



What rainfall rates are most important to wet removal of different aerosol types? Yong Wang^{1*}, Wenwen Xia¹ and Guang J. Zhang² ¹Ministry of Education Key Laboratory for Earth System Modeling & Department of Earth System Science, Tsinghua University, Beijing, 100084 China ²Scripps Institution of Oceanography, La Jolla, CA, USA *e-mail: yongw@mail.tsinghua.edu.cn





Abstract. Both frequency and intensity of rainfall affect aerosol wet deposition. With a stochastic 23 24 deep convection scheme implemented into two state-of-the-art global climate models (GCMs), a 25 recent study found that aerosol burdens are increased globally by reduced climatological mean wet removal of aerosols due to suppressed light rain. Motivated by their work, a novel approach is 26 developed in this study to detect what rainfall rates are most efficient for wet removal (scavenging 27 amount mode) of different aerosol species in different sizes in GCMs and applied to the National 28 29 Center for Atmospheric Research Community Atmosphere Model version 5 (CAM5) with and without the stochastic convection cases. Results show that in the standard CAM5, no obvious 30 differences in the scavenging amount mode are found among different aerosol types. However, the 31 scavenging amount modes differ in the Aitken, accumulation and coarse modes showing around 32 10-12, 8-9, and 7-8 mm d⁻¹, respectively over the tropics. As latitude increases poleward, the 33 scavenging amount mode in each aerosol mode is decreased substantially. The scavenging amount 34 35 mode is generally smaller over land than over ocean. With stochastic convection, the scavenging 36 amount mode for all aerosol species in each mode is systematically increased, which is the most prominent along the Intertropical Convergence Zone exceeding 20 mm d⁻¹ for small particles. 37 Regardless of whether the stochastic convection scheme is used, convective precipitation has 38 higher efficiency in removing aerosols than large-scale precipitation over the globe even though 39 convection is infrequent over high-latitudes. The scavenging amount modes in the two cases are 40 41 both smaller than individual rainfall rates associated with the most accumulated rain (rainfall amount mode), further implying precipitation frequency is more important than precipitation 42 intensity for aerosol wet removal. The notion of the scavenging amount mode can be applied to 43 other GCMs to better understand the relation between rainfall and aerosol wet scavenging, which 44 45 is important to better simulating aerosols.





47 1. Introduction

Wet deposition through scavenging by rainfall is an important sink for atmospheric aerosols 48 and soluble gases (Atlas and Giam, 1988; Radke et al., 1980). A correlation between the total 49 rainfall amount or rainfall intensity and air pollution has been documented in many studies (Cape 50 et al., 2012; Pye et al., 2009; Tai et al., 2012). For instance, Dawson et al. (2007) found a strong 51 sensitivity of the particulate matter with diameters less than 2.5 µm (PM_{2.5}) concentrations to 52 53 rainfall intensity over a large region of the eastern United States from sensitivity tests using a regional numerical model. Besides precipitation intensity, precipitation frequency also influences 54 aerosol wet deposition. In the Geophysical Fluid Dynamics Laboratory (GFDL) chemistry-climate 55 model AM3, Fang et al. (2011) found wet scavenging has a stronger spatial correlation with rainfall 56 frequency than intensity over the United States in January. Mahowald et al. (2011) explored the 57 role of precipitation frequency in dust wet deposition based on model simulations and noted the 58 frequency of precipitation rather than the amount of precipitation controls the fraction of dust wet 59 vs dry deposition outside dust source regions. 60

Hou et al. (2018) investigated the sensitivity of wet scavenging of black carbon (BC) to 61 precipitation intensity and frequency respectively in the Goddard Earth Observing System 62 63 Chemistry (GEOS-Chem) model. The frequency and intensity of precipitation from the GEOS-5 run were used to drive the GEOS-Chem. With the sensitivity tests by artificially perturbating 64 65 precipitation frequency and intensity respectively, they found that the deposition efficiency and hence the lifetime of BC have higher sensitivities to rainfall frequencies than to rainfall intensities. 66 Even with the same mean total rainfall, a different combination of precipitation intensity and 67 frequency results in different removal efficiency of BC. Although these studies investigate the 68 impacts of precipitation intensity and frequency on aerosol wet removal, it is not clear yet 69 70 climatologically what rainfall rates contribute the most to aerosol wet deposition.

Wang et al. (2021) recently found that the frequency of total rainfall in the range from 1 to 20 71 mm d⁻¹ plays a critical role in regulating the annual mean wet deposition rates of aerosols, 72 especially over the tropics and subtropics. By suppressing the too frequent occurrence of 73 convection in this rainfall intensity range with the introduction of a stochastic deep convection 74 75 scheme (Wang et al., 2016), the aerosol burdens in two global climate models (GCMs) were 76 significantly increased, with the simulated aerosol optical depth (AOD) agreeing better with 77 observations. Based on their work, several interesting questions on the relation between rainfall and aerosol wet removal can be asked: (1) climatologically, what rain rates have the highest 78





efficiency in removing atmospheric aerosols? (2) how much convective and large-scale precipitation contribute to it? (3) for different aerosol types and sizes, does the rain rate most efficient in washing out aerosols differ? (4) also, does it differ over different latitudes and continents/oceans?

