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1 **Impacts of an aerosol layer on a mid-latitude continental system of cumulus clouds:**

2 **how do these impacts depend on the vertical location of the aerosol layer?**

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4 Seoung Soo Lee[1,2], Junshik Um[3,4], Won Jun Choi[5], Kyung-Ja Ha[2,4,6], Chang Hoon Jung[7],

5 Jianping Guo[8], Youtong Zheng[9]

6

7 [1]Earth System Science Interdisciplinary Center, University of Maryland, Maryland, USA

8 [2]Research Center for Climate Sciences, Pusan National University, Busan, Republic of

9 Korea

10 [3]Department of Atmospheric Sciences, Pusan National University, Busan, Republic of

11 Korea

12 [4]BK21 School of Earth and Environmental Systems, Pusan National University, Busan,

13 Republic of Korea

14 [5]National Institute of Environmental Research, Incheon, Republic of Korea

15 [6]Center for Climate Physics, Institute for Basic Science, Busan, Republic of Korea

16 [7]Department of Health Management, Kyungin Women's University, Incheon, Republic of

17 Korea

18 [8]State Key Laboratory of Severe Weather, Chinese Academy of Meteorological Sciences,

19 Beijing, China

20 [9]The Program in Atmospheric and Oceanic Sciences, Princeton University, Princeton,

21 New Jersey, USA

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25   Corresponding authors: Seoung Soo Lee and Junshik Um

26   E-mail: cumulss@gmail.com, slee1247@umd.edu, jjunum@pusan.ac.kr

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52 **Abstract**

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54 Using the large-eddy simulation framework, effects of an aerosol layer on warm cumulus

55 clouds in the Korean Peninsula when the layer is above or around the cloud tops in the

56 upper atmosphere are examined. Also, these effects are compared to effects of an aerosol

57 layer when it is around or below the cloud bases in the low atmosphere. Simulations show

58 that when the aerosol layer is in the low atmosphere, aerosols absorb solar radiation and

59 radiatively heat up air enough to induce greater instability, stronger updrafts and more

60 cloud mass than when the layer is in the upper atmosphere. As aerosol concentrations in

61 the layer decrease, the aerosol radiative heating gets weaker to lead to less instability,

62 weaker updrafts and less cloud mass when the layer is in the low atmosphere. This in turn

63 makes differences in cloud mass, which are between a situation when the layer is in the

64 low atmosphere and that when the layer is in the upper atmosphere, smaller. It is found that

65 the transportation of aerosols by updrafts reduces aerosol concentrations in the aerosol

66 layer, which is in the low atmosphere, and in turn reduces the aerosol radiative heating,

67 updraft intensity and cloud mass. It is also found that the presence of aerosol impacts on

68 radiation suppresses updrafts and reduces clouds. Aerosols affect not only radiation but

69 also aerosol activation. In the absence of aerosol impacts on radiation, aerosol impacts on

70 the droplet nucleation increases cloud mass when the layer is in the low atmosphere as

71 compared to a situation when the layer is in the upper atmosphere. As aerosol impacts on

72 radiation team up with those on the droplet nucleation, differences in cloud mass, which

73 are between a situation when the layer in the low atmosphere and that when the layer is in

74 the upper atmosphere, get larger. This is as compared to a situation when there is no aerosol

75 impacts on radiation and only aerosol impacts on the droplet nucleation.

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1. Introduction

Warm cumulus clouds play an important role in global hydrologic and energy circulations (Warren et al., 1986; Stephens and Greenwald, 1991; Hartmann et al., 1992; Hahn and Warren, 2007; Wood, 2012). With industrialization, there have been significant increases in concentrations of aerosols acting as cloud condensation nuclei (CCN) and these increases are known to decrease droplet size (Twomey, 1974, 1977). Increases in concentrations of aerosols acting as radiation absorbers are also known to enhance the radiative heating of air by aerosols. These aerosol-induced changes in droplet size and radiative heating affect updrafts, cloud mass, cloud albedo and precipitation (Albrecht, 1989; Hansen et al., 1997). Global hydrologic and energy circulations are eventually affected by these aerosol effects. However, these effects, which are particularly on warm cumulus clouds, are highly uncertain and thus act to cause the highest uncertainty in the prediction of future climate (Ramaswamy et al., 2001; Forster et al., 2007).

In recent years, people start to take interest in effects of aerosol layers above or around the tops of clouds on clouds (e.g., de Graaf et al., 2014; Xu et al., 2017). This interest is motivated by aerosol layers that are originated from biomass burning sites in the southern Africa. These layers are lifted and transported to the southeast Atlantic (SEA) region and located above or around the top of a large deck of warm cumulus and stratocumulus clouds that play an important role in global hydrologic and energy circulations. Note that aerosols in the transported aerosol layers contain organic carbon (OC) and black carbon (BC) that act as radiation absorbers as well as cloud condensation nuclei (CCN). When these aerosols act as radiation absorbers, they absorb solar radiation and heat up the atmosphere to change atmospheric stability. This in turn affects the cumulus clouds, the hydrologic and energy circulations, and climate. When these aerosols act as CCN, they have an impact on aerosol activation, subsequent microphysical and precipitation processes in the cumulus clouds, those circulations and climate. Reflecting the interest and an associated potential importance of aerosol layers above or around cloud tops in the circulations and climate, to better understand roles of aerosol layers above or around cloud tops in cloud development and its impacts on climate, there were international field campaigns in the SEA such as the NASA ObseRvations of Aerosols above CLouds and their intEractionS (ORACLES;

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114   https://espo.nasa.gov/oracles/content/ORACLES), the UK Clouds and Aerosol Radiative

115   Impacts and Forcing (CLARIFY; Redemann et al., 2021) and the French Aerosol,

116   Radiation and Clouds in southern Africa (AEROCLO-sA; Formenti et al., 2019)

117   campaigns.

118   It is well-known that the relative vertical location of an aerosol layer and a cloud deck

119   can affect cloud properties (e.g., updrafts, cloud mass and albedo) that responds to aerosol

120   absorption, subsequent changes in atmospheric stability, aerosol activation and subsequent

121   changes in microphysics and precipitation (Haywood and Shine, 1997; Johnson et al., 2004;

122   McFarquhar and Wang, 2006). However, despite above-mentioned field campaigns,

123   previous studies on aerosol-cloud interactions have focused mainly on effects of aerosols

124   around or below cloud bottoms on clouds. Effects of aerosols above or around cloud tops

125   on clouds have not been examined as much. This contributes to the low-level understanding

126   of effects of the relative location of an aerosol layer and a cloud deck on the cloud deck.

127   Improving this understanding, which is about going beyond the traditional approach that

128   focuses on around- or below-cloud-bottom aerosol layers, is likely to contribute to the more

129   comprehensive understanding of aerosol-cloud-radiation interactions and thus more

130   general parameterizations of those interactions for climate models. Hence, this study aims

131   to enhance our understanding of effects of the relative location of an aerosol layer and a

132   cloud deck on the cloud deck. This aim is pursued by investigating aerosol-cloud-radiation

133   interactions in a typical situation where an aerosol layer is around or below the bottom of

134   a system of warm cumulus clouds. Then, to fulfill the aim, these interactions in the typical

135   situation are compared to a situation where an aerosol layer is around or above the top of

136   the system. In this study, the investigation is performed by using simulations adopting the

137   large-eddy simulation (LES) framework and an idealized setup, which is based on

138   observation, for the aerosol layer.

