1 2 2	Examination of aerosol impacts on convective clouds and precipitation in two metropolitan areas in East Asia; how varying depths of convective clouds between
3 4	the areas diversity those aerosol effects?
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### 52 Abstract

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This study examines the role played by aerosols which act as cloud condensation nuclei (CCN) in the development of clouds and precipitation in two metropolitan areas in East Asia that have experienced substantial increases in aerosol concentrations over the last decades. These two areas are the Seoul and Beijing areas and the examination has been done by performing simulations using the Advanced Research Weather Research and Forecasting model as a cloud-system resolving model. CCN are advected from the continent to the Seoul area and this increases aerosol concentrations in the Seoul area. These increased CCN concentrations induce the enhancement of condensation that in turn induces the enhancement of deposition and precipitation amount in a system of less deep convective clouds as compared to those in the Beijing area. In a system of deeper clouds in the Beijing area, increasing CCN concentrations also enhance condensation but reduce deposition. This leads to CCN-induced negligible changes in precipitation amount. Also, in the system, there is a competition for convective energy among clouds with different condensation and updrafts. This competition results in different responses to increasing CCN concentrations among different types of precipitation, which are light, medium and heavy precipitation in the Beijing area. CCN-induced changes in freezing play a negligible role in CCN-precipitation interactions as compared to the role played by CCN-induced changes in condensation and deposition in both of the areas.

85 With increasing aerosol loading or concentrations, cloud-particle sizes can be changed. In 86 general, with increasing droplet sizes, the efficiency of collision and collection among 87 droplets increases. Increasing aerosol loading is known to make the droplet size smaller and thus make the efficiency of collision and collection among droplets lower. This leads 88 89 to less droplets or cloud liquid forming raindrops and there is more cloud liquid present in the air to be evaporated or frozen. Studies have shown that increases in cloud-liquid mass 90 91 due to increasing aerosol loading can enhance the freezing of cloud liquid and parcel 92 buoyancy, which lead to the invigoration of convection (Rosenfeld et al., 2008; Fan et al., 93 2009). Via the invigoration of convection, precipitation can be enhanced. The dependence 94 of aerosol-induced invigoration of convection and precipitation enhancement on aerosol-95 induced increases in condensational heating in the warm sector of a cloud system has been 96 shown (e.g., van den Heever et al., 2006; Fan et al., 2009; Lee et al., 2018). Increasing 97 cloud-liquid mass induces increasing evaporation, which intensifies gust fronts. This in 98 turn strengthens convective clouds and increases the amount of precipitation (Khain et al., 99 2005; Tao et al., 2007; Storer et al., 2010; Tao et al., 2012; Lee et al., 2017; Lee et al., 100 2018). It is notable that aerosol-induced precipitation enhancement is strongly sensitive to 101 cloud types that can be defined by cloud characteristics such as cloud depth (e.g., Tao et 102 al., 2007; Lee et al., 2008; Fan et al., 2009).

103 Since East Asia was industrialized, there have been substantial increases in aerosol 104 concentrations over the last decades in East Asia (e.g., Lee et al., 2013; Lu et al., 2011; Oh 105 et al., 2015; Dong et al., 2019). These increases are far greater than those in other regions 106 such as North America and Europe (e.g., Lu et al., 2011; Dong et al., 2019). While those 107 increasing aerosols affect clouds, precipitation and hydrologic circulations in the 108 continental East Asia, the increase in the advected aerosols from the continent to the 109 Korean Peninsula affect clouds, precipitation and hydrologic circulations in the Korean 110 Peninsula (Kar et al., 2009). This study aims to examine effects of the increasing aerosols, 111 which particularly act as cloud condensation nuclei (CCN), and their advection on clouds 112 and precipitation in East Asia. This study focuses on aerosols which act as CCN, but not ice-nucleating particles (INPs), to examine those effects, based on the fact that CCN 113

114 account for most of aerosol mass that affects clouds and precipitation, and CCN, but not 115 INPs, are associated with above-described aerosol-induced invigoration of convection and 116 intensification of gust fronts. Note that these aerosol-induced invigoration and 117 intensification are two well-established major theories of aerosol-cloud interactions. As a 118 first step to the examination, this study focuses on two metropolitan areas in East Asia which are the Beijing and Seoul areas. The population of each of the Beijing and Seoul 119 120 areas is  $\sim 20$  millions. Associated with this, these areas have lots of aerosol sources (e.g., 121 traffic) and have made a substantial contribution to the increases in aerosol concentrations 122 in East Asia. Hence, we believe that these two cities can represent overall situations related 123 to increasing aerosol concentrations in East Asia.

As mentioned above, aerosol-cloud interactions (and their impacts on precipitation) are strongly dependent on cloud types and thus to gain a more general understanding of those interactions, we select cases from the Beijing and Seoul areas with different cloud types. A selected case from the Beijing area involves deep convective clouds that reach the tropopause, while a selected case from the Seoul area involves comparatively shallow (or less deep) convective clouds. Via comparisons between these two cases, we aim to identify mechanisms that control varying aerosol-cloud interactions with cloud types.

To examine impacts of aerosols, which act as CCN, on clouds and precipitation in the cases, numerical simulations are performed, as a way of fulfilling above-described aim. These simulations use a cloud-system resolving model (CSRM) that has reasonably high resolutions to resolve cloud-scale processes that are related to cloud microphysics and dynamics. Hence, these simulations are able to find process-level mechanisms in association with cloud-scale processes.

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### 2. Case description

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In the Seoul area, South Korea, there is an observed mesoscale convective system (MCS) for a period from 03:00 LST (local solar time) to 18:00 LST December 24<sup>th</sup> 2017. During this period, there is a recorded moderate amount of precipitation and its maximum precipitation rate reaches  $\sim 13$  mm hr<sup>-1</sup>. Here, precipitation in the Seoul area is measured by rain gauges in automatic weather stations (AWSs) (King, 2009). The measurement is 145 performed hourly with a spatial resolution that ranges from  $\sim 1$  km to  $\sim 10$  km. The Seoul 146 area is marked by an inner rectangle in Figure 1a and dots in the rectangle in Figure 1a 147 mark the selected locations of rain gauges. At 21:00 LST December 23rd 2017, synopticscale features develop in favor of the formation and development of the selected MCS and 148 149 associated moderate rainfall. These features involve the southwesterly low-level jets that 150 transport warm and moist air to the Korean Peninsula. The southwesterly low-level jet plays an important role in the formation and development of rainfall events in the Korean 151 152 Peninsula by fetching warm and moist air (Hwang and Lee 1993; Lee et al. 1998; Seo et 153 al. 2013; Oh et al. 2018).

154 There was another observed MCS case in the Beijing area, China for a period from 14:00 LST on July 27th to 00:00 LST July 28th 2015. There is a substantial recorded amount 155 156 of precipitation for this period and its maximum precipitation rate reaches  $\sim 45$  mm hr<sup>-1</sup>. Here, similar to the situation in the Seoul area, precipitation in the Beijing area is measured 157 158 by rain gauges in AWSs hourly with a spatial resolution that ranges from  $\sim 1$  km to  $\sim 10$  km. 159 The Beijing area is marked by an inner rectangle in Figure 1b and dots in the rectangle in 160 Figure 1b mark the selected locations of rain gauges. At 09:00 LST July 27th 2015, 161 synoptic-scale features develop in favor of the formation and development of the selected 162 MCS. These features involve the southerly low-level jet that develops heavy rainfall events 163 in the Beijing area by transporting warm and moist air to the area. Synoptic features which 164 are described here are based on reanalysis data that are produced by the Met Office Unified Model (Brown et al., 2012) every 6 hours with a  $0.11^{\circ} \times 0.11^{\circ}$  resolution. 165

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### 3. CSRM and simulations

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### 169 **3.1 CSRM**

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The Advanced Research Weather Research and Forecasting (ARW) model (version 3.3.1)
is used as a CSRM. The ARW model is a compressible model with a nonhydrostatic status.
A 5th-order monotonic advection scheme is used to advect microphysical variables (Wang et al., 2009). The Rapid Radiation Transfer Model (RRTMG; Mlawer et al., 1997; Fouquart and Bonnel, 1980) is adopted to parameterize shortwave and longwave radiation in

simulations. A microphysics scheme that is used in this study calculates the effective sizes
of hydrometeors that are fed into the RRTMG, and the RRTMG simulates how these
effective sizes affect radiation.

179 The CSRM adopts a bin scheme as a way of parameterizing microphysics. The 180 Hebrew University Cloud Model (HUCM) detailed in Khain et al. (2011) is the bin scheme. A set of kinetic equations is solved by the bin scheme to represent a size distribution 181 182 function for each of seven classes of hydrometeors and aerosols acting as CCN. Hence, 183 there are seven size distribution functions for hydrometeors. The seven classes of 184 hydrometeors are water drops, three types of ice crystals, which are plates, columns and 185 dendrites, snow aggregates, graupel and hail. Drops whose radius is smaller (larger) than 186 40 µm are categorized to be droplets (raindrops). There are 33 bins for each size 187 distribution in a way that the mass of a particle  $m_i$  in the j bin is to be  $m_i = 2m_{i-1}$ .

188 A cloud-droplet nucleation parameterization based on Köhler theory represents cloud-189 droplet nucleation. Arbitrary aerosol mixing states and aerosol size distributions can be fed 190 this parameterization. To represent heterogeneous ice-crystal nucleation, to 191 parameterizations by Lohmann and Diehl (2006) and Möhler et al. (2006) are used. In these 192 parameterizations, contact, immersion, condensation-freezing, and deposition nucleation 193 paths are all considered by taking into account the size distribution of INPs, temperature freezing 194 supersaturation. Homogeneous and droplet is 195 considered following the theory developed by Koop et al. (2000).

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### 3.2 Control runs

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199 For a three-dimensional CSRM simulation of the observed case of convective clouds in the 200 Seoul (Beijing) area, i.e., the control-s (control-b) run, a domain just over the Seoul 201 (Beijing) area, which is shown in Figure 1a (1b), is used. This domain adopts a 300-m 202 resolution. The control-s run is for a period from 03:00 LST to18:00 LST December 24th 203 2017, while the control-b run is for a period from 14:00 LST on July 27th to 00:00 LST July 28<sup>th</sup> 2015. The length of the domain is 170 (140) km in the east-west (north-south) 204 205 direction for the control-s run, and 280 (240) km for the control-b run. There are 100 206 vertical layers and these layers employ a sigma coordinate that follows the terrain. The top 207 pressure of the model is 50 hPa for both of the control-s and control-b runs. On average,
208 the vertical resolution is ~200 m.

Reanalysis data, which are produced by the Met Office Unified Model (Brown et al.,
2012), represent the synoptic-scale features, provide initial and boundary conditions of
variables such as wind, potential temperature, and specific humidity for the simulations.
The simulations adopt an open lateral boundary condition. The Noah land surface model
(LSM; Chen and Dudhia, 2001) calculates surface heat fluxes.