To address these questions, this study develops a novel approach to identify the rainfall 83 intensity associated with the most efficient aerosol wet scavenging and applies it to different 84 85 aerosol species at different aerosol sizes in the NCAR CAM5. The paper is organized as follows. Section 2 presents the CAM5 model, experiments, observations and methods. In section 3, 86 precipitation characteristics especially for the amount (defined by daily cumulative rainfall) 87 distributions in two simulations evaluated with observations are presented first. With distinct 88 precipitation features (e.g., frequency and amount) in two simulations, their aerosol wet deposition 89 90 features and mass concentrations are shown. Discussion and conclusions are given in section 4.

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92 2. Experiments, methods and observations

93 2.1. Model and simulations

This study uses the National Center for Atmospheric Research (NCAR) Community 94 95 Atmosphere Model version 5.3 (CAM5.3). As the atmosphere model of the NCAR CESM, CAM5.3 in a standard configuration has a vertical resolution of 30 levels from the surface to 3.6 96 97 hPa and a horizontal resolution of $1.9^{\circ} \times 2.5^{\circ}$ using finite volume dynamical core. Deep convection is parameterized using the Zhang and McFarlane (1995) (ZM) scheme with dilute 98 convective available potential energy (CAPE) modification by Neale et al. (2008) while the 99 shallow convection scheme uses Park and Bretherton (2009). The Bretherton and Park (2009) 100 moist turbulence parameterization is used to present the stratus-radiation-turbulence interactions. 101 102 The Morrison and Gettelman (2008) (MG) scheme is for large-scale stratiform cloud microphysics. The radiative transfer calculations are based on the Rapid Radiative Transfer Model (RRTM) 103 (Iacono et al., 2008). The properties and process of major aerosol species (sulfate, mineral dust, 104 sea salt, primary organic matter, secondary organic aerosol and black carbon) are treated in the 105 modal aerosol module (MAM) in which distributions of aerosol size are represented by three 106 lognormal modes (MAM3): Aitken, accumulation and coarse modes (Liu et al., 2012). The number 107 108 mixing ratio of each mode and the associated mass mixing ratios of aerosol types in each mode 109 are predicted.

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Aerosol wet removal consists of in-cloud scavenging and sub-cloud scavenging, which are





111 both treated by the aerosol wet removal module. For in-cloud scavenging, the rainfall production rates and cloud water mixing ratios are used to calculate first-order loss rates for cloud water, 112 which is further multiplied by "solubility factors" to obtain aerosol first-order loss rates. The 113 solubility factors can be interpreted as (tuning factor) \times (aerosol fraction in cloud droplets). The 114 stratiform in-cloud scavenging only influences the stratiform-cloud-borne aerosol particles with 115 solubility factors of 1.0 (0 for interstitial aerosols). The convective in-cloud scavenging is 116 117 computed both using an in-convection activation fraction and a solubility factor. For stratiform and convective sub-cloud scavenging, the first-order removal rate of interstitial aerosols is equal 118 to (rain rate) \times (scavenging coefficient) \times (solubility factor). There is no sub-cloud scavenging 119 for stratiform-cloud-borne aerosols. 120

We use the CAM5.3 simulation output in Wang et al. (2021) for our analysis. The runs with the default ZM scheme (referred to as CAM5) and the stochastic deep convection scheme (referred to as STOC) (Plant and Craig, 2008; Wang et al., 2016) are Atmospheric Model Intercomparison Project (AMIP) type simulations with the present-day (PD) aerosol emission scenario. The prescribed, seasonally varying climatological present-day (averaged over 1982-2001) sea surface temperatures (SSTs) and sea ice extent, recycled yearly force the two simulations which are run for 6 years and the last 5 years are used for analysis.

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129 **2.2. Methods**

Both precipitation frequency and intensity contribute to the rainfall amount. Wang et al. (2016, 2021) show that the occurrence frequency of observed and simulated precipitation varies with precipitation intensity largely following exponential functions. Therefore, using a log-linear coordinate system to examine the contribution from each rainfall interval will allow an easier comparison among different rainfall intensity ranges. The contributions from different rainfall rates to the total rainfall amount can be described using the following form (Kooperman et al. 2018):

$$P(R_i) = \frac{1}{\Delta \ln(R)} \frac{1}{N_T} \sum_{k=1}^{N_T} r_k \cdot I\left(R_i^l \le r_k < R_i^r\right)$$

where *i* is the bin index, *r* is the daily rain rate, R_i is the rainfall bin center with bounds R_i^l and R_i^r which is logarithmically spaced covering 4 orders of magnitude of rainfall intensity from 0.1 to 1000 mm d⁻¹. The bin width is set to $\Delta \ln(R) = \Delta R/R = 0.1$, meaning that the bin interval is 1/10 of the center value (*R*). N_T is the total number of days, and *I* is a binary operator that has a

(1)