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140   **2. Case, model and simulations**

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142   **2.1 LES model**

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144   The Advanced Research Weather Research and Forecasting (ARW) model is for LES

145   simulations in this study. The ARW model is a compressible model with a nonhydrostatic

146   status. A 5th-order monotonic advection scheme is used to advect microphysical variables

147   (Wang et al., 2009). The ARW adopts a bin scheme to parameterize microphysics. The

148   Hebrew University Cloud Model (HUCM) detailed in Khain et al. (2011) is the bin scheme.

149   A set of kinetic equations is solved by the bin scheme to represent size distribution

150   functions for each class of hydrometeors and aerosols acting as cloud condensation nuclei

151   (CCN). The hydrometeor classes are water drops, ice crystals (plate, columnar and branch

152   types), snow aggregates, graupel and hail. There are 33 bins for each size distribution in a

153   way that the mass of a particle m_j in the j bin is to be $m_j = 2m_{j-1}$.

154   Aerosol sinks and sources control the evolution of aerosol size distribution. These

155   sinks and sources include advection and aerosol activation (Fan et al., 2009). Activated

156   particles are emptied in the corresponding bins of the aerosol spectra. Aerosol mass

157   included in hydrometeors, after activation, is moved to different classes and sizes of

158   hydrometeors through collision-coalescence and removed from the atmosphere once

159   hydrometeors that contain aerosols reach the surface.

160   The Rapid Radiation Transfer Model (RRTM; Mlawer et al., 1997) has been coupled

161   to the bin microphysics scheme described above. Aerosols before their activation can affect

162   radiation by changing the reflection, scattering, and absorption of radiation. This radiative

163   effect of aerosol is represented following Feingold et al. (2005). The internal aerosol

164   mixture and the ARW relative humidity are used to calculate the hygroscopic growth of

165   the aerosol particles as well as their optical properties, represented by extinction, single

166   scattering albedo, and asymmetry factor. Aerosol uptake of water vapor is considered over

167   the range of relative humidity in the domain. In practice, hygroscopic growth and optical

168   property calculations are performed offline prior to simulation and stored in lookup tables.

169   Calculations are done for the prescribed aerosol size distribution and composition, and unit

170   concentration. During model runtime, grid-point number concentration and relative

171   humidity determine the look-up table entries that specify the grid-point aerosol optical

172   properties. The effective sizes of hydrometeors are calculated in an adopted microphysics

173   scheme and the calculated sizes are transferred to the RRTM to consider effects of the

174   effective sizes on radiation.

175   The presence of the aerosol perturbs the radiative fluxes reaching the surface, and its

176   subsequent partitioning into sensible and latent heat fluxes (i.e., the Bowen ratio). This is

177   accounted for with the interactive Noah land surface model (Chen and Dudhia, 2001).

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179   **2.2 Case and simulations**

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181   **2.2.1 Case and standard simulations**

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183   There is an observed system of warm cumulus clouds in a domain in the Korean Peninsula,

184   as marked in Figure 1, for a period between 10:00 and 18:00 LST on April 13th, 2015. This

185   system is simulated to fulfill the goal of this study. For a three-dimensional simulation (i.e.,

186   the control run) of the system in the domain over the period, a 50-m resolution is used for

187   the horizontal domain. The length of the domain in the east-west (north-south) direction is

188   20 (20) km. In the vertical domain, the resolution coarsens with height. The resolution in

189   the vertical domain is 20 m just above the surface and 100 m at the model top that is at ~

190   4.5 km in altitude. Initial and boundary conditions of potential temperature, specific

191   humidity, and wind for the simulation are provided by reanalysis data. These data are

192   produced by the Met Office Unified Model (Brown et al., 2012) every 6 hours on a 0.11°

193   × 0.11° grid. These data represent the synoptic-scale environment. Figure 2 depicts the

194   vertical distributions of potential temperature and water-vapor mixing ratio at 09:00 LST

195   on April 15th, 2015 in radiosonde sounding that is obtained near the domain. This vertical

196   distribution represents initial environmental conditions for the development of the clouds

197   in the control run. The conditional instability is present in the vertical profiles and this

198   favors the development of warm cumulus clouds. An open lateral boundary condition is

199   employed for the control run.

200   There is a site of the aerosol robotic network (AERONET; Holben et al., 2001) in the

201   domain. At 10:00 LST when clouds in the domain start to develop, there is an aerosol layer

202   advected from the East Asia and this layer causes aerosol pollution in the domain. This

203   advection is monitored by aerosol-measuring stations in the Yellow sea as described in Lee

204   et al. (2021). According to the AERONET measurement, which is 1 hour before the

205   observed cumulus clouds start to form, aerosol particles in the layer, on average, are an

206   internal mixture of 70 % ammonium sulfate, 22 % organic compound and 8% black carbon.

207   Aerosol chemical composition in this study is assumed to be represented by this mixture

208   in the whole domain during the whole simulation period. Based on the AERONET

209   observation, the size distribution of background aerosols acting as CCN is assumed to

210   follow a bi-modal log-normal distribution. Modal radius of this distribution is 0.11 and 1.2

211   μm and standard deviation of this distribution is 1.71 and 1.92, while the partition of

212   aerosol number, which is normalized by the total aerosol number of the size distribution,

213   is 0.999 and 0.001 for accumulation and coarse modes, respectively. It is assumed that the

214   size distribution of background aerosols acting as CCN in all parts of the domain during

215   the whole simulation period is assumed to follow the bi-modal size distribution.   The

216   average aerosol concentration in the layer over the domain at 10:00 LST is ~15000 cm^{-3}.

217   This average concentration is applied to all grid points in the layer at the first time step of

218   the control run. This aerosol layer is idealized to be located around and below cloud bases

219   between the surface and 1.0 km as shown in Figure 3a. Cloud bases are located around 1

220   km. Above the layer, aerosol concentration is assumed to be 150 cm^{-3}.

221   This study aims to understand differences in aerosol effects on warm cumulus clouds

222   between the situation where an aerosol layer is above and around the cloud tops and that

223   where the aerosol layer is around and below the cloud bases. To fulfill this goal, we repeat

224   the control run by an idealized setup where the aerosol layer is moved upward to altitudes

225   between 2.5 and 3.5 km as shown in Figure 3b. Altitudes between 2.5 and 3.5 km are places

226   where cloud tops are located frequently. Note that the simulated maximum cloud-top height

227   is 3.3 km. This repeated run is referred to as the aro-above-cld run. As shown in Figures

228   3a and 3b, aerosol concentrations in the aerosol layer are 15000 cm^{-3} in both of the runs.

229   Outside the main aerosol layer, aerosol concentration is set at 150 cm^{-3} in both of the runs

230   (Figures 3a and 3b). Here, we see that the depth of the main aerosol layer and aerosol

231   concentrations in the main layer are identical between the runs.

232   It is well-known that aerosol-cloud-radiation interactions are strongly dependent on

233   aerosol concentrations (Tao et al., 2012). We want to test how results in the control and

234   aro-above-cld runs are sensitive to aerosol concentrations in the main aerosol layer. For

235   the test, the control and aro-above-cld runs are repeated with 10 times lower aerosol

236   concentrations in the main aerosol layer but with no changes in aerosol concentrations in

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237 the domain outside the main aerosol layer. In these repeated runs, the aerosol concentration

238 in the main aerosol layer at the first time step is 1500 cm^{-3}. Henceforth, the repeated control

239 and aro-above-cld runs are referred to as the control-1500 and aro-above-cld-1500 runs.