214 The current version of the ARW model is not able to consider the spatiotemporal 215 variation of aerosol properties. In order to take into account the spatiotemporal variation of 216 aerosol properties, which is typical in metropolitan areas, such as composition and number 217 concentration, an aerosol preprocessor, which is able to consider the variability of aerosol 218 properties, is developed and used in the simulations. This aerosol preprocessor interpolates 219 or extrapolates background aerosol properties in observation data such as aerosol mass 220 (e.g.,  $PM_{2.5}$  and  $PM_{10}$ ) into grid points and time steps in the model. In this study, the inverse 221 distance weighting method is used for the extrapolation and interpolation of observation 222 data including aerosol mass into grid points and time steps in the model. PM stands for 223 particulate matter. The mass of aerosols with diameter smaller than  $2.5 (10.0) \mu m$  per unit 224 volume of the air is  $PM_{2.5}$  ( $PM_{10}$ ).

225 There are surface observation sites, which measure aerosol properties, in the domains 226 and these sites are classified into two types; the selected locations of these sites are marked 227 by dots in the inner rectangles in Figure 1. The distance between the observation sites 228 ranges from  $\sim 1$  km to  $\sim 10$  km and the time interval between observations is  $\sim 10$  minutes. 229 More than 90% of the sites belong to the first type of the sites. These first-type sites are 230 managed by the government in South Korea or China, and measure PM2.5 or PM10 but not 231 other aerosol properties such as aerosol composition and size distributions. Less than 10% 232 of the sites belong to the second type of the sites. These second-type sites are a part of 233 aerosol robotic network (AERONET; Holben et al., 2001) and measure aerosol 234 composition and size distributions. The production of aerosol data in these second-type or 235 AERONET sites is viable only in the presence of the sun. The first-type sites observe PM<sub>2.5</sub> 236 or PM<sub>10</sub> using the beta-ray attenuation method (Eun et al., 2016; Ha et al., 2019) and hence, produce  $PM_{2.5}$  or  $PM_{10}$  data whether the sun is present or not.  $PM_{2.5}/PM_{10}$  data from the 237

first-type sites are used to represent the spatiotemporal variability of aerosols over the domains and the simulation periods. To represent aerosol composition and size distributions, data from the AERONET sites are employed.

241 The AERONET data are averaged over the AERONET sites at 02:00 LST December 24th 2017 (13:00 LST July 27th 2015), which is 1 hour before the observed MCS forms, for 242 243 the Seoul (Beijing) case. Based on the average data, it is assumed that aerosol particles are 244 internally mixed with 70 (80) % ammonium sulfate and 30 (20) % organic compound for 245 the Seoul (Beijing) case. This mixture is assumed to represent aerosol chemical 246 composition in the whole domain and during the entire simulation period. Since ammonium 247 sulfate and organic compound are representative components of CCN, it is assumed that 248 PM<sub>2.5</sub> and PM<sub>10</sub>, which are from the first-type sites, represent the mass of aerosols that act 249 as CCN for the Seoul and Beijing areas, respectively. Aerosols reflect, scatter and absorb 250 shortwave and longwave radiation before they are activated. This type of aerosol-radiation 251 interactions is not taken into account in this study. This is mainly based on the fact that in 252 the mixture, there is insignificant amount of radiation absorbers; black carbon is a 253 representative radiation absorber. The average AERONET data indicate that the size 254 distribution of background aerosols acting as CCN follows the bi-modal log-normal 255 distribution for both of the Seoul and Beijing cases. Based on the average AERONET data, 256 it is assumed that for the whole domain and simulation period, the size distribution of 257 background aerosols acting as CCN follows a shape of distribution with specific size 258 distribution parameters (i.e., modal radius and standard deviation of each of accumulation 259 and coarse modes, and the partition of aerosol number among those modes) for each of the 260 cases. Modal radius of the shape of distribution is 0.110 (0.085) and 1.413 (1.523)  $\mu$ m, 261 while standard deviation of the shape of distribution is 1.54 (1.63) and 1.75 (1.73) for 262 accumulation and coarse modes, respectively, in the Seoul (Beijing) case. The partition of 263 aerosol number, which is normalized by the total aerosol number of the size distribution, 264 is 0.999 and 0.001 for accumulation and coarse modes, respectively, in both of the cases. 265 By using  $PM_{2.5}$  or  $PM_{10}$ , which is not only from the first-type sites but also interpolated 266 and extrapolated to grid points immediately above the surface and time steps, and based on 267 the assumption of aerosol composition and size distribution above, which is in turn based 268 on data from the AERONET sites, the background number concentrations of aerosols

269 acting as CCN are obtained for the simulation for each of the cases. There is no variation 270 with height in background concentrations of aerosols acting as CCN from immediately 271 above the surface to the top of the planetary boundary layer (PBL). However, it is assumed 272 that they decrease exponentially with height from the PBL top upward. With this 273 exponential decrease, when the altitude reaches the tropopause, background concentrations 274 of aerosols acting as CCN reduce by a factor of  $\sim 10$  as compared to those at the PBL top. 275 The size distribution and composition of aerosols acting as CCN do not vary with height. Once background aerosol properties (i.e., aerosol number concentrations, size distribution 276 277 and composition) are put into each grid point and time step, those properties at each grid 278 point and time step do not change during the course of the simulations.

For the control-s and control-b runs, aerosol properties of INPs are not different from those of CCN except for the fact that the concentration of background aerosols acting as CCN is 100 times higher than the concentration of background aerosols acting as INPs at each time step and grid point, following a general difference between CCN and INPs in terms of their concentrations (Pruppacher and Klett, 1978).

284 Once clouds form and background aerosols start to be in clouds, those aerosols are 285 not background aerosols anymore and the size distribution and concentrations of those 286 aerosols begin to evolve through aerosol sinks and sources that include advection and 287 aerosol activation (Fan et al., 2009). For example, once aerosols are activated, they are 288 removed from the corresponding bins of the aerosol spectra. In clouds, after aerosol 289 activation, aerosol mass starts to be inside hydrometeors and via collision-collection, it 290 transfers to different types and sizes of hydrometeors. In the end, aerosol mass disappears 291 in the atmosphere when hydrometeors with aerosol mass touches the surface. In non-cloudy 292 areas, aerosol size and spatial distributions are designed to be identical to the size and 293 spatial distributions of background aerosols, respectively. In other words, for this study, 294 we use "the aerosol recovery method". In this method, at any grid points, immediately after 295 clouds disappear entirely, aerosol size distributions and number concentrations recover to 296 background properties that background aerosols at those points have before those points 297 are included in clouds. In this way, we can keep concentrations of background aerosols 298 outside clouds in the simulations at observed counterparts. This enables spatiotemporal 299 distributions of background aerosols in the simulations to mimic those distributions that

300 are observed and particularly associated with observed aerosol advection in reality. In the 301 aerosol recovery method, there is no time interval between the cloud disappearance and the 302 aerosol recovery. Here, when the sum of mass of all types of hydrometeors (i.e., water 303 drops, ice crystals, snow aggregates, graupel and hail) is not zero at a grid point, that grid 304 point is considered to be in clouds. When this sum becomes zero, clouds are considered to 305 disappear. Many studies using CSRM have employed this aerosol recovery method. They 306 have proven that with the recovery method, reasonable simulations of overall cloud and 307 precipitation properties are accomplished (e.g., Morrison and Grabowski, 2011; Lebo and 308 Morrison, 2014; Lee et al., 2016; Lee et al., 2018).

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### 3.3 Additional runs

312 We repeat the control-s run by getting rid of aerosol-advection induced increases in 313 concentrations of aerosols acting as CCN as a way of investigating how the aerosol 314 advection affects the cloud system in the Seoul area. This repeated run is named the low-315 aerosol-s run. An aerosol layer, which is advected from East Asia or from the west of the 316 Seoul area to it, increases aerosol concentrations in the Seoul area. There are stations in 317 islands in the Yellow Sea that monitor the aerosol advection (Eun et al., 2016; Ha et al., 2019). To monitor and identify the aerosol advection, PM<sub>2.5</sub> which is measured by a station 318 319 in Baekryongdo island in Yellow Sea are compared to those which are measured in stations 320 in and around the Seoul area. In Figure 1a, a dot outside the inner rectangle marks the 321 island. The time evolution of PM<sub>2.5</sub> measured by the station on the island and the average PM<sub>25</sub> over stations in the Seoul area, between 07:00 LST on December 22<sup>nd</sup> and 21:00 LST 322 323 on December 24<sup>th</sup> in 2017 when there is the strong advection of aerosols from East Asia to the Seoul area, is shown in Figure 2. At 09:00 LST on December 22<sup>nd</sup>, the advection of 324 aerosols from East Asia enables aerosol mass to start going up and attain its peak around 325 05:00 LST on December 23<sup>rd</sup> on the island. Following this, aerosol mass starts to increase 326 327 in the Seoul area around 01:00 LST on December 23<sup>rd</sup>, and the mass attains its peak at 15:00 LST on December 23rd in the Seoul area. This is because aerosols, which are 328 329 advected from East Asia, move through the island to reach the Seoul area.

330 In the low-aerosol-s run, as a way of getting rid of aerosol-advection induced increases 331 in concentrations of aerosols acting as CCN, it is assumed that PM<sub>2.5</sub>, which is assumed to 332 represent the mass of aerosols acting as CCN, and the associated background concentration 333 of aerosols acting as CCN after 01:00 LST on December 23<sup>rd</sup> do not evolve with the aerosol 334 advection in the Seoul area. Hence, the background concentration of aerosols acting as CCN is assumed to have that at 01:00 LST on December 23<sup>rd</sup> at each time step and grid 335 point at the beginning of the simulation period. However, to isolate CCN effects on clouds, 336 337 background aerosol concentration acting as INPs at each time step and grid point in the 338 low-aerosol-s run is not different from that in the control-s run during the simulation period. 339 In the observed PM data for the Seoul area, there is reduction in PM by a factor of  $\sim 10$  on average over a period between ~07:00 and ~14:00 LST on December 24th, since 340 341 precipitation scavenges aerosols (Figure 2). To emulate this scavenging and reflect it in 342 background aerosols acting as CCN for the low-aerosol-s run, PM<sub>2.5</sub> and corresponding 343 background concentrations of aerosols acting as CCN at each grid point is gradually 344 reduced for the period between 07:00 and 14:00 LST on December 24<sup>th</sup>. This reduction is 345 done in a way that background concentrations of aerosols acting as CCN at each grid point at 14:00 LST on December 24th is 10 times lower than that at 07:00 LST on December 24th 346 in the low-aerosol-s run. Then, PM2.5 and corresponding background concentrations of 347 aerosols acting as CCN at each grid point at 14:00 LST on December 24th maintains until 348 349 the end of the simulation period. This results in the evolution of the average PM<sub>2.5</sub> over the 350 Seoul area in the low-aerosol-s run as shown in Figure 2. Here, the concentration of 351 background aerosols acting as CCN, which is averaged over the whole domain and 352 simulation period, in the control-s run is 3.1 times higher than that in the low-aerosol-s run. 353 Via comparisons between the runs, how the increasing concentration of background 354 aerosols acting as CCN due to the aerosol advection has an impact on clouds can be 355 examined. The concentration of background aerosols acting as CCN is different among 356 grid points and time steps in the control-s run. Hence, the ratio of the concentration of 357 background aerosols acting as CCN between the runs is different among grid points and 358 time steps.