142 value of 1 within the rainfall bin of interest and 0 outside. Thus, $P(R_i)$ is the contribution to the total precipitation by the rainfall rates centered at R_i . Graphically, the area under the curve of P in 143 a log-linear plot gives the total amount of mean precipitation. Similarly, within the total 144 precipitation rate bin centered at R_i , the contributions from convective (P_c) and large-scale (P_L) 145 146 precipitation are given respectively by:

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$$P_{C}(R_{i}) = \frac{1}{\Delta \ln(R)} \frac{1}{N_{T}} \sum_{k=1}^{N_{T}} r_{k}^{C} \cdot I\left(R_{i}^{l} \le r_{k} < R_{i}^{r}\right) \quad (2)$$
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$$P_{L}(R_{i}) = \frac{1}{\Delta \ln(R)} \frac{1}{N_{T}} \sum_{k=1}^{N_{T}} r_{k}^{L} \cdot I\left(R_{i}^{l} \le r_{k} < R_{i}^{r}\right) \quad (3)$$

where r^{C} and r^{L} are the convective and large-scale rainfall contributions respectively to the 149 total rainfall within the bin r_k . Eqs. (1) through (3) allow us to determine contributions to aerosol 150 151 wet scavenging from convective and stratiform clouds separately.

152 Note that Eqs. (2) and (3) are different from those used in previous studies (e.g., O'Brien et al., 2016, Wang et al. 2021), where the rainfall bin used for occurrence count is specified using 153 convective and large-scale rainfall separately. The use of total precipitation to define the rainfall 154 bin has the advantage of allowing us to derive partitioned frequency distributions conditioned on 155 156 total precipitation rates.

A similar approach can be used to relate the wet removal of aerosols to rainfall intensity. The 157 amount distribution of wet removal (W) for a given aerosol type under different rainfall intensity 158 is calculated at each model grid point before area-weighted averaging over regions of interest: 159

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$$W(R_i) = \frac{1}{\Delta \ln(R)} \frac{1}{N_T} \sum_{k=1}^{N_T} d_k \cdot I\left(R_i^l \le r_k < R_i^r\right) \quad (4)$$

where d is the daily wet deposition rate for a given aerosol type, including in- and below-cloud 161 wet deposition fluxes from both convective and stratiform clouds. Akin to the amount distribution 162 of precipitation, the amount distribution of aerosol wet scavenging graphically depicts how much 163 164 accumulated wet deposition is produced by different rain rates, where the area under the distribution is the total mean wet deposition rate. The rainfall intensity band that contributes the 165 most to the total rainfall or aerosol wet scavenging will be referred to as the rainfall or scavenging 166 amount mode, respectively. 167

With Eq. (4), the synergy of frequency and intensity of rainfall on the wet deposition of 168 169 aerosols are included. The rainfall intensity associated with the peak amount of wet removal can be determined accordingly, telling us what precipitation intensity is most efficient in removing 170 aerosols from the atmosphere. Applying it to different aerosol types in different aerosol size modes, 171





172 individual precipitation intensity most effective in aerosol scavenging is obtained.

The amount distribution of total wet removal of aerosols under different total precipitation intensity can be further decomposed into contributions of wet deposition fluxes from convective and stratiform clouds respectively, similar to the decomposition of precipitation amount:

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$$W_{C}(R_{i}) = \frac{1}{\Delta \ln(R)} \frac{1}{N_{T}} \sum_{k=1}^{N_{T}} d_{k}^{C} \cdot I\left(R_{i}^{l} \le r_{k}^{T} < R_{i}^{r}\right)$$
(5)

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$$W_L(R_i) = \frac{1}{\Delta \ln(R)} \frac{1}{N_T} \sum_{k=1}^{N_T} d_k^L \cdot I\left(R_i^l \le r_k^T < R_i^T\right)$$
(6)

where d^T , d^C and d^L is the daily wet deposition rates from all, convective and stratiform clouds respectively. Thus, for each precipitation bin, the sum of wet removal from convective clouds (W_c) and that from stratiform clouds (W_L) is equal to the total wet deposition rate (W). As a result, the fractional contribution of aerosol wet scavenging from individual cloud processes (i.e., W_C/W and W_L/W) can be obtained.

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184 2.3. Observations

The precipitation characteristics in the two simulations are evaluated with observations. 185 Among them, the total rainfall mean state is evaluated against the Global Precipitation Climatology 186 Project (GPCP) monthly product (version 2.1) at a resolution of 2.5° (Adler et al., 2003) and the 187 Tropical Rainfall Measuring Mission (TRMM) 3B43 monthly observations at a resolution of 1° 188 over (50°S, 50°N) (Huffman et al., 2012a) while the TRMM 3A12 monthly observations at a 189 resolution of 0.5° (Huffman et al., 2007) is used to evaluate the mean convective and large-scale 190 precipitation. A daily estimate of GPCP version 1.2 at 1° horizontal resolution (GPCP 1DD) 191 (Huffman et al., 2001, 2012b) and the TRMM 3B42 version 7 daily observations at a resolution of 192 193 0.25° over (50°S, 50°N) (Huffman et al., 2007) are used in the evaluation of the precipitation frequency and amount distribution. To make a consistent comparison with the model simulations, 194 observations are regridded to the same CAM5 grid points. 195