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241 **2.2.2 Additional simulations**

242

243 Clouds affect aerosols by cloud processes such as nucleation of droplets and aerosol

244 transportation (or advection) by cloud-induced wind. Updrafts and downdrafts comprise

245 cloud-induced wind and transport aerosols upward and downward, respectively. Here, we

246 are interested in impacts of clouds on aerosols and how these impacts in turn affect aerosol

247 effects on clouds. To examine these aspects of aerosol-cloud interactions, the above-

248 mentioned four standard simulations (e.g., the control, aro-above-cld, control-1500 and

249 aro-above-cld-1500 runs) are repeated by preventing aerosol evolution with time at each

250 grid point. In other words, in these repeated runs, aerosol concentrations at each grid point,

251 which are set at the first time step, do not vary with time or are not affected by cloud

252 processes such as nucleation and advection. These repeated runs are referred to as the

253 control-novary, aro-above-cld-novary, control-1500-novary, and aro-above-cld-1500-

254 novary runs. By comparing the standard simulations to these repeated ones, roles of cloud

255 impacts on aerosols in aerosol-layer impacts on clouds are identified.

256 In this study, we aim to better understand roles of the interception (e.g., reflection,

257 scattering and absorption) of radiation by aerosols, which results in phenomena such as

258 radiative heating of air by aerosols. These roles are referred to as aerosol radiative effects.

259 To better understand aerosol radiative effects, the above four standard simulations are

260 repeated again by turning off aerosol radiative effects. These repeated runs are the control-

261 norad, aro-above-cld-norad, control-1500-norad, aro-above-cld-1500-norad runs. The

262 summary of simulations in this study is given in Table 1.

263

264 **3. Results**

265

266 **3.1 The control and aro-above-cld runs**

267

268   Figure 4 shows the time- and area-averaged vertical distributions of cloud-liquid mass

269   density that represents cloud mass for the standard simulations. In Figure 4, the cloud layer

270   is between 1.0 and 3.3 km in the control run and between 0.8 and 2.6 km in the aro-above-

271   cld run. The time- and domain-averaged cloud-liquid mass density is 0.7 and 1.3×10^{-3} g

272   m^{-3} in the control run and in the aro-above-cld run, respectively. Hence, we see that clouds

273   are thicker with their higher tops and have greater mass in the control run than in the aro-

274   above-cld run. This is despite the fact that aerosol concentrations in the main aerosol layer

275   in the control run are identical to those in the aro-above-cld run.

276   Figure 5a shows the time series of the domain-averaged liquid-water path, which is

277   the vertical integral of cloud-liquid mass density and thus, represents cloud mass, for the

278   standard simulations. During the initial stage of the cloud development between 12:50 and

279   13:50 LST, the average cloud mass is slightly higher in the control run than in the aro-

280   above-cld run. Also, the average non-zero cloud mass starts to appear earlier in the control

281   run than in the aro-above-cld run. Over the period between 13:50 and 14:10 LST, there is

282   a jump (or rapid increase or surge) in the average cloud mass in the control run but not in

283   the aro-above-cld run. During this period with the jump, at some specific time points, the

284   average mass is ~one order of magnitude higher in the control run than in the aro-above-

285   cld run. Of interest is that just after the jump and at 14:10 LST, the average mass in the

286   control run starts to decrease and at 14:40 LST, becomes lower than that in the aro-above-

287   cld run. Hence, the greater time- and domain-averaged cloud mass in the control run is

288   mainly attributed to the jump in cloud mass. As seen in Figures 5b and 5c that show the

289   time series of the domain-averaged updraft speed and condensation rates, respectively, the

290   average updraft mass fluxes and associated condensation rates in the control run are also

291   slightly higher in the control run than in the aro-above-cld run for the period between 12:50

292   and 13:50 LST. The average updraft speed and associated condensation rates in the control

293   run jump during the period between ~13:50 and ~14:10 LST, hence, these speed and rates

294   are much higher in the control run than in the aro-above-cld run during the period between

295   ~13:50 and ~14:10 LST (Figures 5b and 5c). After the jump, the speed and rates decrease

296   rapidly and become lower in the control run than in the aro-above-cld run (Figures 5b and

297   5c). Taking into account the fact that condensation is the only source of cloud mass and

298   the updraft speed strongly control the amount of condensation, the updraft speed,

299   condensation rate and cloud mass in each of the runs and differences in those variables

300   between the runs are similar in terms of their temporal evolution.

301   Figure 5d shows the time series of the domain-averaged convective available potential

302   energy (CAPE) for the control and aro-above-cld runs. Considering that updrafts grow by

303   consuming buoyancy energy, updraft intensity is proportional to CAPE that is the integral

304   of the buoyancy energy in the vertical domain. Hence, the evolution of CAPE in each of

305   the runs is similar to that of the updraft speed, associated condensation rates and cloud

306   mass. Accordingly, the evolution of differences in CAPE between the runs is similar to that

307   of those differences in the updraft speed, associated condensation rates and cloud mass.

308   This similarity includes the jump not only in CAPE but also in those speed, rates and mass

309   during the period between 13:50 and 14:10 LST in the control run. (052022)

310   In Figures 5, the peaks (or the maximum values) of the domain-averaged CAPE,

311   updrafts, condensation rates and cloud mass in the control run occur around 14:10 LST and

312   this occurrence is earlier than that which occurs around 14:50 LST in the aro-above-cld

313   run. This means that the cloud system in the control run reaches its mature stage earlier

314   than that in the aro-above-cld run. After the peak around 14:10 LST, the system enters its

315   dissipating stage in the control run, while around 14:10 LST in the aro-above-cld run, the

316   system still evolves to enter the mature stage and it enters its dissipating stage after 14:50

317   LST. Hence, the cloud system in the control run matures and demises faster as compared

318   to that in the aro-above-cld run. Stated differently, the cloud system in the control run has

319   a shorter life cycle than that in the aro-above-cld run.

320   To find mechanisms controlling the jump in CAPE which acts as a main cause of the

321   greater cloud mass in the control run, the analysis of the results is done for an initial period

322   between 10:00 LST when the simulation starts and 13:50 LST which is immediately before

323   the jump starts to occur. The thermodynamic condition for the CAPE jump is established

324   during this initial period. The average net shortwave fluxes at the surface are shown in

325   Table 2 for the initial period in the control and aro-above-cld runs. Table 2 shows that

326   during the initial period in the control run, there is a smaller amount of shortwave radiation

327   that are incident on the surface in the control run than in the aro-above-cld run. The aerosol

328   layer intercepts solar radiation and reduces solar radiation which reaches the surface. In

329   spite of the fact that the depth of the main aerosol layer and aerosol concentrations in the

330   layer are identical between the runs, results here indicate that the aerosol layer in the low

331   atmosphere is more efficient in the interception of solar radiation than that in the upper

332   atmosphere. Due to the less solar radiation reaching the surface, the time- and area-

333   averaged net surface heat fluxes, which are the sum of the surface sensible and latent-heat

334   fluxes, become lower in the control run than in the aro-above-cld run during the initial

335   period (Table 2). Hence, the surface fluxes favor more instability or higher CAPE and

336   associated more intense subsequent updrafts and cloud mass in the aro-above-cld run than

337   in the control run.

338   The vertical distributions of the time- and domain-averaged radiative heating rates are

339   obtained for the initial period for each of the control and aro-above-cld runs. For the initial

340   period, the average radiative heating rate is much higher in the control run than in the aro-

341   above-cld run particularly at altitudes between 0.0 and ~ 1.0 km where cloud bases are

342   located (Figure 6a). This is associated with the fact that the main aerosol layer is located at

343   altitudes between 0.0 and 1.0 km in the control run. This more radiative heating in the low

344   atmosphere favors subsequent higher CAPE, which involves its jump in the control run, in

345   the control run than in the aro-above-cld run after the initial period. The average radiative

346   heating rate is higher in the aro-above-cld run than in the control run at altitudes between

347   ~2.5 and 3.5 km. This is associated with the fact that the main aerosol layer is located at

348   altitudes between 2.5 and 3.5 km in the aro-above-cld run. However, this higher radiative

349   heating rate is in the upper part of the domain and tends to stabilize the atmosphere more

350   in the aro-above-cld run than in the control run. Thus, the higher radiative heating rate in

351   the aro-above-cld run contributes to lower CAPE, less intense updrafts and lower cloud

352   mass in the aro-above-cld run especially for the period when the jumps occur in the control

353   run.