For the Beijing case, to examine how aerosols acting as CCN affect clouds and precipitation, we repeat the control-b run with simply reduced concentrations of 361 background aerosols acting as CCN at each time step and grid point by a factor of 3.1. This 362 repeated run is named the low-aerosol-b run. The 3.1-fold increase in aerosol 363 concentrations from the low-aerosol-b run to the control-b is based on the 3.1-fold increase 364 in the average concentration of background aerosols acting as CCN from the low-aerosol-365 s run to the control-s run. However, as in the control-s and low-aerosol-s runs, to isolate 366 CCN effects on clouds, background aerosol concentration acting as INPs at each time step 367 and grid point in the low-aerosol-b run is identical to that in the control-b run during the 368 simulation period. Hence, on average, a pair of the control-s and low-aerosol-s runs has the same perturbation of aerosols acting as CCN as in a pair of the control-b and low-aerosol-369 370 b runs. Here, we define aerosol perturbation as a relative increase in aerosol concentration 371 when compared to that before the increase occurs. The brief summary of all simulations in 372 this study is given in Table 1.

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### 4.1 Cumulative precipitation

4. Results

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We compare the observed precipitation to the simulated counterpart in the control-s run for the Seoul case and in the control-b run for the Beijing case. For this comparison, the observed and simulated precipitation rates at the surface are averaged over the domain for each of the Seoul and Beijing cases (Figures 3a and 3b). Here, the simulated precipitation rates are smoothed over 1 hour. The comparison shows that the evolution of the simulated precipitation rate does not deviate from the observed counterpart significantly (Figures 3a and 3b).

In the Seoul case, overall, the precipitation rate is higher in the control-s run than in the low-aerosol-s run. As a result of this, the domain-averaged cumulative precipitation amount at the last time step is 14.1 mm and 12.0 mm in the control-s run and the lowaerosol-s run, respectively. The control-s run shows  $\sim 20$  % higher cumulative precipitation amount. In the Beijing case, the evolution of the mean precipitation rate in the control-b run is not significantly different from that in the low-aerosol-b run. Due to this, the controlb run shows only  $\sim 2$  % higher cumulative precipitation amount, despite the fact that the 392 concentrations of background aerosols acting as CCN are ~3 times higher in the control-b 393 run than in the low-aerosol-b run. Note that in the Seoul case, the time- and domain-394 averaged concentration of background aerosols acting as CCN is also ~3 times higher in 395 the control-s run than in the low-aerosol-s run. Despite this, the difference in the cumulative 396 precipitation amount between the runs with different concentrations of background 397 aerosols acting as CCN is greater in the Seoul case than in the Beijing case.

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### 4.2 Precipitation, and associated latent-heat and dynamic processes

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401 Figures 4a and 4b show the cumulative frequency distributions of precipitation rates at the 402 last time step in the simulations for the Seoul and Beijing cases, respectively. In each of 403 those figures, the observed frequency distribution is shown and compared to the simulated 404 distribution. The observed distribution is obtained by interpolating and extrapolating the 405 observed precipitation rates to grid points and time steps in each of the control-s and 406 control-b runs. The observed maximum precipitation rates are 13.0 and 44.5 mm hr<sup>-1</sup> for 407 the Seoul and Beijing cases, respectively, and these maximum rates are similar to those in 408 the control-s and control-b runs, respectively. Overall, the observed and simulated 409 frequency distributions are in good agreement for each of the cases. This enables us to 410 assume that results in the control-s (control-b) run are benchmark results to which results 411 in the low-aerosol-s (low-aerosol-b) run can be compared to identify how aerosols acting 412 as CCN have an impact on clouds and precipitation for the Seoul (Beijing) case. Here, it is 413 notable that for the Beijing case, while differences in the cumulative precipitation amount 414 between the control-b and low-aerosol-b runs are not significant, features in the frequency 415 distribution of precipitation rates between those runs are substantially different (Figure 4b).

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### 1) Seoul case

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### a. Precipitation Frequency distributions

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421 Regarding precipitation whose rates are higher than  $\sim 2 \text{ mm hr}^{-1}$ , the cumulative 422 precipitation frequency at the last time step is higher in the control-s run as compared to 423 that in the low-aerosol-s run (Figure 4a). In particular, for the precipitation rate of 11.4 mm 424  $h^{-1}$ , there is an increase in the cumulative frequency by a factor of as much as ~10 in the 425 control-s run. When it comes to precipitation rates above 11.5 mm hr<sup>-1</sup>, precipitation is 426 present in the control-s run and precipitation is absent in the low-aerosol-s run. Regarding 427 precipitation whose rates are lower than  $\sim 2$  mm hr<sup>-1</sup>, differences in the cumulative frequency between the runs are insignificant. Hence, we see that there are significant 428 429 increases in the frequency of relatively heavy precipitation whose rates are above  $\sim 2 \text{ mm}$ 430 hr<sup>-1</sup> in the control-s run when compared to that in the low-aerosol-s run. At the last time step, this results in a larger amount of cumulative precipitation in the control-s run than in 431 432 the low-aerosol-s run.

433 The time evolution of the cumulative precipitation frequency is shown in Figure 5. At 06:00 LST December 24<sup>th</sup> 2017, which corresponds to the initial stage of the precipitation 434 development, the maximum precipitation rate reaches  $\sim 3 \text{ mm hr}^{-1}$  and there is the greater 435 436 frequency over most of precipitation rates in the control-s run than in the low-aerosol-s run 437 (Figure 5a). With the time progress from 06:00 LST to 10:00 LST, the maximum precipitation rate increases to reach 12 mm hr<sup>-1</sup> and the cumulative frequency is higher over 438 precipitation whose rates are higher than ~3 mm hr<sup>-1</sup> in the control-s run, while for 439 440 precipitation whose rates are lower than ~3 mm hr<sup>-1</sup>, differences in the cumulative 441 frequency between the runs are negligible (Figures 5a and 5b). When time reaches 12:00 442 LST, which is around time when the peak in the evolution of the area-averaged 443 precipitation rates occurs and thus the system is at its mature stage, the maximum 444 precipitation rate increases up to  $\sim 13$  mm hr<sup>-1</sup> (Figures 3a and 5c). The basic patterns of 445 differences in the cumulative precipitation frequency between the runs with the maximum 446 precipitation rate around 13 mm hr<sup>-1</sup>, which are established at 12:00 LST, maintain until 447 the end of the simulation period (Figures 4a and 5c).

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### b. Condensation, deposition, updrafts and associated variables

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451 Note that the source of precipitation is precipitable hydrometeors which are raindrops, 452 snow, graupel and hail particles. Droplets and ice crystals are the source of those 453 precipitable hydrometeors mostly via collision and coalescence processes. Droplets and ice 454 crystals gain their mass mostly via condensation and deposition. Based on this, to explain 455 the greater cumulative precipitation amount in the control-s run than in the low-aerosol-s 456 run, the evolutions of differences in condensation, deposition and associated updrafts 457 between the runs are analyzed. The vertical profiles of differences in the area-averaged 458 condensation, deposition and freezing rates, updraft mass fluxes and the associated mass 459 density of each class of hydrometeors between the runs at 03:20, 03:40, 06:00 and 12:00 460 LST are shown in Figure 6. In Figure 6, differences in freezing rates are added for a more 461 comprehensive understanding of processes that are related to differences in cumulative 462 precipitation amount between the runs. Freezing includes riming processes between liquid 463 and solid hydrometeors and these riming processes act as a source of precipitable 464 hydrometeors. Cloud fractions are 0.32 (0.30), 0.85 (0.82), 0.93 (0.92) and 1.00 (1.00) in 465 the control-s (low-aerosol-s) run at 03:20, 03:40, 06:00 and 12:00 LST, respectively. We 466 see that cloud fraction varies 0-~6% between the runs. Note that in all of figures, which 467 display snow and hail mass density and include Figure 6, snow mass density includes ice-468 crystal mass density, while hail mass density includes graupel mass density for the sake of 469 the display brevity. In Figure 6, horizontal black lines represent the altitudes of freezing 470 and melting.

471 Condensation rates in the control-s run start to be larger than that in the low-aerosol-472 s run at 03:20 LST (Figure 6a). Higher aerosol or CCN concentrations induce more 473 nucleation of droplets, higher cloud droplet number concentration (CDNC) and associated 474 greater integrated surface of droplets in the control-s run. CDNC, which is averaged over 475 grid points and time steps with non-zero CDNC, is 1050 and 352 cm<sup>-3</sup> in the control-s and 476 low-aerosol-s runs, respectively. Hence, more droplet surface is provided for water vapor 477 to condense onto in the control-s run. This leads to more condensation in the control-s run. 478 This establishes stronger feedbacks between updrafts and condensation, leading to greater 479 droplet (or cloud-liquid) mass at 03:20 LST in the control-s run (Figure 6a). Then, these 480 stronger feedbacks, which involve stronger updrafts particularly above 2 km in altitude, 481 subsequently induce greater deposition and snow mass as time progresses from 03:20 LST 482 to 03:40 LST, while more condensation and greater droplet mass maintain in the control-s 483 run with the time progress to 03:40 LST (Figure 6b). These stronger updrafts enable clouds 484 to grow higher in the control-s run. This eventually leads to a situation where the maximum

485 cloud depth is  $\sim$ 7 km in the control-s run and this depth is  $\sim$ 5 % deeper than that in the low-486 aerosol-s run for the whole simulation period.

487 Through aerosol-induced stronger feedbacks between condensation, deposition and 488 updrafts in the control-s run, while more condensation and more overall deposition 489 maintain in the control-s run, differences in condensation and deposition between the 490 control-s and low-aerosol-s runs increase as time progresses from 03:40 LST to 06:00 LST 491 (Figures 6b and 6c). Associated with this, the greater mass of raindrops and hail particles 492 appears up, while the greater mass of droplets and snow in the control-s run than in the 493 low-aerosol-s run maintains with the time progress from 03:40 LST to 06:00 LST (Figure 494 6c). At 06:00 LST, there is more freezing starting to occur in the control-s run than in the 495 low-aerosol-s run. However, differences in freezing are ~one and ~two orders of magnitude 496 smaller than those in deposition and condensation, respectively. After 06:00 LST until time 497 reaches 12:00 LST when the overall differences in the cumulative precipitation frequency 498 between the runs are established, differences in freezing become  $\sim 3$  times smaller than 499 those in deposition and ~one order of magnitude smaller than those in condensation 500 (Figures 6c and 6d). The greater mass of hydrometeors in the control-s run also continues 501 after 06:00 LST until time reaches 12:00 LST (Figures 6c and 6d). At 12:00 LST, 502 condensation, deposition and freezing rates are still higher in the control-s run. Here, we 503 see that CCN-induced more cumulative precipitation amount and associated differences in 504 the precipitation frequency distribution between the control-s and low-aerosol-s runs are 505 primarily associated with CCN-induced more condensation, which induce CCN-induced 506 more deposition and higher mass density of hydrometeors as sources of precipitation, but 507 weakly connected to CCN-induced changes in freezing. This is supported by the fact that 508 the time- and domain-averaged differences in freezing rate are ~one to ~two order of 509 magnitude smaller than those in condensation and deposition rates.