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197 **3. Results**

198 **3.1. Precipitation**

Figure 1 shows the latitudinal distributions of total, convective and large-scale precipitation in GPCP, TRMM, CAM5 and STOC. Overall, the total mean precipitation distributions in CAM5 and STOC runs are comparable, except over the northern tropics where the STOC run simulates mean rainfall slightly larger than the CAM5 run. In comparison with observations, the total





precipitation in both simulations is overestimated in the tropics and subtropics while that in midand high-latitudes agrees well (Fig. 1a). The overestimated total precipitation over the tropics and
subtropics in both simulations is dominantly from the overestimated convective precipitation (Fig.
1b). Nonetheless, compared to the extremely small large-scale rainfall contribution in the CAM5
run, the increased large-scale precipitation in the STOC run, though mainly contributing to the
further increase of total precipitation in the northern tropics, results in a better agreement with the
TRMM observations.

The distributions of total rainfall amount for GPCP, TRMM, CAM5 and STOC over the 210 tropics (20°S, 20°N), subtropics and midlatitudes (20°N, 50°N), and high-latitudes (50°N, 90°N) 211 are shown in Figure 2a-c. Over the tropics, the distribution in STOC exhibits more rainfall from 212 more intense rain rate and less rainfall from light rain than that in CAM5, thus the rainfall amount 213 mode in STOC (around 40 mm d^{-1}) is much stronger than that in CAM5 (~20 mm d^{-1}), falling 214 between the TRMM and GPCP observed rainfall amount mode (30-50 mm d⁻¹) (Fig. 2a). The weak 215 216 amount mode of total rainfall in CAM5 is controlled by convective precipitation rather than largescale precipitation in terms of their respective distributions and fractional contributions at rain rates 217 ranging from 1 to 20 mm d⁻¹ (Fig. 2d&g). In contrast, convective and large-scale rainfall in STOC 218 both represents the observed amount mode of total rain. The shift of the total rainfall amount mode 219 to a larger value in STOC is due to the increased (decreased) fractional contribution of convective 220 precipitation at rain rates larger (smaller) than ~20 mm d⁻¹ (Fig. 2g). Over the subtropics and 221 midlatitudes, the amount mode of total rainfall in CAM5 is comparable to that over the tropics 222 $(\sim 20 \text{ mm d}^{-1})$. Again, compared with CAM5, the rainfall amount mode in the STOC run shifts 223 rightward better matching GPCP and TRMM observations (Fig. 2b). The representation of 224 convective and large-scale precipitation for the observed amount mode of total rainfall in the two 225 226 simulations is the same as that over the tropics except large-scale precipitation in CAM5 which represents the observed amount mode of total rain as well (Fig. 2e). In contrast to the tropics, the 227 228 difference of the fractional contribution between large-scale and convective precipitation at rain rates between 1 to 20 mm d⁻¹ in the CAM5 run is reduced due to the decreased convective and 229 increased large-scale fractional contributions (75% vs. 25%) (Fig. 2h). With the introduction of 230 the stochastic deep convection parameterization, the STOC run suppresses the sub-tropical and 231 midlatitude convection, further decreasing their fractional contributions relative to CAM5. At rain 232 rates larger than 20 mm d⁻¹, although STOC enhances the fractional contribution of convection, 233 large-scale precipitation, as in CAM5, still makes more contributions. Since large-scale 234





precipitation dominates the total precipitation over high latitudes, the amount distributions of total rainfall are similar between the two simulations (Fig. 2c). Despite this, the amount of convective rainfall and the associated fractional contribution between 1 and 10 mm d⁻¹ are reduced in the STOC run compared with that in the CAM5 run (Fig. 2f&i).

For a given rain rate, its amount contribution is determined by frequency (f) only $(P(R_i) =$ 239 $f(R_i)R_i$). The frequency distributions of the total precipitation in observations and simulations, 240 and contributions from convective and large-scale precipitation in CAM5 and STOC runs are 241 shown in Figure 3. Over the tropics, where there is frequent convection, although the frequency of 242 total precipitation in the STOC run is slightly higher than that in the CAM5 run at rain rates 243 between 0.1 and 2 mm d^{-1} , the frequency of rain rates between 2 and 20 mm d^{-1} in STOC is greatly 244 reduced, much closer to GPCP and TRMM. Furthermore, for rain rates larger than 20 mm d⁻¹, the 245 simulated frequency in STOC matches TRMM very well (Fig. 3a). These changes in the total 246 rainfall frequency can be explained by those in individual large-scale and convective components, 247 i.e., a decrease of the frequency of convective precipitation is the main contributor to the frequency 248 change of total rain rates between 2 and 20 mm d⁻¹ while both large-scale and convective 249 precipitation is responsible for the frequency increase of total rain rates larger than 20 mm d⁻¹ (Fig. 250 3d). These results are consistent with Wang et al. (2021). As the latitude increases poleward 251 associated with the decreasing frequency contribution of convection, the difference of the 252 253 frequency of total rainfall between CAM5 and STOC runs becomes less prominent (Fig. 3b&c). However, relative to the frequency of convective precipitation in the CAM5 run, similar changes 254 to those over the tropics in the STOC run are still evident (Fig. 3e&f). A chain linking the changes 255 of frequency and amount from CAM5 to STOC is summarized here: with the stochastic deep 256 convection parameterization, the frequency of convection for rain rates between 1 and 20 mm⁻¹ is 257 258 reduced in STOC, resulting in the decreased amount of total rain within this range and thus the associated shift of the rainfall amount mode to larger rainfall intensity. 259