354   Effects of greater radiative heating in the low atmosphere on CAPE outweigh those

355   effects of solar radiation which are incident on the surface and the associated surface heat

356   fluxes during the initial stage in the control run. This leads to more intense clouds with

357   more cloud mass for the rest of periods, in turn leading to the more time- and domain-

358   averaged cloud mass in the control run than in the aro-above-cld run.

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360   **3.2 Comparisons between simulations with different aerosol concentrations**

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362   With the lower concentration of aerosols in the main layer, there are much more solar

363   radiation reaching the surface and resultant higher surface fluxes in the control-1500 run

364   than in the control run and in the aro-above-cld-1500 run than in the aro-above-cld run

365   (Table 2). This makes CAPE higher in the control-1500 run than in the control run over

366   most of the simulation period except for the period between 13:50 and 14:20 LST during

367   which the jump in CAPE in the control run exists, and in the aro-above-cld-1500 run than

368   in the aro-above-cld run throughout the simulation period (Figure 5d). Then, there are

369   stronger updrafts and greater condensation rate and cloud-liquid mass density developing

370   in the control-1500 run than in the control run over most of the simulation period, which

371   are except for the period between 13:50 and 14:20 LST during which the jump in updrafts

372   and cloud-liquid mass in the control run exists, and in the aro-above-cld-1500 run than in

373   the aro-above-cld run throughout the simulation period (Figures 5a, 5b and 5c). This leads

374   to the greater time- and domain-averaged cloud-liquid mass density in the control-1500 run

375   than in the control run and in the aro-above-cld-1500 run than in the aro-above-cld run

376   (Figure 4). Regarding the control and control-1500 runs, this is despite the fact that aerosol

377   radiative heating in the main aerosol layer in the low atmosphere is higher due to higher

378   aerosol concentrations there in the control run than in the control-1500 run (Figure 6). In

379   Figure 4, it is seen that the time- and domain-averaged cloud-liquid mass in the aro-above-

380   cld-1500 run is higher than in the control run. This is due to more solar radiation reaching

381   the surface in the aro-above-cld-1500 run than in the control run and despite the fact that

382   aerosol concentrations and thus aerosol radiative heating in the low atmosphere is much

383   higher in the control run than in the aro-above-cld-1500 run (Table 2 and Figure 6). In

384   Figure 4, it is also seen that the time- and domain-averaged cloud-liquid mass in the

385   control-1500 run is higher than in the aro-above-cld run. This is due to higher aerosol

386   concentrations and more aerosol radiative heating in the low atmosphere (in the upper

387   atmosphere) in the control-1500 run (aro-above-cld run) than in the aro-above-cld run

388   (control-1500 run).

389   Similar to the situation between the control and aro-above-cld runs, there is less solar

390   radiation reaching the surface in the control-1500 run than in the aro-above-cld-1500 run

391   (Table 2). In association with this, there is the less surface heat fluxes in the control-1500

392 run than in the aro-above-cld-1500 run. This favors higher CAPE and more invigoration of

393 updrafts and associated convection in the aro-above-cld-1500 run than in the control-1500

394 run. However, overall, CAPE is higher in the control-1500 run than in the aro-above-cld-

395 1500 run (Figure 5d). This is because similar to the situation between the control and aro-

396 above-cld runs, more aerosols are in the low atmosphere in the control-1500 run than in

397 the aro-above-cld-1500 run. These more aerosols heat up the low atmosphere more and

398 increase the instability there more (Figure 6c). This induces increases in CAPE, which

399 compensates for decreases in CAPE due to the smaller amount of solar radiation reaching

400 the surface in the control-1500 run, and leads to overall higher CAPE in the control-1500

401 run than in the aro-above-cld-1500 run.

402   Associated with higher CAPE, there is greater cloud-liquid mass density in the control-

403 1500 run than in the aro-above-cld-1500 run, which is similar to the situation between the

404 control and aro-above-cld runs. However, differences in the mass density between these

405 repeated runs are smaller than those between the control and aro-above-cld runs (Figure 4).

406 As seen in Figure 5a which shows the time series of the domain-averaged cloud-liquid

407 mass density, the control-1500 run does not show a jump in the mass density unlike the

408 situation in the control run. This contributes to smaller differences in cloud-liquid mass

409 density between the control-1500 and aro-above-cld-1500 runs than between the control

410 and aro-above-cld runs. The CAPE evolution of the control-1500 and aro-above-cld-1500

411 runs show that there is no jump in CAPE and thus updrafts in the control-1500 run (Figure

412 5d). This mainly contributes to smaller differences in CAPE and updrafts, which in turn

413 contributes to smaller differences in cloud mass between the control-1500 and aro-above-

414 cld-1500 runs than between the control and aro-above-cld runs.

415   In addition, remember that the cloud system in the control run has a shorter life cycle

416 than in the aro-above-cld run. However, as seen in Figure 5, the cloud system the conrol-

417 1500 run has a similar life cycle to that in the aro-above-cld-1500 run. In the control run,

418 the instability or CAPE accumulates or increases rapidly to reach its peak, which forms the

419 jump, for a period between 13:50 and 14:10 LST, while in the control-1500 run, CAPE

420 increases gradually to reach its peak from ~12:00 LST to ~14:30 LST (Figure 5d). For a

421 period between ~14:10 and ~14:50 LST, CAPE reduces rapidly down back to the CAPE

422 value around ~13:00 LST in the control run, while CAPE decreases gradually and never

423   drops back to CAPE value at ~12:00 LST until the end of the simulation period in the
424   control-1500 run. This leads to the shorter life cycle or lifetime of the system not only in
425   the control run than in the aro-above-cld run but also in the control run than in the control-
426   1500 run. Here, we see that as aerosol concentration increases in the main aerosol layer in
427   the low atmosphere, the time scale of the accumulation and consumption of the instability
428   or convective energy gets shorter, leading to the shorter lifetime of the cloud system. When
429   aerosol concentration in the main aerosol layer in the low atmosphere is relatively low as
430   in the control-1500 run, the relatively long lifetime of the cloud system in the control-1500
431   run is similar to the lifetime in the aro-above-cld-1500 run. However, when aerosol
432   concentration in the main aerosol layer in the low atmosphere is relatively high as in the
433   control run, the relatively short lifetime of the cloud system in the control run is shorter
434   than the lifetime in the aro-above-cld run.

435   Comparisons among the above four standard simulations show that with increasing
436   aerosol concentrations in the main aerosol layer, there are decreases in solar radiation
437   reaching the surface due to the increasing interception of solar radiation by aerosols,
438   leading to decreases in cloud-liquid mass density, whether the main aerosol layer is in the
439   low atmosphere or in the upper atmosphere. Also, when there is the main aerosol layer in
440   the low atmosphere, there is radiative heating of air by aerosols around and below cloud
441   bases and this enables more instability, stronger updrafts and more cloud-liquid mass
442   despite less solar radiation reaching the surface than when there is the main aerosol layer
443   in the upper atmosphere, whether aerosol concentrations in the main layer is or low
444   (initially set at 1500 cm^{-3}) or high (initially set at 15000 cm^{-3})

445   The increase in cloud-liquid mass from the aro-above-cld-1500 run to the control-1500
446   run is smaller than that from the aro-above-cld run to the control run. This means that with
447   increasing concentrations of aerosols, the effects of radiative heating of aerosols, which is
448   in the low atmosphere, on instability and cloud-liquid mass enhances. This enhancement
449   is closely linked to the jump in the CAPE and updrafts that appears when concentration of
450   aerosols in the low atmosphere is high. Stated differently, the jump in the CAPE does not
451   occur when concentration of aerosols in the low atmosphere is low, meaning that there is
452   a critical value of initial aerosol concentrations above which the jump occurs.

