cloud fraction and albedo changes on CRF'*param* and Twomey, LWP, and

 cloud fraction effects to SW\_CLD responses after uniform fixed injection of 10<sup>-9</sup> kg m<sup>-2</sup> s<sup>-1</sup> sea-salt

 aerosols over five ocean regions.



**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<sup>-2</sup> s<sup>-1</sup> 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<sup>-2</sup> s<sup>-1</sup> 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.

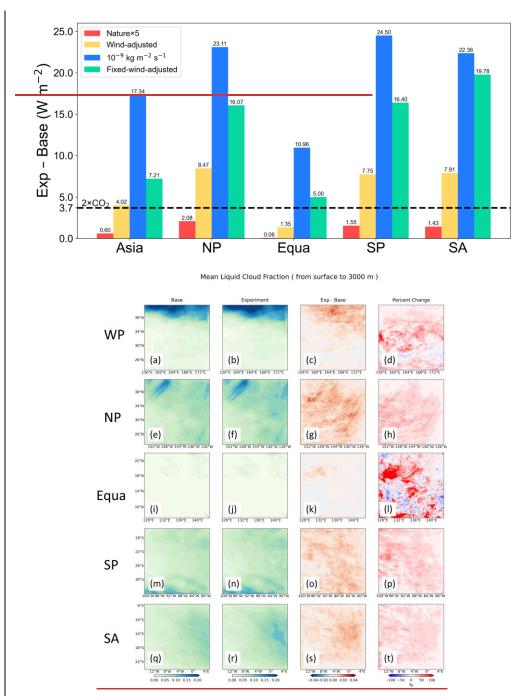
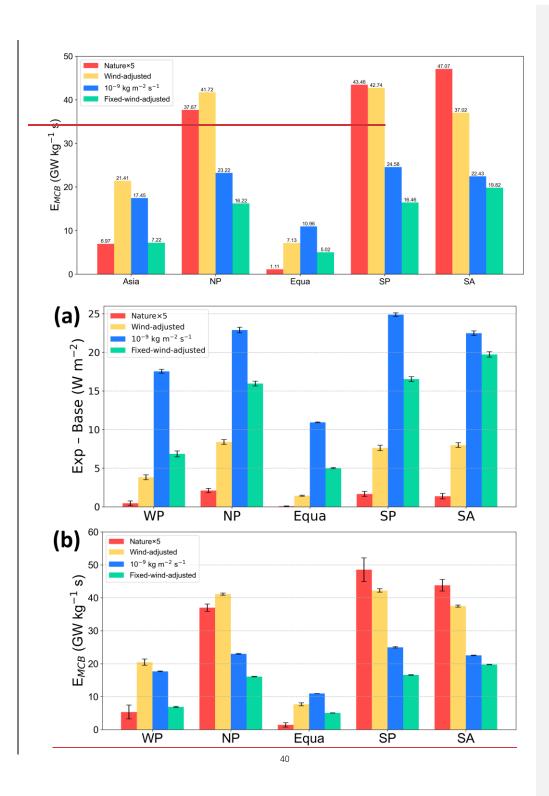
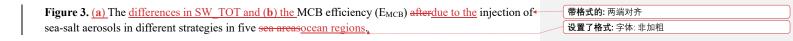
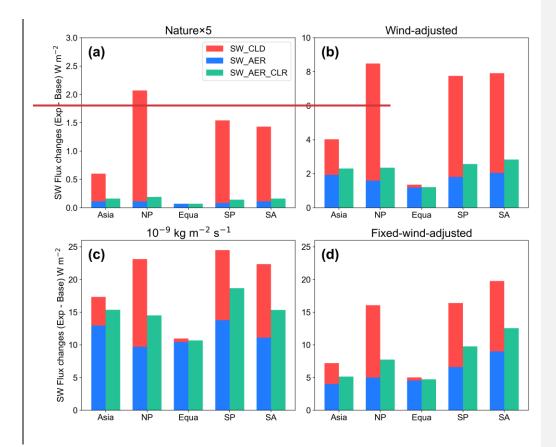