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261 **3.2. Wet deposition of aerosols**

With precipitation features in CAM5 and STOC runs in mind, aerosol wet deposition in the two simulations is explored. Figure 4 demonstrates the simulated distributions of wet removal of different aerosol species in different modes over the tropics. Overall, the shape of the distributions of wet removal for all aerosol species in the three modes in both simulations resembles that of the rainfall distribution. Nonetheless, the scavenging amount modes are not equal to the amount modes





of total rainfall as shown in Fig. 2a, especially for large particles. Specifically, in CAM5, for sulfate, 267 sea salt and secondary organic aerosol (SOA) in the Aitken mode, the scavenging amount modes 268 are around 10-12 mm d^{-1} , smaller than the rainfall amount mode of ~20 mm d^{-1} . As the aerosol 269 size increases, the scavenging amount modes decrease to 8-9 and 7-8 mm d⁻¹ in the accumulation 270 and coarse modes, respectively, implying larger particles are easier to be removed by lighter 271 rainfall. The feature that the scavenging amount mode is smaller than the amount mode of total 272 273 rain suggests the frequency of light precipitation plays a more important role in regulating the amount of aerosol wet scavenging than that of rainfall. Additionally, in contrast to other aerosols, 274 the wet removal of sea salt is more sensitive to light precipitation. With the rain rate increasing 275 beyond 1 mm d⁻¹, the wet deposition rate of sea salt increases more rapidly than that of other 276 aerosols (i.e., steeper curve). As a response to the shift of the amount mode of total rainfall to a 277 larger value from CAM5 to STOC, the scavenging amount modes for all aerosols in the three 278 modes in STOC are increased accordingly. Owing to the decreased rainfall amount and the high 279 280 occurrence frequency at rain rates smaller than 20 mm d⁻¹ (Fig. 3a&d), the decrease of wet removal in this rainfall range overwhelms the wet deposition increase at rain rates beyond 20 mm d⁻¹. As a 281 result, compared to CAM5, the net decreases of regionally averaged wet removal for all aerosols 282 in the three modes in STOC are found. The largest relative decreases in the Aitken, accumulation 283 and coarse modes are found in black carbon (-33.3% from 0.03 to 0.02 mg/m²/day), SOA (-50% 284 285 from 0.004 to 0.002 mg/m²/day), and dust (-20.9% from 7.60 to 6.01 mg/m²/day), respectively.

The distributions for the subtropics and midlatitudes, and high latitudes are shown in Figures 286 5 and 6, respectively. Same as in the tropics, the similar distributions of different aerosol species 287 in different modes over these two regions are found except for dust in the coarse mode in the 288 subtropics and midlatitudes where two peaks are found: one located at the rain rate around 0.8 mm 289 290 d^{-1} and the other around 8 mm d^{-1} (Fig. 5). With the suppression of the total rainfall amount between 1-10 mm d⁻¹ (Fig. 2b), for dust in the coarse mode over (20°N, 50°N), the amount magnitudes of 291 292 two peaks are comparable in the STOC run in contrast to the distinctly different magnitudes of two peaks in the CAM5 run. The scavenging amount modes for all aerosols over these two latitudinal 293 belts are smaller than the rainfall amount modes as well (Fig. 2b&c). In comparison with CAM5, 294 again, the scavenging amount mode shifts rightward and the regional mean of wet removal for all 295 aerosols is reduced in the STOC run (Figs. 5&6). Due to a decrease of mean rain as latitude 296 297 increases, the scavenging amount mode and mean wet removal for all aerosols are increasingly 298 reduced.





299 Besides the scavenging amount mode different from the amount mode of total rainfall, the fractional contributions of wet deposition rates from stratiform and convective clouds differ more 300 significantly from the fractional contributions of convective and large-scale precipitation to the 301 total rainfall amount. Over the tropics (Figure 7), for all aerosols in the Aitken and accumulation 302 modes, in the range of rain rates from 0.1 to 100 mm d⁻¹, the total wet removal is almost all from 303 convective clouds for both CAM5 and STOC despite the fact that the fractional contribution of 304 305 large-scale rainfall to the total rainfall amount reaches as much as 25% at rain rates greater than 20 mm d^{-1} (Fig. 2g). For rain rates higher than 100 mm d^{-1} , while the large-scale contribution to 306 the total rainfall amount is up to 50-60% in two runs, only for sulfate, sea salt, dust, black carbon 307 and primary organic matter (POM) in the accumulation mode in STOC does the fractional 308 contribution of wet removal from stratiform clouds reach 50%. In contrast, for large aerosol 309 particles (i.e., sulfate, sea salt and dust in the coarse mode), the role of stratiform clouds becomes 310 important. For example, at rain rates ranging from 0.1 to 10 mm d⁻¹ in which the large-scale 311 contribution to the total rainfall amount can almost be neglected in both simulations, wet 312 deposition from stratiform clouds accounts for 10-25% in CAM5 and 25-40% in STOC. This is 313 because large particles in stratiform clouds can be easily removed (Abdul-Razzak and Ghan, 2000) 314 by acting as cloud condensation nuclei. As a response to a rapid increase of the large-scale 315 fractional contribution to the total rainfall amount when rain rates exceed 100 mm d⁻¹ in STOC, 316 317 the fractional contribution of wet removal from the stratiform clouds rockets up to 100%.