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## c. Condensation frequency distributions and horizontal distributions of condensation and precipitation

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514 Based on the importance of condensation for CCN-induced changes in precipitation, the 515 horizontal distribution of the column-averaged condensation rates over the domain and the 516 cumulative frequency distribution of the column-averaged condensation rates at each time 517 step is obtained. To better visualize the role of condensation in precipitation, the horizontal 518 distribution of the column-averaged condensation rates is superimposed on that of 519 precipitation rates (Figure 7). At 03:40 LST, condensation mainly occurs around the 520 northern part of the domain as marked by a yellow rectangle. The synoptic wind condition 521 in the marked area favors the collision between northward and southward wind and the 522 associated convergence around the surface (Figures 7a and 7b). This convergence induces 523 updrafts and condensation in the marked area. In the marked area, more aerosols acting as 524 CCN induce more and more extensive condensation, which leads to the higher domain-525 averaged condensation rates in the control-s run than in the low-aerosol-s run (Figures 6b, 526 7a and 7b). More droplets are formed on more aerosols acting as CCN and more droplets 527 provide more surface areas where condensation occurs and this enables more and more 528 extensive condensation in the control-s run than in the low-aerosol-s run (Figures 6b, 7a 529 and 7b).

530 At 06:20 LST, a precipitating system is advected into the domain via the western 531 boundary, and as seen in Figures 7c and 7d for 08:40 LST, as time progresses to 08:40 LST, 532 the advected precipitating system is further advected to the east and extended mostly over 533 areas in the northern part of the domain where condensation mainly occurs. This confirms 534 that condensation is the main source of cloud mass and precipitation. In the eastern part of 535 the domain, there are mountains and in particular, higher mountains are on the northeastern 536 part of the domain than in the other parts of the domain. These higher mountains induce 537 forced convection and associated condensation more effectively in the northeastern part 538 than in the other parts. This is in favor of the precipitating system that extends further to 539 the east in the northern part of the domain. Due to more aerosols acting as CCN, 540 condensation, which is induced by forced convection over mountains, is more and more 541 extensive in the control-s run (Figures 7c and 7d). In association with this, there is more 542 extension of the precipitating system in the control-s run than in the low-aerosol-s run. This 543 enables the system in the control-s run to reach the eastern boundary at 08:40 LST, which 544 is earlier than in the low-aerosol-s run (Figures 7c and 7d). The system in the low-aerosol-545 s run reaches the eastern boundary at 09:00 LST. Here, we see that although aerosols acting 546 as CCN do not change overall locations of the precipitation system, they affect how fast

the system extends to the east by affecting the amount of condensation which is produced by forced convection. Associated with this, as seen in Figure 8, the control-s run has the much higher cumulative condensation frequency than the low-aerosol-s run over all of condensation rates during the period between 07:20 and 09:00 LST. Contributed by this, the higher precipitation frequency over most of precipitation rates occurs in the control-s run during and after the period (Supplementary Figures 1a and 1b and Figures 5b and 5c).

553 At 10:00 LST, in the southern part of the domain, there is a precipitating area forming 554 as marked by a yellow rectangle (Figures 7e and 7f). The precipitation area in the southern 555 part of the domain extends and merges into the advecting main precipitating system in the 556 northern part of the domain. The merge leads to precipitation that occupies most of the 557 domain at 12:00 LST (Figures 7g and 7h). After 10:00 LST, associated with this merge, the maximum precipitation rate increases to 13 mm hr<sup>-1</sup> at 12:00 LST (Figures 5c). After 558 559 13:00 LST, the precipitation enters its dissipating stage and its area reduces and nearly 560 disappears. Even after the merge, CCN-induced more condensation maintains and this in 561 turn contributes to a situation where the control-s run has the greater precipitation 562 frequency over most of precipitation rates than in the low-aerosol-s run until the 563 simulations progress to their last time step (Figures 4a, 5c and 6d).

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2) Beijing case

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567 Stronger convection and deeper clouds develop in the Beijing case than in the Seoul case. 568 The maximum cloud depth is  $\sim$ 7 and  $\sim$ 12 km in the control-s and control-b runs, 569 respectively. In the Seoul case, clouds do not reach the tropopause, while they reach the 570 tropopause in the Beijing case. Deeper clouds in the Beijing case produce the maximum 571 precipitation rate of  $\sim$ 45 mm hr<sup>-1</sup> in the control-b run. However, less deep clouds in the 572 Seoul case produce the maximum precipitation rate of  $\sim$ 13 mm hr<sup>-1</sup> in the control-s run 573 (Figure 4).

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### a. Precipitation frequency distributions

When it comes to precipitation whose rates are higher than  $\sim 12$  mm hr<sup>-1</sup>, the control-b run 577 578 has the higher cumulative precipitation frequency at the last time step than the low-aerosol-579 b run (Figure 4b). Particularly, for the precipitation rates of 28.1 and 30.0 mm hr<sup>-1</sup>, the 580 cumulative frequency increases by a factor of as much as ~10. Moreover, regarding precipitation rates higher than ~33 mm hr<sup>-1</sup>, precipitation is present in the control-b run, 581 582 however, precipitation is absent in the low-aerosol-b run. Hence, we see that the frequency of comparatively heavy precipitation whose rates are higher than  $\sim 12$  mm hr<sup>-1</sup> rises 583 significantly in the control-b run as compared to that in the low-aerosol-b run. Below  $\sim 2$ 584 mm hr<sup>-1</sup>, there is also the greater precipitation frequency in the control-b run than in the 585 586 low-aerosol-b run. Unlike the situation for precipitation rates above ~12 mm hr<sup>-1</sup> and below 587  $\sim 2 \text{ mm hr}^{-1}$ , for precipitation rates from  $\sim 2 \text{ mm hr}^{-1}$  to  $\sim 12 \text{ mm hr}^{-1}$ , the control-aerosol-b 588 run has the lower precipitation frequency than in the low-aerosol-b run. Here, we see that the higher precipitation frequency above  $\sim 12 \text{ mm hr}^{-1}$  and below  $\sim 2 \text{ mm hr}^{-1}$  balances out 589 590 the lower precipitation frequency between  $\sim 2$  and  $\sim 12$  mm hr<sup>-1</sup> in the control-b run. This 591 results in the similar cumulative precipitation amount between the runs.

592 Figure 9 shows the time evolution of the cumulative precipitation frequency. When 593 precipitation starts around 16:00 LST, the higher precipitation frequency occurs over most 594 of precipitation rates in the low-aerosol-run-b run than in the control-b run (Figure 9a). At 16:00 LST, the maximum precipitation rate is lower than 1.0 mm hr<sup>-1</sup> for both of the runs. 595 596 As time progresses to 17:00 LST, the maximum precipitation rate increases to ~17 mm hr<sup>-</sup> <sup>1</sup> and the higher (lower) cumulative precipitation frequency over precipitation rates higher 597 598 than  $\sim 12 \text{ mm hr}^{-1}$  (between  $\sim 2$  and  $\sim 12 \text{ mm hr}^{-1}$ ) in the control-b run than in the low-599 aerosol-b run, which is described above as shown in Figure 4b for the last time step, starts 600 to emerge (Figure 9b). At 17:20 LST, the higher frequency for precipitation rates below 2 mm hr<sup>-1</sup> in the control-b run, which is also described above as shown in Figure 4b for the 601 602 last time step, starts to show up, while the higher (lower) frequency for precipitation rates higher than  $\sim 12$  mm hr<sup>-1</sup> (between  $\sim 2$  and  $\sim 12$  mm hr<sup>-1</sup>) in the control-b run, which is 603 604 established at 17:00 LST, maintains as time progresses from 17:00 LST to 17:20 LST (Figure 9c). At 17:20 LST, the maximum precipitation rate increases to 42 (19) mm hr<sup>-1</sup> in 605 606 the control-b (low-aerosol-b) run (Figure 9c). At 19:00 LST, the maximum precipitation rate increases to ~45 (33) mm hr<sup>-1</sup> for the control-b (low-aerosol-b) run, while the 607

qualitative nature of differences in the precipitation frequency distributions with the tipping precipitation rates of  $\sim 2$  and  $\sim 12$  mm hr<sup>-1</sup> between the runs does not vary much between 17:20 and 19:00 LST (Figures 9c and 9d). The qualitative nature of differences in the cumulative precipitation frequency between the runs and the maximum precipitation rates in each of the runs, which are established at 19:00 LST, do not vary significantly until the end of the simulation period (Figures 4b and 9d).

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### b. Condensation, deposition, updrafts and associated variables

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617 As done for the Seoul case, as a way of better understanding differences in the cumulative 618 precipitation amount and frequency between the control-b and low-aerosol-b runs, the 619 evolutions of differences in the vertical distributions of the area-averaged condensation 620 rates, deposition rates, freezing rates, the mass density of each class of hydrometeors and 621 updrafts mass fluxes are obtained and shown in Figures 10. Cloud fractions are 0.12 (0.11), 622 0.25 (0.22), 0.36 (0.32), 0.43 (0.40) and 0.48 (0.47) in the control-b (low-aerosol-b) run at 623 14:20, 15:40, 16:00, 17:20 and 19:00 LST, respectively. Here, we see that cloud fraction 624 varies by ~2-12% between the runs. In Figure 10, horizontal black lines represent the 625 altitudes of freezing and melting. As seen in Figure 3b, precipitation starts around 16:00 626 LST but differences in condensation rates start at 14:20 LST with higher condensation rates 627 in the control-b run (Figure 10a). Similar to the situation in the Seoul case, higher 628 concentrations of aerosols acting as CCN induce more nucleation of droplets, higher 629 CDNC and associated greater integrated surface of droplets in the control-b run. CDNC, 630 which is averaged over grid points and time steps with non-zero CDNC, is 992 and 341 631 cm<sup>-3</sup> in the control-b and low-aerosol-b runs, respectively. Hence, more droplet surface is 632 provided for water vapor to condense onto in the control-b run. This leads to more 633 condensation in the control-b run. Due to this, cloud-liquid or droplet mass becomes greater 634 in the control-b run at 14:20 LST (Figure 10a). Increased condensation rates induce 635 increased condensational heating and thus intensified updrafts (Figure 10a). These 636 updrafts enable the maximum cloud depth to be ~ 12 km in the control-b run and this depth 637 is just  $\sim 1$  % deeper than that in the low-aerosol-b run for the whole simulation period. This negligible difference in the maximum cloud depth between the runs is due to the fact that 638

639 640 clouds with the maximum depth reach the tropopause in both of runs and thus there is not much wiggle room to make significant differences in cloud depth between the runs.