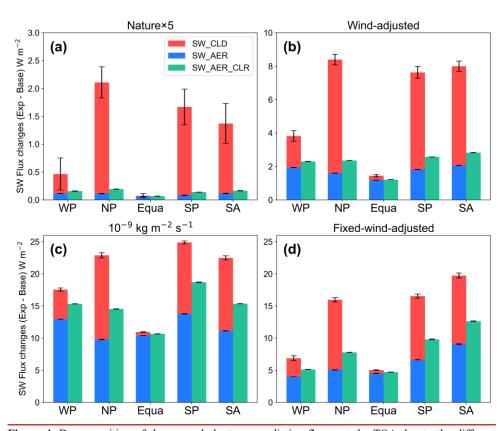
Figure 2. Differences in SW\_TOT due to Column mean liquid cloud fraction from the surface to 3000 m

altitude for five regions. The first to fourth columns are Base, the sensitivity experiment with a uniform injection of  $10^{-9}$  kg m<sup>-2</sup> s<sup>-1</sup> sea-salt aerosols in different ways in five ocean regions. The black dashed line is the radiative flux response required to offsetover the 3.7 W m<sup>-2</sup> radiative forcing caused by entire region, Exp - Base, and the doubling percent change of atmospheric  $CO_2$  concentrations since industrialization Exp - Base, respectively.

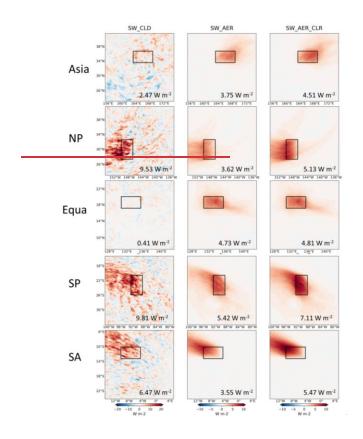


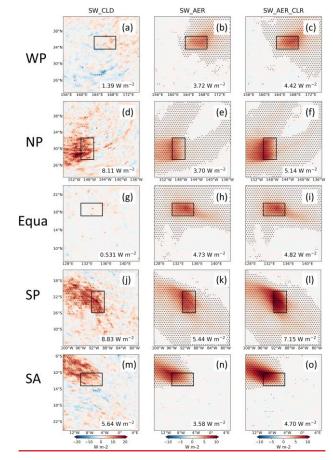




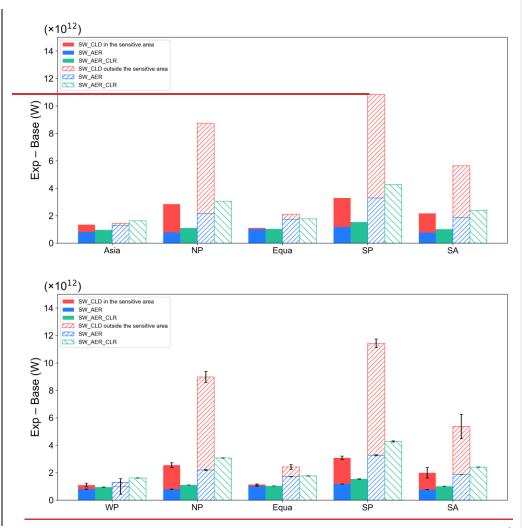


**Figure 4.** Decomposition of the upward shortwave radiative fluxes at the TOA due to the different waysstrategies of injecting sea-salt aerosols in the five regions. Note that the vertical coordinatey-axis ranges are not consistent.

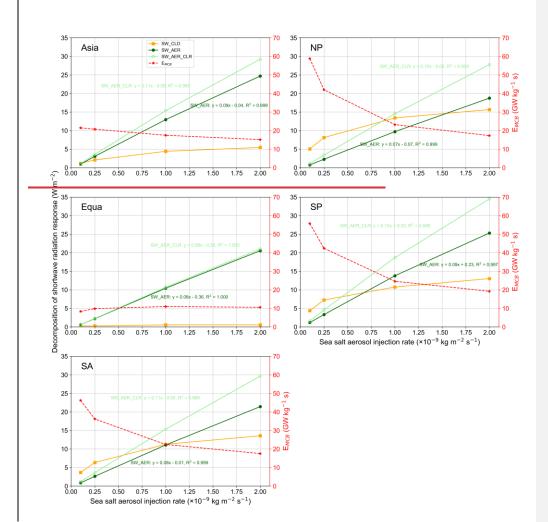




**Figure 5.** Spatial distribution of SW\_CLD (first column), SW\_AER (second column), and SW\_AER\_CLR (third column) responses resulting from the injection of 10<sup>-9</sup> kg m<sup>-2</sup> s<sup>-1</sup> sea-salt aerosols in the sensitive areas over five ocean regions. The values of the radiative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