As for the subtropics and midlatitudes (Figure 8), as rain rates increase, the changes of the 318 fractional contributions from convective and stratiform clouds in the two simulations follow the 319 changes of the fractional contributions to the total rainfall amount well. However, their fractional 320 contributions to rainfall and aerosol wet scavenging differ dramatically. Take rainfall rates between 321 1 to 10 mm d⁻¹ for example. Although the fractional contribution of wet removal of aerosols in the 322 Aitken and accumulation modes from stratiform clouds increases slightly in the two simulations 323 (~12% in STOC larger than ~5% in CAM5), this still shows a large contrast to the large-scale 324 fractional contribution to the total rainfall amount (>25%) (Fig. 2h). Different from the tropics, 325 after rain rates exceed 10 mm d⁻¹, the fractional contributions from stratiform clouds for all aerosols 326 in these two modes in CAM5 and STOC climb to 25%. For aerosols in the coarse mode between 327 1 and 10 mm d⁻¹, the fractional contribution from stratiform clouds in CAM5 is larger than 25% 328 329 but still much smaller than that from convective clouds. Associated with the decreased (increased) convective (large-scale) precipitation in STOC, the individual fractional contributions to the total 330





wet removal from stratiform and convective clouds are comparable. As rain rates increase beyond
 20 mm d⁻¹, the fractional contribution from stratiform clouds in two runs becomes dominant with

a larger contribution from convective clouds in STOC than in CAM5.

In high latitudes (Figure 9), even though precipitation is mainly from large-scale rainfall with 334 little convection (Figs. 2i & 3f), it is surprising that the aerosol particles in the Aitken and 335 accumulation modes at rain rates between 0.3-20 mm d⁻¹ in both simulations are still mainly 336 337 removed by convective clouds. This is largely attributed to the fact that in-cloud interstitial aerosols are removed by convection only (see section 2.2). Only for total rainfall larger than 20 mm d⁻¹ does 338 wet removal from stratiform clouds dominate over that from convective clouds. In contrary to the 339 behavior of small aerosol particles, the wet scavenging of aerosol particles in the coarse mode in 340 CAM5 and STOC behave consistently across the entire rainfall range, with the fractional 341 contribution from large-scale overwhelming that from convective clouds (exceeding 75% in STOC 342 larger than in CAM5). 343

With these aerosol wet deposition features and the associated rainfall amount and frequency 344 characteristics shown in section 3.1, the cause for the decrease of the mean wet removal in STOC 345 compared to CAM5 is summarized as follows. For all aerosol species in three modes over three 346 latitudinal belts, the rain rates at which there is a large amount of wet removal range from 1 to 20 347 mm d⁻¹ although the individual scavenging amount mode differs (Figs. 4-6). In this rainfall 348 349 intensity range, the frequency decrease of convective precipitation and unchanged large-scale precipitation (Fig. 3) result in the reduced amount of this total rainfall intensity band (Fig. 2). This 350 change of the total/convective rainfall amount and the behavior that aerosols especially for 351 particles in the Aitken and accumulation modes are mainly removed from convective clouds 352 (except sulfate, sea salt and dust in the coarse mode in high latitudes) (Figs. 7-9) work together for 353 354 the climatological mean wet deposition decrease.

The framework proposed in section 2.3 is difficult to use for assessing the geographic 355 distribution of the scavenging amount mode because it is based on discrete logarithmic bins that 356 can under-sample the data in some regions with little precipitation. In this regard, an alternative 357 approach is proposed. At each grid point, the daily precipitation intensity during the entire N_T 358 days is sorted in an ascending order with which the corresponding wet deposition rate is 359 accumulated accordingly. Then the rainfall intensity associated with the median accumulated wet 360 361 removal is used as a complementary statistic of the scavenging amount mode which is independent of the rainfall bin structure. In CAM5 (Fig. 10), the geographic patterns in general resemble that 362





of annual mean precipitation (Wang and Zhang, 2016), showing maximum centers (~6-10 mm d⁻ 363 ¹) along the Intertropical Convergence Zone (ITCZ), the South Pacific Convergence Zone and in 364 the Indian Ocean. Besides these regions, the scavenging amount mode for SOA in the Aitken mode 365 also peaks over the north Pacific and Amazonia. Even though rainfall intensity between 1 and 20 366 mm d⁻¹ occurs more frequently over oceans than over land (Wang et al., 2016), it is easier for 367 aerosols over land except the Tibetan Plateau to be removed by lighter rainfall. In comparison with 368 CAM5, increases of the simulated scavenging amount mode in STOC are found across the globe 369 but most significant along the ITCZ where for some small aerosol particles (e.g., sulfate, sea salt 370 and SOA in the Aitken and accumulation modes) it can exceed 20 mm d⁻¹ (Fig. 11). 371