453

3.3 Comparisons between simulations with predicted and prescribed aerosol concentrations

Figure 7 shows the vertical distributions of aerosol concentrations, which are averaged over the horizontal domain and simulation period, for the standard and repeated runs with no temporal variation of aerosols (e.g., the control-novary, aro-above-cld-novary, control-1500-novary, and aro-above-cld-1500-novary runs). Comparisons between the control and control-novary runs (between the control-1500 and control-1500-novary runs) show that due to the upward transportation of aerosols by updrafts in the control (control-1500) run, aerosol concentrations in the main aerosol layer in the low atmosphere reduces and those in the air above the main aerosol layer increases. This is as compared to the situation in the control-novary (control-1500-novary) run where aerosols are assumed not to be affected by cloud-induced wind (Figures 7a and 7c). Note that the low atmosphere is where cloud-induced updrafts develop and grow, hence, the upward transportation of aerosols by them is dominant. Due to the higher concentration of aerosols in the low atmosphere between 0.0 and ~1.0 km, there is more radiative heating of air by aerosols in the control-novary run than in the control run and in the control-1500-novary run than in the control-1500 run in the low atmosphere.

Comparisons between the aro-above-cld and aro-above-cld-novary runs (between the aro-above-cld-1500 and aro-above-cld-1500-novary runs) show that due to the transportation of aerosols by downdrafts in the aro-above-cld (aro-above-cld-1500) run, aerosol concentrations in the main aerosol layer in the upper atmosphere reduces and those in the air below the main aerosol layer increases. This is as compared to the situation in the aro-above-cld-noary (aro-above-cld-1500-novary) run where aerosols are assumed not to be affected by cloud-induced wind (Figures 7b and 7d). Note that the upper atmosphere is where cloud-induced updrafts decelerate and turn into downdrafts, and the downward transportation of aerosols by them is dominant. However, those increases in aerosol concentrations in the air below the main aerosol layer mainly occur for the atmosphere between ~1.5 and ~2.5 km and aerosol concentrations in the low atmosphere between 0.0 and ~1.0 km do not change significantly (Figures 7b and 7d). Hence, these transported aerosols by downdrafts do not affect instability in the low atmosphere, which tends to have

485   more impacts on CAPE than instability in other parts of the atmosphere, significantly. This

486   leads to similar instability in the low atmosphere and CAPE, which in turn leads to similar

487   updrafts and cloud mass between the aro-above-cld and aro-above-cld-novary runs and the

488   aro-above-cld-1500 and aro-above-cld-1500-novary runs (Figure 8a). However, due to

489   more aerosols and their more radiative heating of air in the low atmosphere in the control-

490   novary run than in the control run and in the control-1500-novary run than in the control-

491   1500 run, there are higher CAPE (or greater instability), stronger updrafts and higher cloud

492   mass in the control-novary run than in the control run and in the control-1500-novary run

493   than in the control-1500 run (Figure 8a). It is notable that cloud mass in the control-novary

494   run is so large that its maximum value in the vertical profile as shown in Figure 8a exceeds

495   that even in the control-1500-novary run (Figure 8a). Associated with this, there are only

496   ~20 % changes in cloud mass between the control-1500 and control-1500-novary runs,

497   while there are as much as ~200 % changes in cloud mass between the control and control-

498   novary runs. This indicates that as aerosol concentration increases in the low atmosphere,

499   the sensitivity of responses of cloud mass to changes in aerosol concentrations, which are

500   induced by cloud-induced wind, in the low atmosphere increases substantially.

501

502   **3.4 Comparisons between simulations with aerosol radiative effects and those with**

503   **no aerosol radiative effects**

504

505   Figure 8b shows that with no aerosol radiative effects, differences in cloud mass between

506   the control-norad and aro-above-cld-norad runs are much smaller than those differences

507   between the control and aro-above-cld runs with aerosol radiative effects. However, as in

508   the control and aro-above-cld runs, there is higher cloud mass in the control-norad run,

509   which has the main aerosol layer in the low atmosphere, than in the aro-above-cld-norad

510   run, which has the main aerosol layer in the upper atmosphere. Figure 8b shows that cloud

511   mass in each of the control and aro-above-cld runs increases significantly when aerosol

512   radiative effects are turned off. Figure 8b shows that with no aerosol radiative effects,

513   differences in cloud mass between the control-1500-norad and aro-above-cld-1500-norad

514   runs are also smaller than those differences between the control-1500 and aro-above-cld-

515   1500 runs with aerosol radiative effects. However, as in the control-1500 and aro-above-

18

516   cld-1500 runs, there is higher cloud mass in the control-1500-norad run, which has the

517   main aerosol layer in the low atmosphere, than in the aro-above-cld-norad run, which has

518   the main aerosol layer in the upper atmosphere (Figure 8b). Figure 8b shows that cloud

519   mass in each of the control-1500 and aro-above-cld-1500 runs increases when aerosol

520   radiative effects are turned off, although these increases are a lot smaller than those in each

521   of the control and aro-above-cld runs. Here, we see that aerosol radiative effects suppress

522   clouds and reduce cloud mass. This means that effects of aerosol-induced reduction in the

523   surface-reading solar radiation and subsequently in the surface heat fluxes on clouds are

524   dominant over those of radiative heating of air by aerosols, when it comes to the

525   explanation of differences in cloud mass between a situation with aerosol radiative effects

526   and that with no aerosol radiative effects. The suppression of clouds and reduction in cloud

527   mass increase with increasing aerosol concentrations in the main aerosol layer, whether the

528   main layer is in the low atmosphere or in the upper atmosphere, since more aerosols reduce

529   the surface-reaching solar radiation and surface heat fluxes more.

530   More aerosols and their activation (or nucleation of droplets) produce higher cloud

531   droplet number concentration (CDNC) in the low atmosphere in the control-1500-norad

532   run than in the aro-above-cld-1500-norad run. Note that aerosol activation mainly occurs

533   around cloud bases which are located in the low atmosphere. Droplets act as a source of

534   condensation, since individual droplets provide their surface areas onto which water vapor

535   condenses. Hence, higher CDNC induces more condensation and this in turn induces

536   stronger updrafts and more cloud mass in the control-1500-norad run than in the aro-above-

537   cld-1500-norad run. These effects of more aerosols, which induces more condensation and

538   stronger updrafts, are generally referred to as aerosol microphysical effects (Lee et al.,

539   2016). The differences in CDNC are greater due to greater differences in aerosols in the

540   low atmosphere between the control-norad and aro-above-cld-norad runs than those

541   between the control-1500-norad and aro-above-cld-1500-norad runs. This leads to greater

542   aerosol microphysical effects or greater differences in condensation, associated updrafts

543   and cloud mass between the control-norad and aro-above-cld-norad runs than those

544   between the control-1500-norad and aro-above-cld-1500-norad runs. With aerosol

545   radiative effects, radiative heating of air in the low atmosphere works in tandem with

546   aerosol microphysical effects. Hence, it is shown that as compared to the situation with no

547   aerosol radiative effects, with aerosol radiative effects, differences in cloud mass between

548   the run with the main aerosol layer in the low atmosphere and that with the layer in the

549   upper atmosphere are greater, whether aerosol concentrations are low (initially set at 1500

550   cm^{-3}) or high (initial set at 15000 cm^{-3}) in the layer. In addition to the jump in CAPE,

551   updrafts and condensation in the control run as described in Section 3.1, greater aerosol

552   microphysical effects in the low atmosphere when aerosol concentrations are high (initial

553   set at 15000 cm^{-3}) in the aerosol layer contributes to greater differences in cloud mass

554   between the control and aro-above-cld runs than between the control-1500 and aro-above-

555   cld-1500 runs when aerosol concentrations are low (initial set at 1500 cm^{-3}) in the aerosol

556   layer.