**Figure 6.** Total SW\_CLD, SW\_AER, and SW\_AER\_CLR responses resulting from the injection of 10<sup>-9</sup> kg m<sup>-2</sup> s<sup>-1</sup> 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 a-slash stylehatching indicate the total radiative response outside the sensitive areas.



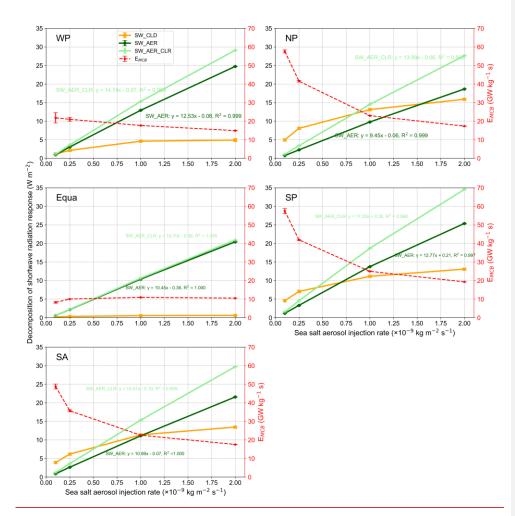
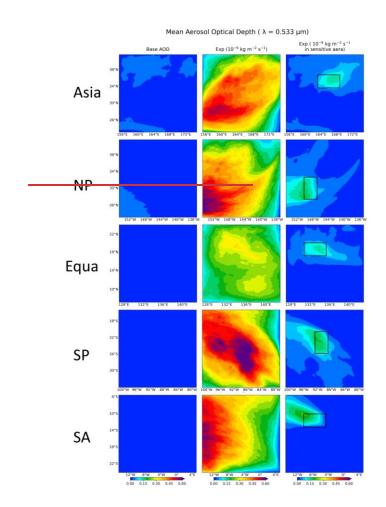
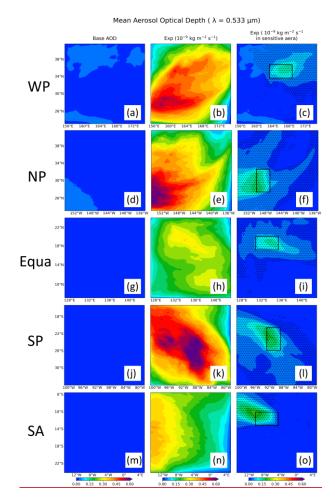


Figure 7. Changes in SW\_CLD, SW\_AER, and SW\_AER\_CLR radiative responses due to sea-salt aerosols uniformly injected in varying amounts in five ocean regions, and corresponding changes in  $E_{MCB}$ . SW\_AER and SW\_AER\_CLR are labeled with the results of the corresponding linear regression analysis.





**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<sup>-9</sup> kg m<sup>-2</sup> s<sup>-1</sup>, and the third column is the AOD after uniform injection in sensitive areas. <u>Areas labeled with dots indicate mean</u> differences that are significant at the 95% confidence level. The black rectangles are sensitive areas.