641 When time reaches 15:40 LST, deposition rates and snow mass start to show 642 differences between the runs, while higher condensation rates and droplet mass maintain 643 in the control-b run with the time progress from 14:20 LST to 15:40 LST. However, unlike 644 the situation in the Seoul case, higher concentrations of aerosols acting as CCN result in 645 lower deposition rates and snow mass in the control-b run (Figure 10b). When time 646 progresses from 15:40 LST to 16:00 LST, differences in freezing start to occur and freezing 647 rates are lower (higher) at altitudes between ~6 and ~8 km (~4 and ~6 km), while higher condensation rates and droplet mass, and lower snow mass maintain in the control-b run 648 649 (Figure 10c). Due to stronger updrafts, which are mainly ascribed to more condensation, 650 deposition rates start to be higher at altitudes between ~7 and ~9 km and freezing rates are 651 higher at altitudes between  $\sim 4$  and  $\sim 6$  km in the control-b run with the time progress from 652 15:40 LST to 16:00 LST (Figure 10c). Differences in freezing rates are similar to those in 653 deposition and ~two orders of magnitude smaller than those in condensation at 16:00 LST 654 (Figure 10c). At 16:00 LST, differences in hail mass between the runs appear up and hail 655 mass is slightly lower in the control-b run (Figure 10c). At 17:20 LST, overall, freezing 656 rates are lower at altitudes between ~4 and ~8 km, while overall, snow and hail mass is still 657 lower, and droplet mass is still higher in the control-b run (Figure 10d). Differences in 658 freezing rates are ~2 times smaller than those in deposition and ~one order of magnitude smaller than those in condensation at 17:20 LST (Figure 10d). Due to more condensation 659 660 and droplet mass, greater raindrop mass appears up in the control-b run at 17:20 LST 661 (Figure 10d). As the time progresses to 19:00 LST, deposition rates become lower at the 662 altitudes from ~7 km to ~12 km and overall freezing rates become higher at altitudes from 663  $\sim$  4 km to  $\sim$ 10 km in the control-b run (Figure 10e). Overall, lower snow and hail mass 664 maintains in the control-b run as time progresses from 17:20 LST to 19:00 LST. As time 665 progresses from 17:20 LST to 19:00 LST, overall higher condensation rates, droplet and 666 raindrop mass maintain in the control-b run (Figure 10e). Here, while the time- and 667 domain-averaged deposition (condensation and freezing) rates are lower (higher) in the 668 control-b run over the whole simulation period, the average differences in freezing rates 669 are ~one to ~two orders of magnitude smaller than those in deposition and condensation

670 rates between the runs. Hence, more condensation (but not deposition and freezing) is a 671 main cause of stronger updrafts in the control-b run. More condensation and more freezing 672 tend to induce increases in the mass of precipitable hydrometeors in the control-b run. Less 673 deposition tends to induce decreases in the mass of precipitable hydrometeors in the 674 control-b run. This competition between condensation, deposition and freezing leads to negligible differences in the cumulative precipitation amount at the last time step between 675 676 the control-b and low-aerosol-b runs, although roles of freezing in this competition are 677 negligible as compared to those of condensation and deposition.

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- condensation and precipitation, and condensation-precipitation correlations

c. Condensation frequency distributions, horizontal distributions of

683 Figure 11 shows the horizontal distribution of the column-averaged condensation rates over 684 the domain and Figure 12 shows the cumulative frequency distributions of column-685 averaged condensation rates at selected times. As in the Seoul case, the horizontal 686 distribution of condensation rates is superimposed on that of precipitation rates and the 687 terrain in Figure 11. At 14:20 LST, condensation starts to occur in places with mountains, 688 which induce forced convection, and condensation is concentrated around the center of the 689 domain as marked by a yellow circle (Figures 11a and 11b). Note that condensation does 690 not occur in the plain area which is the south of the 100-m terrain-height contour line 691 (Figures 11a and 11b). Due to higher concentrations of aerosols acting as CCN, there is 692 more condensation around the center in the control-b run than in the low-aerosol-b run 693 (Figures 11a and 11b). This leads to a situation where the control-b run has the higher area-694 averaged condensation rates than the low-aerosol-b run (Figure 10a). Then, as time 695 progresses to 17:20 LST, the condensation area extends to the eastern and western parts of 696 the domain mostly over mountain areas (Figures 11c and 11d). Hence, the main source of 697 condensation is considered to be forced convection over mountains. As seen in Figures 11c 698 and 11d, higher concentrations of aerosols acting as CCN induce the control-b run to have 699 much more condensation spots and thus much bigger areas with condensation than the low-700 aerosol-b run at 17:20 LST. Associated with this, CCN-induced more condensation in the

- control-b run maintains with the time progress to 17:20 LST (Figure 10d). At 17:20 LST,
  precipitation mainly occurs in a spot which is in the western part of areas with relatively
  high condensation rates (Figures 11c and 11d).
- 704 At 17:20 LST, as seen in the cumulative frequency of condensation rates, the controlb run has the higher condensation frequency above condensation rate of  $\sim 10 \times 10^{-3}$  g m<sup>-3</sup> s<sup>-</sup> 705 <sup>1</sup> and below that of  $\sim 3 \times 10^{-3}$  g m<sup>-3</sup> s<sup>-1</sup> than the low-aerosol-b run (Figure 12a). This pattern 706 707 of differences in the condensation frequency distribution with the tipping condensation-708 rate points at  $\sim 10 \times 10^{-3}$  and  $\sim 3 \times 10^{-3}$  g m<sup>-2</sup> s<sup>-1</sup> continues up to 19:00 LST (Figures 12b). Figure 13 shows the mean precipitation rate over each of the column-averaged 709 710 condensation rates for the period up to 17:20 LST in the control-b run. A column-averaged 711 condensation rate in an air column with a precipitation rate at its surface is obtained and 712 these condensation and precipitation rates are paired at each column and time step. Then, 713 collected precipitation rates are classified and grouped based on the corresponding paired 714 column-averaged condensation rates. The classified precipitation rates corresponding to 715 each of the column-averaged condensation rates are averaged arithmetically to construct 716 Figure 13. There are only less than 10 % differences in the mean precipitation rate for each 717 of the column-averaged condensation rates between the control-b and low-aerosol-b runs 718 (not shown). Figure 13 shows that generally a higher condensation rate is related to a higher 719 mean precipitation rate. It is also roughly shown that, according to the mean precipitation rate for each condensation rate, overall, condensation rates below  $\sim 3 \times 10^{-3}$  g m<sup>-3</sup> s<sup>-1</sup> and 720 above  $\sim 10 \times 10^{-3}$  g m<sup>-3</sup> s<sup>-1</sup> are correlated with precipitation rates below  $\sim 2$  mm hr<sup>-1</sup> and 721 above ~12 mm hr<sup>-1</sup>, respectively, while condensation rates between ~3 and ~ $10 \times 10^{-3}$  g 722 m<sup>-3</sup> s<sup>-1</sup> are correlated with precipitation rates between  $\sim 2$  and  $\sim 12$  mm hr<sup>-1</sup> (Figure 13). 723 724 Hence, on average, the higher frequency of condensation with rates above  $\sim 10 \times 10^{-3}$  g m<sup>-</sup>  $^{3}$  s<sup>-1</sup> and below ~ 3 × 10<sup>-3</sup> g m<sup>-3</sup> s<sup>-1</sup> can be considered to lead to the higher frequency of 725 precipitation whose rates are higher than  $\sim 12$  mm hr<sup>-1</sup> and lower than  $\sim 2$  mm hr<sup>-1</sup> in the 726 727 control-b run, respectively. It can also be considered that the lower condensation frequency between  $\sim 3$  and  $\sim 10 \times 10^{-3}$  g m<sup>-3</sup> s<sup>-1</sup> leads to the lower precipitation frequency between  $\sim 2$ 728 and ~12 mm hr<sup>-1</sup> in the control-b run. It is found that this correspondence between 729 730 condensation and precipitation rates is valid whether analyses to construct Figure 13 are 731 repeated only for a time point at 16:30 LST or for a period between 16:30 and 17:00 LST.

These time point and period are related to analyses of the moist static energy as describedin Section e below.

At 17:20 LST, the larger precipitation frequency between  $\sim 2$  and  $\sim 12$  mm hr<sup>-1</sup> in the low-aerosol-b run nearly offsets the larger precipitation frequency in the other ranges of precipitation rates in the control-b run (Figure 9c). This leads to the similar average precipitation rate between the runs at 17:20 LST and contributes to the similar cumulative precipitation at the last time step between the runs (Figure 3b).

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#### d. Evaporation and gust fronts

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742 As time progresses from 17:00 to 19:00 LST, the precipitation system moves northward 743 (Figure 14). At the core of the precipitation system, due to evaporation and downdrafts, 744 there is the horizontal outflow forming at 17:00 LST (Figures 14a and 14b). The core is 745 represented by the field of precipitation whose rates are higher than 1 mm hr<sup>-1</sup> in Figure 14. 746 At the core, the northward outflow is magnified by the northward synoptic-scale wind, while at the core, the outflow in the other directions is offset by the northward synoptic-747 748 scale wind. Hence, the outflow is mainly northward from 17:00 LST onwards as marked 749 by yellow circles in Figures 14. This enables convergence or a gust front, which is produced 750 by the outflow from the core, to be mainly formed at the north of the core. Note that the 751 intensity of a gust front is proportional to that of outflow from a core of precipitation or 752 convective system (Weisman and Klemp, 1982; Houze, 1993). The strong gust front at the 753 north of the core generates strong updrafts, a significant amount of condensation and 754 precipitation. Then, a subsequent area with clouds and precipitation is formed at the north 755 of the core as time progresses, which means that the precipitation system extends or moves 756 to the north as seen in comparisons between sub-panels with different times in Figure 14. 757 This movement, which is induced by collaborative work between outflow, synoptic wind 758 and gust fronts, is typical in deep convective clouds.