557   The initial concentration of aerosols in the aro-above-cld-norad run is identical to that

558   in the aro-above-cld-1500-norad run in the low atmosphere where most of aerosol

559   activation occurs as seen in the description of initial aerosol distribution in Section 2.2.

560   Due to this, in the low atmosphere, CDNC and condensation in the aro-above-cld-norad

561   run are similar to those in the aro-above-cld-1500-norad run. This leads to similar cloud

562   mass between the runs.

563

564   **4.   Summary and conclusions**

565

566   This study examined differential impacts of an aerosol layer on warm cumulus clouds in

567   the Korean Peninsula between a situation where the main aerosol layer is located around

568   or above the tops of clouds in the upper atmosphere and that where the main aerosol layer

569   is located around or below the bottoms of clouds in the low atmosphere. This study finds

570   that the main layer intercepts more solar radiation which reaches the surface when it is in

571   the low atmosphere than when it is in the upper atmosphere. This makes the surface heat

572   fluxes and associated CAPE lower, which tend to make updrafts weaker and make cloud

573   mass lower when the main aerosol layer is in the low atmosphere. However, with the main

574   aerosol layer in the low atmosphere, there is a greater amount of cloud mass than that with

575   the main layer in the upper atmosphere. This is because the main layer in the low

576   atmosphere heats up the air there more, leading to increases in the instability and CAPE as

577   compared to the situation when the main layer is in the upper atmosphere. These increases

578   in CAPE are larger than reduction in CAPE due to the reduced solar radiation reaching the

579   surface, resulting in more cloud mass when the main layer is in the low atmosphere than

580   when the main layer is in the upper atmosphere.

581   With decreasing concentrations of aerosols in the main aerosol layer, there are

582   decreases in the interception of solar radiation reaching the surface, increases in surface

583   heat fluxes, CAPE and cloud mass whether the main layer is in the low atmosphere or in

584   the upper atmosphere. However, the decreasing concentrations of aerosols cause the jump

585   in instability as seen in the evolution of CAPE to disappear when the main layer is in the

586   low atmosphere. This leads to reducing differences in cloud mass between the situation

587   with the main layer in the low atmosphere and that with the layer in the upper atmosphere.

588   When the main aerosol layer is in the low atmosphere, with increasing aerosol

589   concentrations in the layer, the lifetime of cloud system reduces in a way that the lifetime

590   with the main layer in the low atmosphere gets shorter than that with the layer in the upper

591   atmosphere.

592   Updrafts and downdrafts in clouds transport aerosols. In particular, for the main aerosol

593   layer in the low atmosphere, updrafts transport aerosols in the main layer to places above

594   it. This reduces aerosol concentrations in the main layer, leading to reduction in radiative

595   heating of air by aerosols, CAPE, updrafts and cloud mass. This reduction enhances with

596   increasing aerosol concentrations in the main layer. For the aerosol layer in the upper

597   atmosphere, downdrafts transport aerosols in the layer to places below it. However, this

598   does not affect aerosol concentrations, radiative heating of air in the low atmosphere

599   significantly. This in turn does not affect CAPE and cloud mass significantly.

600   Aerosol radiative effects suppress clouds and reduce cloud mass by reducing solar

601   radiation which reaches the surface as compared to a situation when there are no aerosol

602   radiative effects. This suppression of clouds increases with increasing aerosol

603   concentrations in the main aerosol layer. Aerosol microphysical effects enhance cloud

604   mass and these effects are stronger with higher aerosol concentrations in the main layer.

605   When aerosol radiative effects, which are in terms of radiative heating of air by aerosols,

606   and aerosol microphysical effects work together, differences in cloud mass between a

607   situation where the main layer is in the upper atmosphere and that where the main layer is

608   in the low atmosphere enhance as compared to a situation where only aerosol

609 microphysical effects are present. More aerosols, and thus stronger radiative heating of air

610 and stronger aerosol microphysical effects in the main aerosol layer in the low atmosphere

611 enable this enhancement to be larger when aerosol concentrations are high in the main layer

612 than they are low.

613   This study shows that radiative heating of air by aerosols in the low atmosphere, which

614 are around or below cloud bases, enhances instability, invigorates convection and increases

615 cloud mass, which is contrary to the conventional wisdom of impacts of absorbing aerosols

616 on convection. However, radiative heating of air by aerosols in the upper atmosphere,

617 which are around or above cloud tops, enhances stability, suppresses convection and

618 reduces cloud mass. Aerosols in the low atmosphere intercept more solar radiation reaching

619 the surface, which tend to suppress the surface fluxes and convection, than aerosols in the

620 upper atmosphere. Here, we see that aerosol-induced changes in the surface fluxes and

621 those in radiative heating of air interact with each other in terms of responses of convection

622 and clouds to aerosols. This interaction varies with the varying vertical location of aerosols

623 and the varying cloud-induced wind that is at cloud scale. In general, traditional

624 parameterizations for warm cumulus clouds in climate and weather-forecast models have

625 not been able to consider this dependence of the interaction on the vertical location of

626 aerosols, since in general, those parameterizations for warm clouds do not differentiate

627 aerosol layers based on their vertical locations. In addition, the cloud-scale cloud-induced

628 wind, which is not able to be resolved by general resolutions in climate and weather-

629 forecast models, have not been represented by those parameterizations with good

630 confidence. So, impacts of aerosol transportation by cloud-induced wind on the interaction

631 have not been properly considered in those traditional parameterizations. Results here

632 demonstrate that for more comprehensive representation of interactions between warm

633 cumulus clouds and aerosols, we need to develop a more comprehensive parameterization

634 that is able well represent the varying interaction between aerosol-induced changes in the

635 surface fluxes and those in radiative heating of air with varying vertical locations of

636 aerosols and aerosol transportation by cloud-induced wind.

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Code/Data source and availability

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642 Our private computer system stores the code/data which are private and used in this study.

643 Upon approval from funding sources, the data will be opened to the public. Projects related

644 to this paper have not been finished, thus, the sources prevent the data from being open to

645 the public currently. However, if information on the data is needed, contact the

646 corresponding author Seoung Soo Lee (slee1247@umd.edu).

647

648 **Author contributions**

649 Essential initiative ideas are provided by SSL, JU and WJC to start this work. Simulation

650 and observation data are analyzed by SSL, JU and KJH. CHJ. JG and YZ review the results

651 and contribute to their improvement.

652

653 **Competing interests**

654 The authors declare that they have no conflict of interest.

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810 **FIGURE CAPTIONS**
811
812 Figure 1. An inner rectangle in the map of the Korean Peninsula represents the simulation
813 domain. The green (light blue) represents land (ocean) area in the map.

814
815 Figure 2. Vertical distributions of potential temperature and water-vapor mixing ratio at
816 09:00 LST on April 15th, 2015. These distributions are obtained from radiosonde sounding
817 near the simulation domain in Figure 1.

818
819 Figure 3. Vertical distributions of the area-averaged aerosol concentrations at the first time
820 step of (a) the control run and (b) the aro-above-cld run.