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373 3.3. Aerosol amount changes

374 To investigate the impact of reduced aerosol wet removal on aerosol mass concentrations in 375 the atmosphere, Figure 12 presents latitude-pressure cross-sections of changes in annual mean 376 mass mixing ratios of different aerosol species between CAM5 and STOC. The aerosol concentrations for all species are increased throughout the troposphere. But the peak-heights differ 377 for different aerosol types. Sulfate and sea salt peak near the surface while dust, black carbon, 378 POM and SOA show maxima at around 800 hPa. In terms of the latitudinal variation, the largest 379 changes are broadly located in the tropics and midlatitudes in both hemispheres, corresponding to 380 381 ITCZ convection region and midlatitude cyclone regions. The exception is dust, for which the maximum is between the equator and 30 °N where the Sahara Desert is. In addition to the primary 382 maxima at the lower troposphere, a secondary peak is found at the upper troposphere (~ 200 hPa) 383 for all aerosol species, especially in the tropics. The significant increases of aerosols in the lower 384 troposphere primarily result from reduced light rain. As will be seen in Figure 13 below, convective 385 386 transport also has a substantial contribution. The secondary peak is apparently associated with convective transport. To verify this, Figure 13 shows the difference of convective mass flux 387 between STOC and CAM5 and the vertical transport of selected aerosol types. Although the mass 388 flux in deep convection in the lower troposphere is reduced because of the reduced frequency of 389 convection (Fig. 13a), the increases in aerosol concentrations still lead to the enhancement of the 390 vertical aerosol transport by deep convection (e.g., POM and SOA, Fig. 13c-d). In the upper 391 troposphere, there is an increase in convective mass flux. This is due to the increase of the 392 393 frequency of more intense convection and precipitation (Fig. 3). Correspondingly, there is more vertical aerosol transport in the upper troposphere (Wang and Zhang, 2016). Other aerosol species 394





transported by deep convection have similar results (figure not shown).

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397 4. Discussion and conclusions

This study aims to identify the scavenging amount modes for different aerosol species in 398 different sizes. In the standard CAM5 with too much light precipitation mainly associated with too 399 frequent convection, for a given aerosol mode, there are no obvious differences in the scavenging 400 401 amount modes among different aerosol species. However, as the aerosol size grows, the scavenging amount mode decreases, suggesting that lighter rainfall is more efficient at removing 402 larger particles. Specifically, the scavenging amount modes in the Aitken, accumulation and coarse 403 modes are around 10-12, 8-9 and 7-8 mm d⁻¹, respectively over the tropics. As latitude increases 404 poleward, the scavenging amount mode in each aerosol mode is decreased substantially. In 405 comparison with the scavenging amount modes over the ocean, the values over land are generally 406 smaller. With the effective reduction of too frequent convection by the stochastic deep convection 407 parameterization, STOC systematically increases the scavenging amount mode for all aerosol 408 species in each mode which is the most prominent along the ITCZ exceeding 20 mm d⁻¹ for small 409 particles. For both CAM5 and STOC, the scavenging amount modes of all aerosols are smaller 410 411 than the rainfall amount modes, implying the rainfall intensity associated with the most accumulated rain does not equal the most accumulated wet deposition. The rainfall frequency plays 412 413 a more critical role in regulating the accumulated aerosol wet deposition than in the most accumulated rainfall. 414

This study also demonstrates that convective precipitation has higher efficiency in removing 415 atmospheric aerosols than large-scale precipitation. Even at high latitudes where convection is 416 infrequent, aerosol wet scavenging especially for fine particles is still dominantly from convective 417 418 precipitation. This implies that there is an inconsistency of fractional contributions from convective and stratiform clouds between precipitation and aerosol wet removal, which probably 419 420 will cause a problem in high-resolution GCM simulations. As resolution increases, if convective precipitation theoretically decreases, convection-induced decreases of aerosol wet removal will 421 overwhelm the increases of aerosol wet removal from the large-scale precipitation increase. As a 422 423 result, aerosol burdens might be increasingly enhanced.