As described above, the more droplet nucleation and greater integrated droplet surface induce more condensation before 17:00 LST in the control-b run. This and lower efficiency of collision and collection among droplets enable the control-b run to have a larger amount of cloud liquid or droplets as a source of evaporation. This in turn enables more droplet

763 evaporation, more associated cooling and stronger downdrafts, although less rain 764 evaporation is in the control-b run particularly for the period from 17:00 LST to 19:00 LST. 765 The time- and domain-averaged droplet and rain evaporation rates are 0.72(0.31) and 0.08766 (0.13) g m<sup>-3</sup> h<sup>-1</sup>, respectively, while the time- and domain-averaged downdraft mass flux is 0.15 (0.10) kg m<sup>-2</sup> s<sup>-1</sup> over the period from 17:00 LST to 19:00 LST in the control-b (low-767 768 aerosol-b) run. More evaporation of droplets and associated stronger downdrafts with 769 higher concentrations of aerosols acting as CCN have been shown by the numerous 770 previous studies (e.g., Tao et al., 2007; Tao et al., 2012; Khain et al., 2008; Lee et al., 2018). 771 During the period between 17:00 and 19:00 LST, with the development of convergence 772 or the gust front, as mentioned above, the maximum precipitation rate increases from  $\sim 17$ 773 (17) to ~ 45 (33) mm hr<sup>-1</sup> in the control-b (low-aerosol-b) run (Figure 9). This indicates that the gust-front development contributes to the overall intensification of the precipitation 774 775 system, while it moves northward. If there were only northward synoptic-scale wind with 776 no formation of the gust front, the system would move northward with less intensification. 777 Over the period from 17:00 LST to 19:00 LST, stronger downdrafts and associated stronger outflow generate a stronger gust front and more subsequent condensation in the control-b 778 779 run. This enhances the small initial difference, which is at 17:00 LST, in the frequency of 780 precipitation with rates above  $\sim 12 \text{ mm hr}^{-1}$  between the runs substantially as time 781 progresses from 17:00 LST to 19:00 LST (Figure 9). Associated with this, with the time 782 progress, the nearly identical maximum precipitation rate between the runs at 17:00 LST 783 turns into the significantly higher maximum precipitation rate in the control-b run than in 784 the low-aerosol-b run (Figure 9). Around 19:00 LST, the system enters its dissipating stage, 785 accompanying reduction in the precipitating area and the area-averaged precipitation rate 786 (Figures 3b).

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### e. Moist static energy

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Condensation, which controls droplet mass and precipitation, is controlled by updrafts and updrafts are in turn controlled by instability. One of important factors that maintain instability is the moist static energy. Motivated by this, to better understand differences in the precipitation frequency distribution in association with those in the condensation frequency distribution between the control-b and low-aerosol-b runs, we calculate the fluxof the moist static energy and the flux is defined as follows:

(1),

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797  $\overrightarrow{Fs} = S \times \rho \times \overrightarrow{V}$ 

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where  $\overrightarrow{Fs}$  represents the flux of the moist static energy, S the moist static energy,  $\rho$  the air 799 density and  $\vec{V}$  the horizontal-wind vector. In Eq. (1), we see that the flux is in the vector 800 form and has two components, which are its magnitude and direction. The fluxes of the 801 802 moist static energy in the PBL are obtained over the domain at 16:30 LST, since in general, 803 the moist static energy in the PBL has much stronger effects on instability and updrafts 804 than that above the PBL. In particular, we focus on the PBL fluxes of the energy that cross 805 the boundary over a time step at 16:30 LST between areas with the column-averaged condensation rate from  $3 \times 10^{-3}$  g m<sup>-3</sup> s<sup>-1</sup> to  $10 \times 10^{-3}$  g m<sup>-3</sup> s<sup>-1</sup>, which are referred to as "area 806 807 A", and those with the column-averaged condensation rate above  $10 \times 10^{-3}$  g m<sup>-3</sup> s<sup>-1</sup>, which are referred to as "area B". This is because we are interested in the exchange of the moist 808 809 static energy between areas A and B and this exchange can be seen by looking at those 810 fluxes which cross the boundary between those areas.

811 We are interested in the exchange of the energy, since we hypothesized that the 812 exchange somehow alters instability in each of areas A and B in a way that there are 813 increases (decreases) in instability, the updraft intensity, condensation and precipitation 814 with increasing concentrations of aerosols acting as CCN in area B (A), leading to the higher (lower) frequency of condensation whose rates are higher than  $10 \times 10^{-3}$  g m<sup>-3</sup> s<sup>-1</sup> 815 (between  $3 \times 10^{-3}$  and  $10 \times 10^{-3}$  g m<sup>-3</sup> s<sup>-1</sup>) and precipitation whose rates are higher than 12 816 mm hr<sup>-1</sup> (between 2 and 12 mm hr<sup>-1</sup>) in the control-b run than in the low-aerosol-b run. 817 818 When the PBL fluxes, which crosses the boundary over the time step at 16:30 LST, are 819 summed at 16:30 LST, there is the net flux from area A to area B. This means that there is 820 the net transportation of the moist static energy from areas with condensation rates between  $3 \times 10^{-3}$  and  $10 \times 10^{-3}$  g m<sup>-3</sup> s<sup>-1</sup> to those with condensation rates greater than  $10 \times 10^{-3}$  g m<sup>-3</sup> 821 s<sup>-1</sup> in the PBL at 16:30 LST as shown in Table 2. Table 2 shows the net summed flux of the 822 823 moist static energy which crosses the boundary between areas A and B in the control-b run 824 as well as the low-aerosol-b run. To calculate the net flux at 16:30 LST in Table 2, the

825 fluxes, which cross the boundary between areas A and B over the time step at 16:30 LST, 826 only at grid points in the PBL are summed. For the calculation, the flux from area A to area 827 B has a positive sign, while the flux from area B to area A has a negative sign. Since the 828 net flux is positive for both of the runs as shown in Table 2, there is the net flux from area 829 A to area B in the PBL. The above-described analysis for the fluxes crossing the boundary 830 between areas A and B is repeated for every time step between 16:30 and 17:00 LST and 831 based on this, the net summed flux over the period between 16:30 and 17:00 LST is 832 obtained. As shown in Table 2, the net flux for the period between 16:30 and 17:00 LST is 833 also positive as in the situation only for 16:30 LST. This means that there is the net 834 transportation of the moist static energy from area A to area B in the PBL during the period 835 between 16:30 and 17:00 LST.

At 16:30 LST, condensation with rates above  $10 \times 10^{-3}$  g m<sup>-3</sup> s<sup>-1</sup> starts to develop and 836 837 this forms area B. Area B has stronger updrafts via greater condensational heating than in 838 other areas, including area A, with lower condensation rates. Stronger updrafts in area B 839 induce the convergence of air and associated moist static energy from area A to area B. 840 Since the average condensation rate and updrafts at 16:30 LST over area B are higher and 841 stronger due to increasing concentrations of aerosols acting as CCN, respectively, the air 842 convergence and the associated transportation of the moist static energy in the PBL from 843 area A to area B are stronger and more, respectively, in the control-b run than in the low-844 aerosol-b run (Table 2). Stated differently, area B steals the moist static energy from area 845 A, and this occurs more effectively in the control-b run. This increases instability and 846 further intensifies updrafts in area B, and decreases instability and weakens updrafts in area 847 A, while these increases and decreases (intensification and weakening) of instability 848 (updrafts) are greater in the control-b run for the period from 16:30 LST to 17:00 LST. 849 This increases condensation, cloud mass and precipitation whose rates are higher than 12 850 mm hr<sup>-1</sup> in area B, and decreases condensation, cloud mass and precipitation whose rates are from 2 mm hr<sup>-1</sup> to 12 mm hr<sup>-1</sup> in area A. These increases and decreases occur more 851 852 effectively for the control-b run than for the low-aerosol-b run during the period. This in 853 turn leads to the lower precipitation frequency for the precipitation rates from 2 mm hr<sup>-1</sup> to 12 mm hr<sup>-1</sup> and the higher frequency for the precipitation whose rates are higher than 12 854 mm hr<sup>-1</sup> at 17:00 LST in the control-b run (Figure 9b). The weakened updrafts and reduced 855

condensation turn a portion of precipitation with rates between 2 and 12 mm hr<sup>-1</sup> to precipitation whose rates are below 2 mm hr<sup>-1</sup>, and this takes place more efficiently in the control-b run during the period between 16:30 and 17:00 LST. This eventually increases the frequency of precipitation rates below 2 mm hr<sup>-1</sup> and this increase is greater for the control-b run, leading to the greater precipitation frequency for the precipitation rates below 2 mm hr<sup>-1</sup> in the control-b run at 17:20 LST (Figure 9c).

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### 5. Discussion

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### 5.1 Comparison of the Seoul and Beijing cases

867 In this section, we compare the Seoul case to the Beijing case. For the comparison, 868 remember that on average, a pair of the control-s and low-aerosol-s runs has the same 869 perturbation of aerosols acting as CCN as in a pair of the control-b and low-aerosol-b runs. 870 Associated with the fact that clouds in the Seoul case are less deep than those in the Beijing 871 case, overall, updrafts in the Seoul case are not as strong as those in the Beijing case. Hence, 872 unlike the situation in the Beijing case, stronger updrafts, which accompany higher 873 condensation rates, and associated convergence in the Seoul case are not strong enough to 874 steal the sufficient amount of the moist static energy from weaker updrafts which 875 accompany lower condensation rates. This makes the redistribution of the moist static 876 energy between areas with relatively higher condensation rates and those with relative 877 lower condensation rates, such as that between areas A and B for the Beijing case, 878 ineffective for the Seoul case. Due to this, the sign of CCN-induced changes in the 879 frequency of precipitation rates does not vary throughout all of the precipitation rates 880 except for the range of low precipitation rates where there are nearly no CCN-induced 881 changes in the frequency in the Seoul case as shown in Figure 4a. As seen in Figure 4a, 882 mainly due to increases in condensation and deposition, precipitation frequency increases 883 for most of precipitation rates, although the precipitation frequency does not show 884 significant changes as concentration of aerosols acting as CCN increases for relatively low 885 precipitation rates in the control-s run as compared to that in the low-aerosol-s run. This 886 means that there are no tipping precipitation rates where the sign of CCN-induced changes

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in the frequency of precipitation rates changes in the Seoul case, contributing to the higher cumulative precipitation amount in the simulation with higher concentrations of aerosols acting as CCN for the Seoul case, which are different from the situation in the Beijing case.

890 In the Beijing case with deeper clouds as compared to those in the Seoul case, clouds 891 develop gust fronts via strong downdrafts and associated strong outflow. These gust fronts 892 play an important role in developing strong convection and associated high precipitation 893 rates. Unlike the situation in the Seoul case, there are strong clouds and associated updraft entities that are able to steal heat and moisture (or the moist static energy) as sources of 894 895 instability from areas with relatively less strong clouds and updrafts with medium strength; 896 note that these strong clouds here involve stronger updrafts via greater condensational 897 heating as described in Section e above and this enables these clouds to be thicker and have 898 higher cloud mass than these less strong clouds. This further intensifies strong clouds and 899 weakens less strong clouds with medium strength. Due to this, the cumulative frequency 900 of heavy (medium) precipitation in association with strong clouds (less strong clouds with 901 medium strength) increases (decreases). Some of the weakened clouds eventually produce 902 light precipitation, which increase the cumulative frequency for light precipitation. The 903 intensification of strong clouds and the weakening of less strong clouds with medium 904 strength gets more effective with increasing concentration of aerosols acting as CCN. 905 Hence, in the Beijing case, for medium precipitation in association with less strong clouds, 906 the simulation with higher concentration of aerosols acting as CCN shows the lower 907 cumulative precipitation frequency at the last time step. However, for heavy precipitation, 908 which is associated with strong clouds, and light precipitation, the simulation with higher 909 concentrations of aerosols acting as CCN shows the higher cumulative precipitation 910 frequency at the last time step. These differential responses of precipitation to increasing 911 concentration of aerosols acting as CCN among different types of precipitation occur in the 912 circumstances of the similar cumulative precipitation amount between the simulations with 913 different concentration of aerosols acting as CCN. This similar precipitation amount is due 914 to above-mentioned competition between CCN-induced changes in condensation, 915 deposition and freezing.