821
822 Figure 4. Vertical distributions of the time- and area-averaged cloud-liquid mass density
823 that represents cloud mass for the standard simulations (i.e., the control, aro-above-cld,
824 control-1500 and aro-above-cld-1500 runs).

825
826 Figure 5. Time series of the domain-averaged (a) liquid-water path, (b) updraft speed, (c)
827 condensation rate and (d) convective available potential energy in the standard simulations.

828
829 Figure 6. Vertical distributions of the time- and area-averaged radiative heating rate (a) in
830 the control and aro-above-cld runs over the initial stage between 10:00 and 13:50 LST, (b)
831 in the control and aro-above-cld runs over the whole simulation period and (c) in the
832 control-1500 and aro-above-cld-1500 runs over the whole simulation period.

833
834 Figure 7. Vertical distributions of the time- and area-averaged aerosol concentrations (a)
835 in the control and control-novary runs, (b) aro-above-cld and aro-above-cld-novary runs,
836 (c) control-1500 and control-novary-1500 runs and (d) aro-above-cld-1500 and aro-above-
837 cld-novary-1500 runs.

838
839 Figure 8. Vertical distributions of the time- and area-averaged cloud-liquid mass density.
840 In (a), the control-novary, aro-above-cld-novary, control-1500-novary and aro-above-cld-

841   1500-novary runs and in (b), the control-norad, aro-above-cld-norad, control-1500-norad

842   and aro-above-cld-1500-norad runs are shown together with the standard simulations.

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Simulations	Altitudes of a main aerosol layer (km)	Aerosol concentrations in the main aerosol layer at the first time step (cm^{-3})	Aerosol evolution	Aerosol radiative effects
Control	0 - 1	15000	Present	Present
Aro-above-cld	2.5-3.5	15000	Present	Present
Control-1500	0 - 1	1500	Present	Present
Aro-above-cld-1500	2.5-3.5	1500	Present	Present
Control-novary	0 - 1	15000	Absent	Present
Aro-above-cld-novary	2.5-3.5	15000	Absent	Present
Control-1500-novary	0 - 1	1500	Absent	Present
Aro-above-cld-1500-novary	2.5-3.5	1500	Absent	Present
Control-norad	0 - 1	15000	Present	Absent
Aro-above-cld-norad	2.5-3.5	15000	Present	Absent
Control-1500-norad	0 - 1	1500	Present	Absent
Aro-above-cld-1500-norad	2.5-3.5	1500	Present	Absent

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873   Table 1. Summary of simulations

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Simulations	Net solar radiation flux reaching the surface (W m^{-2})	Surface latent heat fluxes (W m^{-2})	Surface sensible heat fluxes (W m^{-2})	Surface latent heat fluxes plus surface sensible heat fluxes (W m^{-2})
Control	293 (205)	175 (120)	22 (16)	197 (136)
Aro-above-cld	306 (217)	170 (117)	48 (33)	218 (150)
Control-1500	461	250	70	320
Aro-above-cld-1500	467	248	75	323

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887 Table 2. The time- and area-averaged net solar radiation, latent heat, sensible heat and total

888 heat (sensible plus latent heat) fluxes at the surface over the whole simulation period in the

889 standard simulations. Numbers in the parentheses are averaged over the initial period

890 between 10:00 and 13:50 LST for control and aro-above-cld runs.
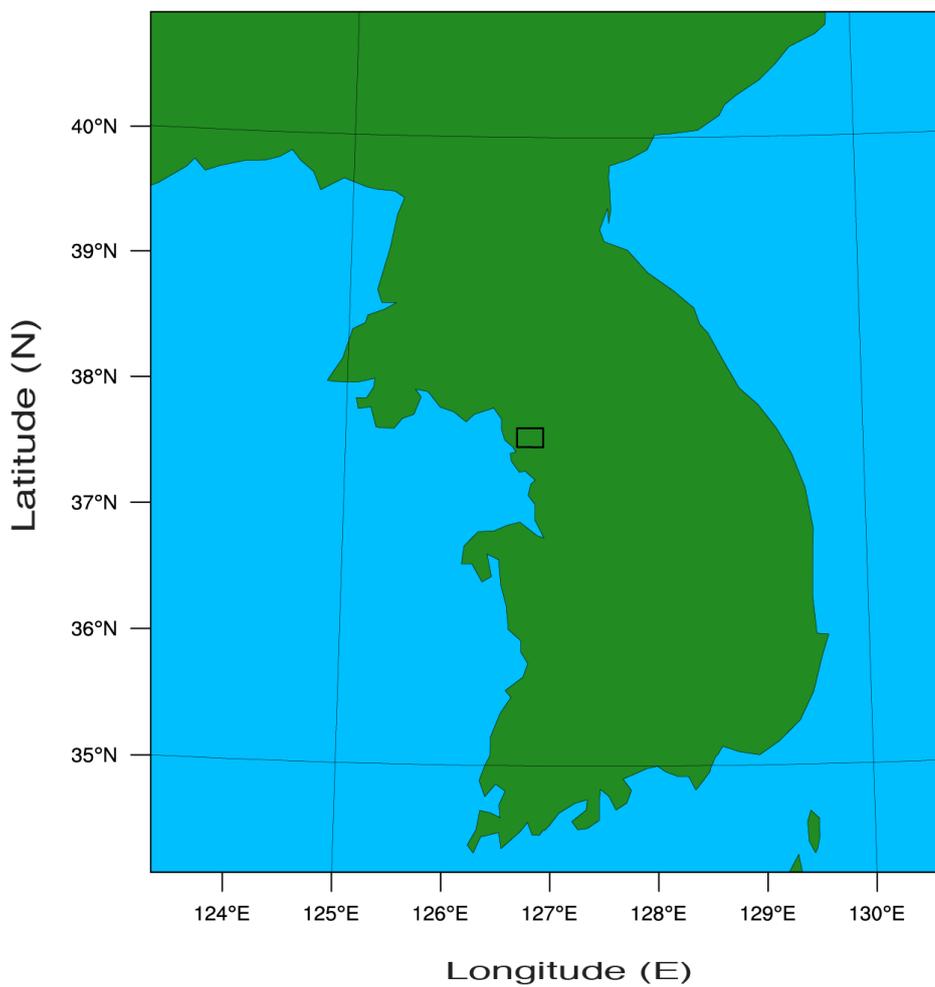
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Figure 1