The approach proposed in this study to determine the scavenging amount mode and the corresponding fractional contributions from stratiform and convective clouds can be applied to other GCMs to better understand the individual relation between rainfall and aerosol wet





427	scavenging, which is of importance to simulating aerosols in GCMs. The high sensitivity of the
428	scavenging amount mode to the representation of the rainfall amount distribution at rain rates
429	between 1 and 20 mm d ⁻¹ and the vital role of aerosol wet removal from convective clouds
430	highlight that the improvement of the aerosol wet deposition in GCMs should focus on not only
431	the parameterization of aerosol wet scavenging itself but also the parameterization of convection.
432	
433	Code availability. The CESM1.2.1-CAM5.3 source code can be downloaded from the CESM
434	official website http://www2.cesm.ucar.edu. The stochastic convection code is accessible from an
435	open repository, Zenodo (https://doi.org/10.5281/zenodo.4543261).
436	
437	Data availability. The GPCP 1DD data is available from NASA GSFC RSD
438	(https://psl.noaa.gov/data/gridded/data.gpcp.html). TRMM data is available from
439	https://gpm.nasa.gov/data/directory. The CAM5 simulation output is provided in an open
440	repository Zenodo (https://doi.org/10.5281/zenodo.4259554).
441	
442	Author contributions. YW conceived the idea. YW conducted the model simulations and
443	performed the analysis. YW and GJZ interpreted the results. YW wrote the paper, with
444	contributions from GJZ. All authors discussed the results and edited the manuscript.
445	
446	Competing interests. The authors declare that they have no conflict of interest.
447	
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541 Figure captions

- 542 Figure 1. Zonal mean (a) total (solid line), (b) convective (solid line) and large-scale (dashed line)
- 543 precipitation in CAM5 (blue), STOC (red) and TRMM (black). Zonal mean total rain in GPCP
- 544 (green) is also shown.
- 545 Figure 2. Amount distributions of (a-c) total, (d-f) convective and large-scale precipitation, and
- 546 (g-i) fractional contributions of convective precipitation to total precipitation over (a, d&g) (20°S,
- 547 20°N), (b, e&h) (20°N, 50°N) and (c, f&i) (50°N, 90°N). Total rainfall amounts are shown for
- 548 CAM5 (blue), STOC (red), GPCP (green) and TRMM (black) while convective (solid line) and
- 549 large-scale (dashed line) rainfall amounts and the fractional contributions of convective
- 550 precipitation are shown for CAM5 and STOC. The amount distributions (units: mm d⁻¹) are scaled
- 551 by $\Delta \ln(R) = \Delta R/R$, which has units of mm d⁻¹/mm d⁻¹ and is a unitless scaling term.
- 552 Figure 3. Frequency distributions of (a-c) total and (d-f) convective and large-scale precipitation,
- 553 over (a&d) (20°S, 20°N), (b&e) (20°N, 50°N) and (c&f) (50°N, 90°N). Total rainfall frequency
- distributions are shown for CAM5 (blue), STOC (red), GPCP (green) and TRMM (black) while
- 555 convective (solid line) and large-scale (dashed line) rainfall frequency distributions are shown for
- 556 CAM5 and STOC. The frequency distributions (units: %) are scaled by $\Delta \ln(R) = \Delta R/R$, which 557 has units of mm d⁻¹/mm d⁻¹ and is a unitless scaling term.
- 558 **Figure 4.** Amount distributions of wet removal of aerosols (units: mg/m²/day) over (20°S, 20°N)
- in CAM5 (blue), and STOC (red) runs. The distributions are scaled by $\Delta \ln(R) = \Delta R/R$, which
- 560 has units of mm d⁻¹/mm d⁻¹ and is a unitless scaling term. Numbers in each subplot are regional
- 561 mean wet deposition rates in two simulations. Note that the y-axis range for each frame is different.
- 562 **Figure 5.** Same as Figure 4, but over (20°N, 50°N).
- 563 **Figure 6.** Same as Figure 4, but over (50°N, 90°N).
- 564 Figure 7. Fractional contributions of wet removal of aerosols from convective clouds to the total
- amount of aerosol wet deposition over (20°S, 20°N) in CAM5 (blue), and STOC (red) runs. The
- distributions are scaled by $\Delta \ln(R) = \Delta R/R$, which has units of mm d⁻¹/mm d⁻¹ and is a unitless
- 567 scaling term.
- 568 **Figure 8.** Same as Figure 7, but over (20°N, 50°N).
- 569 **Figure 9.** Same as Figure 7, but over (50°N, 90°N).
- 570 Figure 10. Global distributions of the rainfall intensity associated with 50% of the accumulated
- 571 wet removal of aerosols for CAM5.
- 572 **Figure 11.** Same as Figure 10, but for STOC.





- 573 Figure 12. Annual and zonal mean cross-sections of changes in different aerosol mass
- 574 concentrations (µg/kg) between STOC and CAM5 runs (STOC CAM5). Areas exceeding 95%
- 575 t-test confidence level are stippled.
- 576 **Figure 13.** Annual and zonal mean cross-sections of changes in (a) mass flux from deep convection
- 577 and (b-c) vertical transport of POM and SOA aerosols by deep convection between STOC and
- 578 CAM5 runs (STOC CAM5). Areas exceeding 95% t-test confidence level are stippled.









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Figure 7. Fractional contributions of wet removal of aerosols from convective clouds to the total amount of aerosol wet deposition over (20°S, 20°N) in CAM5 (blue), and STOC (red) runs. The distributions are scaled by $\Delta \ln(R) = \Delta R/R$, which has units of mm d⁻¹/mm d⁻¹ and is a unitless scaling term.







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639







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