916 In both of the Seoul and Beijing cases, CCN-induced changes in condensation plays an 917 important role in making differences in the precipitation amount and/or the precipitation 918 frequency distribution between the simulations with different concentration of aerosols 919 acting as CCN. It is notable that in less deep clouds in the Seoul case, in addition to 920 condensation, deposition plays a role in precipitation to induce CCN-induced increases in 921 the precipitation amount. CCN-induced increases in condensation initiate the differences 922 in cloud mass and precipitation and then CCN-induced increases in deposition follow to 923 further enhance those differences. In deep clouds in the Beijing case, condensation tends 924 to induce increases in cloud mass and precipitation, while deposition tends to induce 925 decreases in cloud mass and precipitation with increasing concentration of aerosols acting 926 as CCN. Hence, as clouds get shallower and thus ice processes become less active, the role 927 of deposition in CCN-induced changes in precipitation amount turns from CCN-induced 928 suppression of precipitation to enhancement of precipitation. Here, we find that contrary 929 to the traditional understanding, the role of variation of freezing, which is induced by the 930 varying concentration of aerosols acting as CCN but not INPs, in precipitation is negligible 931 as compared to that of condensation and deposition in both of the cases.

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### 6. Summary and conclusions

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This study examines impacts of aerosols, which act as CCN, on clouds and precipitation in two metropolitan areas, which are the Seoul and Beijing areas, in East Asia that has experienced substantial increases in aerosol concentrations over the last decades. The examination is performed via simulations, which use a CSRM. These simulations are for deep clouds which reach the tropopause in the Beijing case and for comparatively less deep clouds which do not reach the tropopause yet grow above the level of freezing in the Seoul case.

In both of the cases, CCN-induced changes in condensation plays a critical role in CCN-induced variation of precipitation properties (e.g., the precipitation amount and the precipitation frequency distribution). In the Seoul case, CCN-induced increases in condensation and subsequent increases in deposition lead to CCN-induced increases in the precipitation frequency over most of precipitation rates and thus in the precipitation amount. However, in the Beijing case, while there are increases in condensation with increasing CCN concentrations, there are decreases in deposition with increasing CCN concentrations. 949 This competition between increases in condensation and decreases in deposition leads to negligible CCN-induced changes in cumulative precipitation amount in the Beijing case. 950 951 In both of the cases, CCN-induced changes in freezing are negligible as compared to those 952 in condensation and deposition. In the Beijing case, there is another competition for the 953 moist static energy among clouds with different updrafts and condensation. This 954 competition results in CCN-induced differential changes in the precipitation frequency 955 distributions. With clouds getting deeper from the Seoul case to the Beijing case, clouds 956 and associated updrafts, which are strong enough to steal the moist static energy from other 957 clouds and their updrafts, appear. This makes strong clouds stronger and clouds with 958 medium strength weaker. With higher CCN concentrations, strong clouds steal more 959 energy, and thus strong clouds become stronger and clouds with medium strength weaker 960 with a greater magnitude. As a result of this, there are more frequent heavy precipitation (whose rates are higher than 12 mm hr<sup>-1</sup>) and light precipitation (whose rates are lower than 961 962 2 mm hr<sup>-1</sup>), and less frequent medium precipitation (with rates from 2 mm hr<sup>-1</sup> to 12 mm 963 hr<sup>-1</sup>) with increasing CCN concentrations in the Beijing case.

964 In both of the Seoul and Beijing cases, there are mountains and they play an important 965 role in how cloud and precipitation evolve with time and space. In both of the cases, the 966 precipitating system moves or expands over mountains which induce forced convection 967 and generate condensation. This important role of mountains and forced convection in the 968 formation and evolution of the precipitation system has not been examined much in the 969 previous studies of aerosol-cloud interactions, since many of those previous studies (e.g., 970 Jiang et al., 2006; Khain et al., 2008; Li et al., 2011; Morrison et al., 2011) have dealt with 971 convective clouds that develop over plains and oceans. Hence, findings in this study, which 972 are related to mountain-forced convection and its interactions with aerosols, can be 973 complementary to those previous studies. Stated differently, this study can shed light on 974 our path to the understanding of aerosol-cloud interactions over more general domains not 975 only with no terrain but also with terrain.

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### Code/Data source and availability

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982 Our private computer system stores the code/data which are private and used in this study. 983 Note that in particular, the stored PM data are provided by the Korea Environment 984 Cooperation in South Korea and State Key Laboratory of Severe Weather in China. Upon 985 approval from funding sources, the data will be opened to the public. Projects related to 986 this paper have not been finished, thus, the sources prevent the data from being open to the 987 public currently. However, if information on the data is needed, contact the corresponding 988 author Seoung Soo Lee (slee1247@umd.edu).

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### 990 Author contributions

991 Essential initiative ideas are provided by SSL, KJH and KHS to start this work. Simulation 992 and observation data are analyzed by SSL, JC and GK. JU and YZ review the results and 993 contribute to their improvement. CHJ and JG perform additional simulations, which are 994 required by the review process, and their basic analyses. CHJ and SKS provide ideas to 995 handle the reviewers' comments.

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### 997 **Competing interests**

### 998 The authors declare that they have no conflict of interest.

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# Brown, A., Milton, S., Cullen, M., Golding, B., Mitchell, J., and Shelly, A.: Unified modeling and prediction of weather and climate: A 25-year journey, Bull. Am Meteorol. Soc. 93, 1865–1877, 2012.

- Chen, F., and Dudhia, J.: Coupling an advanced land-surface hydrology model with the
  Penn State-NCAR MM5 modeling system. Part I: Model description and
  implementation, Mon. Wea. Rev., 129, 569–585, 2001.
- Dong, B., Wilcox, L. J., Highwood, E. J., and Sutton, R. T.: Impacts of recent decadal
  changes in Asian aerosols on the East Asian summer monsoon: roles of aerosol–
  radiation and aerosol–cloud interactions, Clim. Dyn., 53, 3235-3256, 2019.
- Eun, S.-H., Kim, B.-G., Lee, K.-M., and Park, J.-S.: Characteristics of recent severe haze
  events in Korea and possible inadvertent weather modification, SOLA, 12, 32-36,
  2016.
- Fan, J., Yuan, T., Comstock, J. M., et al.: Dominant role by vertical wind shear in regulating
  aerosol effects on deep convective clouds, J. Geophys. Res., 114,
  doi:10.1029/2009JD012352, 2009.
- Fouquart, Y., and Bonnel, B.: Computation of solar heating of the Earth's atmosphere: a
  new parameterization, Beitr. Phys. Atmos., 53, 35-62, 1980.
- Ha, K.-J., Nam, S., Jeong, J.-Y., et al., Observations utilizing Korean ocean research
  stations and their applications for process studies, Bull. Amer. Meteor. Soc., 100,
  2061-2075, 2019.
- Holben, B. N., Tanré, D., Smirnov, et al.: An emerging ground-based aerosol climatology:
  Aerosol optical depth from AERONET, J. Geophys. Res., 106, 12067–12097, 2001.
- 1036 Houze, R. A., Cloud dynamics, Academic Press, 573 pp, 1993.
- Hwang, S.-O., and Lee, D.-K.: A study on the relationship between heavy rainfalls and
  associated low-level jets in the Korean peninsula, J. Korean. Meteorol. Soc., 29, 133–
  146, 1993.
- Jiang, H., Xue, H., Teller, A., Feingold, G., and Levin, Z.: Aerosol effects on the lifetime
  of shallow cumulus, Geophys. Res. Lett., 33, L14806, doi:10.1029/2006GL026024,
  2006.

- Kar, S. K., Lioi, Y.A., and Ha, K.-J. : Aerosol effects on the enhancement of cloud-toground lightning over major urban areas of South Korea, Atmos. Res., 92, 80-87,
  2009.
- Khain, A., BenMoshe, N., and Pokrovsky, A.: Factors determining the impact of aerosols
  on surface precipitation from clouds: Attempt of classification, J. Atmos. Sci., 65,
  1721-1748, 2008.
- Khain, A., Pokrovsky, A., Rosenfeld, D., Blahak, U., and Ryzhkoy, A.: The role of CCN in
  precipitation and hail in a mid-latitude storm as seen in simulations using a spectral
  (bin) microphysics model in a 2D dynamic frame, Atmos. Res., 99, 129–146, 2011.
- 1052 Khain, A., Rosenfeld, D., and Pokrovsky, A.: Aerosol impact on the dynamics and
  1053 microphysics of deep convective clouds, Quart. J. Roy. Meteor. Soc., 131, 2639-266,
  1054 2005.
- Khain, A. D., BenMoshe, N., and A. Pokrovsky, A.: Factors determining the impact of
  aerosols on surface precipitation from clouds: An attempt at classification, J. Atmos.
  Sci., 65, 1721–1748, doi:10.1175/2007JAS2515.1, 2008.
- 1058King,J.:Automaticweatherstations,availableat1059https://web.archive.org/web/20090522121225/http://www.automaticweatherstation.com/index.html, 2009.
- Koop, T., Luo, B. P., Tsias, A., and Peter, T.: Water activity as the determinant for
  homogeneous ice nucleation in aqueous solutions, Nature, 406, 611-614, 2000.
- Lebo, Z. J., and Morrison, H.: Dynamical effects of aerosol perturbations on simulated
  idealized squall lines, Mon. Wea. Rev., 142, 991-1009, 2014.
- 1065 Lee, D.-K., Kim, H.-R., and Hong, S.-Y.: Heavy rainfall over Korea during 1980–1990.
  1066 Korean, J. Atmos. Sci., 1, 32–50, 1998.
- Lee, S., Ho, C.-H., Lee, Y. G., Choi, H.-J. and Song, C.-K.: Influence of transboundary air
  pollutants from China on the high-PM10 episode in Seoul, Korea for the period
  October 16–20, 2008. Atmos. Environ., 77, 430–439, 2013.
- Lee, S. S., Donner, L. J., Phillips, V. T. J., and Ming, Y.: The dependence of aerosol effects
  on clouds and precipitation on cloud-system organization, shear and stability, J.
  Geophys. Res., 113, D16202, 2008.
- 1073 Lee, S. S., Kim, B.-G., and Yum, S. S., et al.: Effect of aerosol on evaporation, freezing and