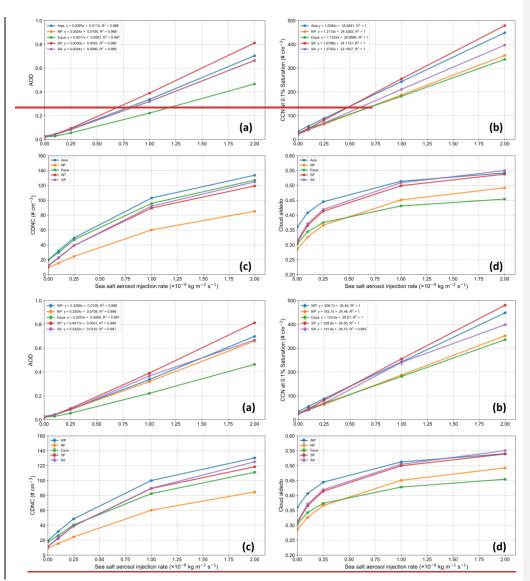
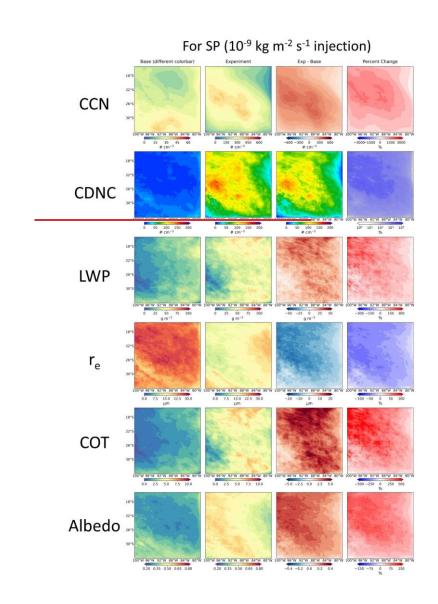
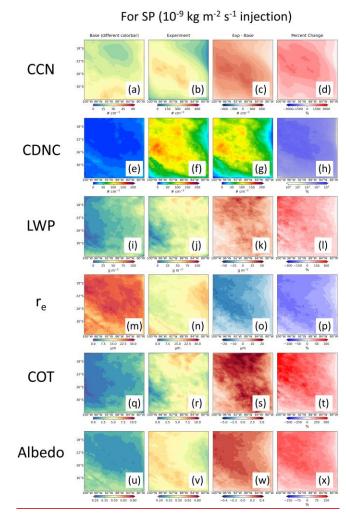
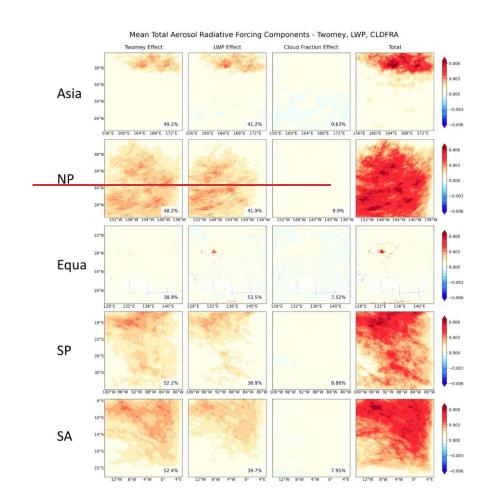


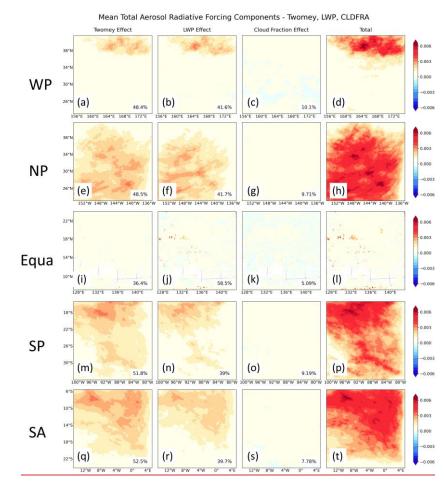
Figure 9. Relationship between changes in regional mean (a) AOD, (b) CCN, (c) CDNC, and (d) cloud albedo due to uniform injection of sea-salt aerosols across the region and the amounts of sea-salt aerosols injected. The results of the linear regression of (a) AOD and (b) CCN on the sea-salt aerosols 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<sup>-2</sup> s<sup>-1</sup> in the SP region. Results are shown for cloud condensation nuclei (CCN, S = 0.1%, # cm<sup>-3</sup>), cloud droplet number concentration (# cm<sup>-3</sup>), liquid water path (LWP, g m<sup>-2</sup>), cloud effective radius ( $r_e$ ,  $\mu$ m), 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.