- 1074 precipitation in a multiple cloud system, Clim. Dyn., 48, 1069-1087, 2016.
- Lee, S. S., Li, Z., Mok, J., et al,: Interactions between aerosol absorption, thermodynamics,
  dynamics, and microphysics and their impacts on clouds and precipitation in a
  multiple-cloud system, Clim. Dyn., <u>https://doi.org/10.1007/s00382-017-3552-x</u>,
  2017.
- Lee, S. S., Kim, B.-G., Li, Z., Choi, Y.-S., Jung, C.-H., Um, J., Mok, J., and Seo, K.-H.:
  Aerosol as a potential factor to control the increasing torrential rain events in urban
  areas over the last decades, Atmos. Chem. Phys., 18, 12531–12550,
  https://doi.org/10.5194/acp-18-12531-2018, 2018.
- Li, Z., Niu, F., Fan, J., Liu, Y., Rosenfeld, D., and Ding, Y.: Long-term impacts of aerosols
  on the vertical development of clouds and precipitation, Nat. Geosci., 4, 888-894,
  2011.
- Lohmann, U. and Diehl, K.: Sensitivity studies of the importance of dust ice nuclei for the
  indirect aerosol effect on stratiform mixed-phase clouds, J. Atmos. Sci., 63, 968-982,
  2006.
- Lu, Z., Zhang, Q., and Streets, D. G.: Sulfur dioxide and primary carbonaceous aerosol
  emissions in China and India, 1996–2010, Atmos. Chem. Phys., 11, 9839–9864, 2011.
- Mlawer, E. J., Taubman, S. J., Brown, P. D., Iacono, M. J., and Clough, S. A.: RRTM, a
  validated correlated-k model for the longwave, J. Geophys. Res., 102, 16663-1668,
  1093 1997.
- 1094 Möhler, O., et al, Efficiency of the deposition mode ice nucleation on mineral dust particles,
  1095 Atmos. Chem. Phys., 6, 3007-3021, 2006.
- Morrison, H., and Grabowski, W. W.: Cloud-system resolving model simulations of aerosol
  indirect effects on tropical deep convection and its thermodynamic environment,
  Atmos. Chem. Phys., 11, 10503–10523, 2011.
- 1099 Oh, H., Ha, K.-J. and Timmermann, A.: Disentangling Impacts of Dynamic and
  1100 Thermodynamic Components on Late Summer Rainfall Anomailes in East Asia, J.
  1101 Geophys. Res., 123, 8623-8633, 2018.
- Oh, H.-R., Ho, C.-H., Kim, J., Chen, D., Lee, S., Choi, Y.-S., Chang, L.-S., and Song, C.K.: Long-range transport of air pollutants originating in China: A possible major cause
  of multi-day high-PM10 episodes during cold season in Seoul, Korea. Atmos.

- 1105 Environ., 109, 23–30, 2015.
- Pruppacher, H. R. and Klett, J. D.: Microphysics of clouds and precipitation, 714pp, D.
  Reidel, 1978.
- Rosenfeld, D., Lohmann, U., Raga, G. B., et al.: Flood or drought, How do aerosols affect
  precipitation? Science, 321, 1309-1313, 2008.
- Storer, R. L., van den Heever, S. C., and Stephens, G. L.: Modeling aerosol impacts on
  convection under differing storm environments, J. Atmos. Sci., 67, 3904-3915, 2010.
- Seo, K.-H., Son, J. H., Lee, J.-H., and Park, H.-S.: Northern East Asian monsoon
  precipitation revealed by air mass variability and its prediction, J. Clim., 28, 62216233, 2013.
- Tao, W.-K., Chen, J.-P., Li, Z., Wang, C., and Zhang, C.: Impact of aerosols on convective
  clouds and precipitation, Rev. Geophys., 50, RG2001, 2012.
- 1117 Tao, W. K., Cloud resolving modeling, J. Meteorol. Soc. Jpn., 85B, 305–330,
  1118 doi:10.2151/jmsj.85B.305, 2007.
- van den Heever, S. C., Carrió, G. G., Cotton, W. R., DeMott, P. J., and Prenni, A. J.:
  Impacts of nucleating aerosol on Florida storms. part I: Mesoscale simulations, J.
  Atmos. Sci., 63, 1752–1775, 2006.
- Wang, H., Skamarock, W. C., and Feingold, G.: Evaluation of scalar advection schemes in
  the Advanced Research WRF model using large-eddy simulations of aerosol-cloud
  interactions, Mon. Wea. Rev., 137, 2547-2558, 2009.
- Weisman, M. L., and Klemp, J. B.: The dependence of numerically simulated convective
  storms on vertical wind shear and buoyancy, Mon. Wea. Rev., 110, 504-520, 1982.
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- 1140 FIGURE CAPTIONS
- 1141

Figure 1. Inner rectangles in (a) and (b) mark the Seoul area in the Korean Peninsula and the Beijing area in the East-Asia continent, respectively. A dot outside the inner rectangle in (a) marks Baekryongdo island. Dots in the inner rectangles in (a) and (b) mark the selected locations where precipitation and aerosol mass are measured. In (a) and (b), the light blue represents the ocean and the green the land area.

1148Figure 2. Time series of  $PM_{2.5}$  observed at the ground station in Backryongdo island (blue1149line) and of the average  $PM_{2.5}$  over ground stations in the Seoul area (red line) between115007:00 LST on December  $22^{nd}$  and 21:00 LST on December  $24^{th}$  in 2017. Note that  $PM_{2.5}$ 1151observed at stations in the Seoul area is applied to the control-s run whose period is marked1152by the dashed rectangle. Time series of the average  $PM_{2.5}$  over stations in the Seoul area in1153the low-aerosol-s run for the simulation period is also shown (black solid line).

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Figure 3. Time series of precipitation rates at the surface, which are averaged over the domain and smoothed over 1 hour, (a) for the control-s and low-aerosol-s runs in the Seoul area and (b) for the control-b and low-aerosol-b runs in the Beijing area. In (a) and (b), the averaged and observed precipitation rates over the observation sites in the Seoul and Beijing areas, respectively, are also shown.

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Figure 4. Observed and simulated cumulative frequency distributions of precipitation rates at the surface for (a) the Seoul case, which are collected over the Seoul area, and (b) the Beijing case, which are collected over the Beijing area, at the last time step. Simulated distributions are in the control-s and low-aerosol-s runs for the Seoul case and in the control-b and low-aerosol-b runs for the Beijing case. The observed distribution is obtained by interpolating and extrapolating the observed precipitation rates to grid points and time steps in the control-s and control-b runs for the Seoul and Beijing cases, respectively. Figure 5. Cumulative frequency distributions of the precipitation rates at the surface in the
control-s and low-aerosol-s runs for the Seoul case at (a) 06:00, (b) 10:00 and (c) 12:00
LST.

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Figure 6. Vertical distributions of differences in the area-averaged condensation, deposition and freezing rates, and cloud-liquid, raindrop, snow and hail mass density, and updraft mass fluxes between the control-s and low-aerosol-s runs at (a) 03:20, (b) 03:40, (c) 06:00 and (d) 12:00 LST. The horizontal black line in each panel represents the altitude of freezing or melting. Here, for the sake of the display brevity, snow mass density includes ice-mass density, while hail mass density includes graupel mass density.

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1180 Figure 7. Spatial distributions of terrain heights, column-averaged condensation rates, 1181 surface wind vectors and precipitation rates at (a) and (b) 03:40, (c) and (d) 08:40, (e) and 1182 (f) 10:00, and (g) and (h)12:00 LST. The distributions in the control-s run are shown in (a), 1183 (c), (e) and (g), and the distributions in the low-aerosol-s run are shown in (b), (d), (f) and 1184 (h). Condensation rates are shaded. Dark-yellow and dark-red contours represent 1185 precipitation rates at 0.5 and 3.0 mm hr<sup>-1</sup>, respectively, while beige, light brown and brown 1186 contours represent terrain heights at 100, 300 and 600 m, respectively. See text for yellow 1187 rectangles in (a), (b), (e) and (f).

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Figure 8. Cumulative frequency distributions of the column-averaged condensation rates in the control-s and low-aerosol-s runs for the Seoul case at (a) 07:20 and (b) 09:00 LST.

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Figure 9. Cumulative frequency distributions of the precipitation rates at the surface in the
control-b and low-aerosol-b runs for the Beijing case at (a) 16:00, (b) 17:00, (c) 17:20, and
(d) 19:00 LST.

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1196 Figure 10. Same as Figure 6 but for differences between the control-b and low-aerosol-b

1197 runs at (a) 14:20, (b) 15:40, (c) 16:00, (d) 17:20 and (e) 19:00 LST.

Figure 11. Spatial distributions of terrain heights, column-averaged condensation rates, surface wind vectors and precipitation rates at (a) and (b) 14:20, and (c) and (d) 17:20 LST. (a) and (c) are for the control-b run and (b) and (d) are for the low-aerosol-b run. Condensation rates are shaded. Dark-yellow and dark-red contours represent precipitation rates at 1.0 and 2.0 mm hr<sup>-1</sup>, respectively, while beige, light brown, brown and dark brown contours represent terrain heights at 100, 500, 1000 and 1500 m, respectively. See text for vellow circles in (a) and (b). Figure 12. Cumulative frequency distributions of the column-averaged condensation rates in the control-b and low-aerosol-b runs for the Beijing case at (a) 17:20 and (b) 19:00 LST. Figure 13. Mean precipitation rates corresponding to each column-averaged condensation rate for the period between 14:00 and 17:20 LST in the control-b run. One standard deviation of precipitation rates is represented by a vertical bar at each condensation rate. Figure 14. Spatial distributions of precipitation rates (shaded) and wind vectors (arrows) for the Beijing case at (a) and (b) 17:00, and (c) and (d) 19:00. The distributions in the control-b run are in (a) and (c). The distributions in the low-aerosol-b run are in (b) and (d). 

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Simulations	Site	Concentrations of background aerosols acting as CCN
Control-s run	Seoul area	Observed and affected by the aerosol advection
Low-aerosol-s run	Seoul area	Same as those in the control-s run but unaffected by the aerosol advection
Control-b run	Beijing area	Observed
Low-aerosol-b run	Beijing area	Reduced by a factor of 3.1 as compared to those observed

1230 Table 1. Summary of simulations

	The net flux of the moist static energy		
	which crosses the boundary between areas		
Simulations	A and B		
	$(J m^{-2} s^{-1})$		
	At 16:30 LST	16:30 to 17:00 LST	
Control-b run	$1.57 \times 10^{12}$	$1.07 \times 10^{15}$	
Low-aerosol-b run	$1.15 \times 10^{12}$	7.55 ×10 <sup>14</sup>	

- 1234 Table 2. The net flux of the moist static energy which crosses the boundary between areas
- 1235 A and B at 16:30 LST and for a period from 16:30 to 17:00 LST.







Figure 3









Figure 5



Figure 6











### Seoul case



### Beijing case







Figure 10







### Beijing case