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Figure 2

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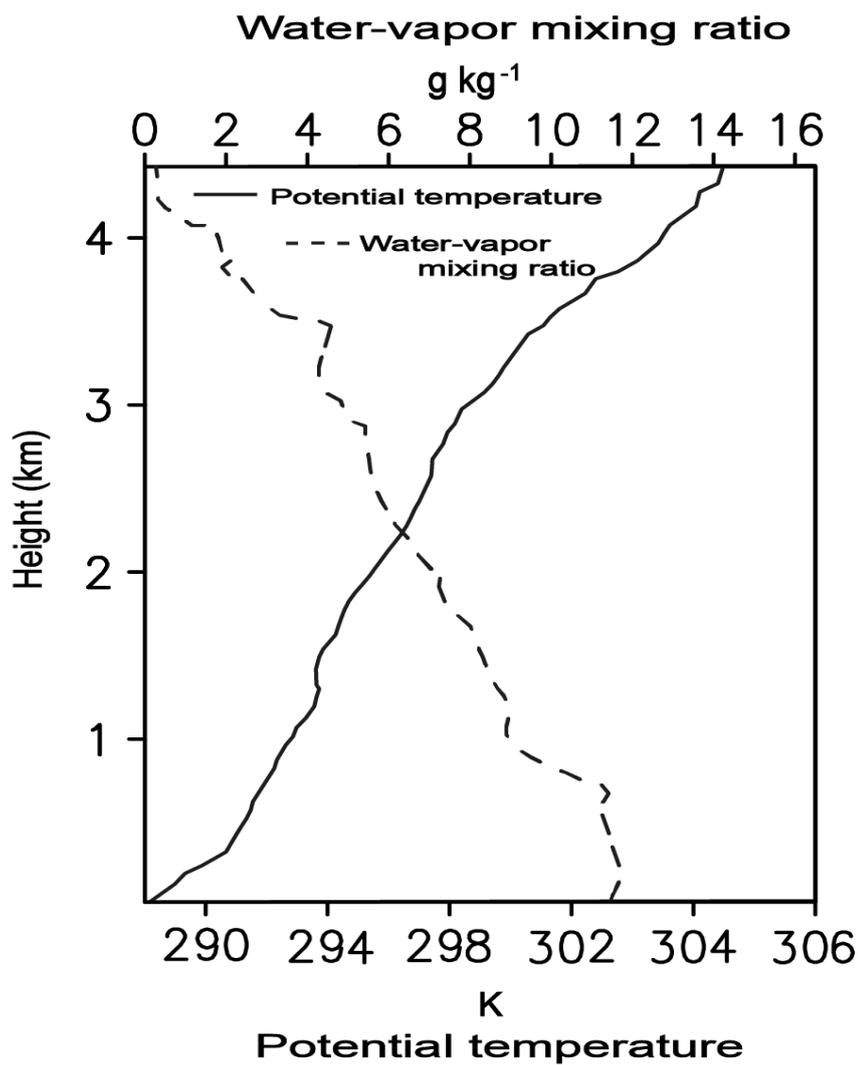
Vericial distributions of aerosol concentrations

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905 **Figure 3**

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Cloud mass

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Figure 4

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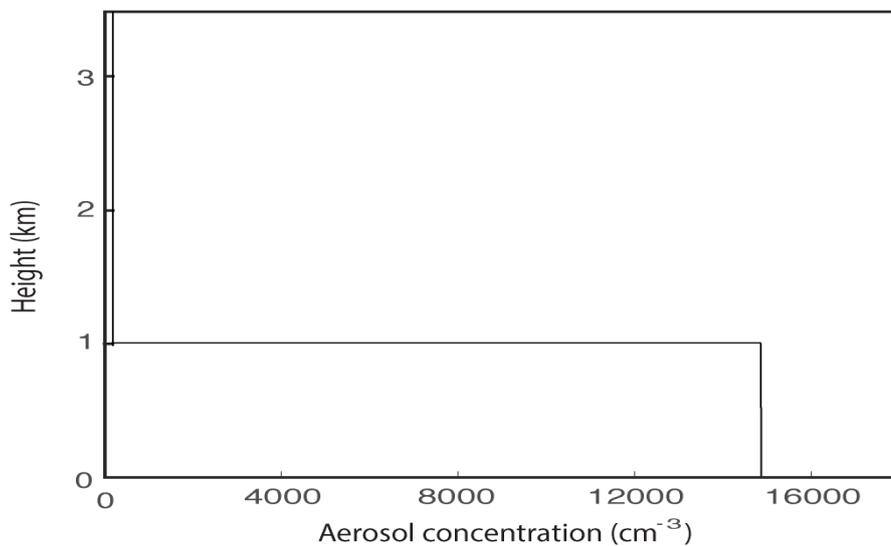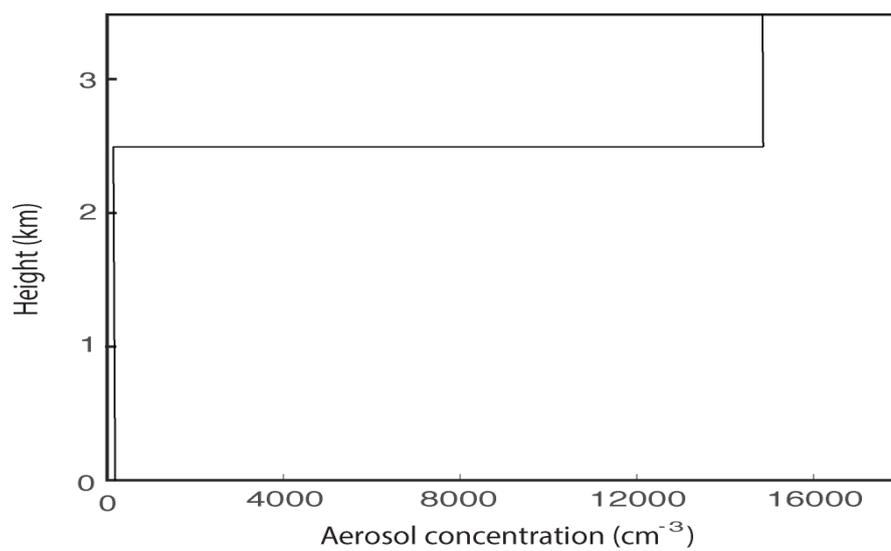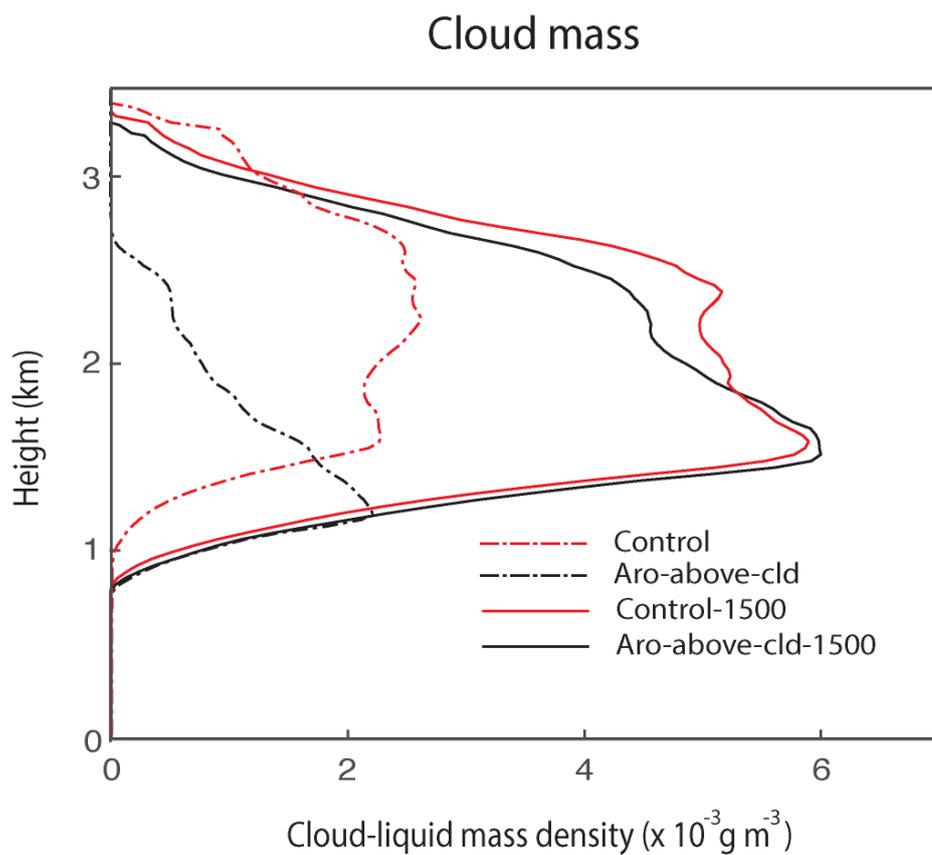
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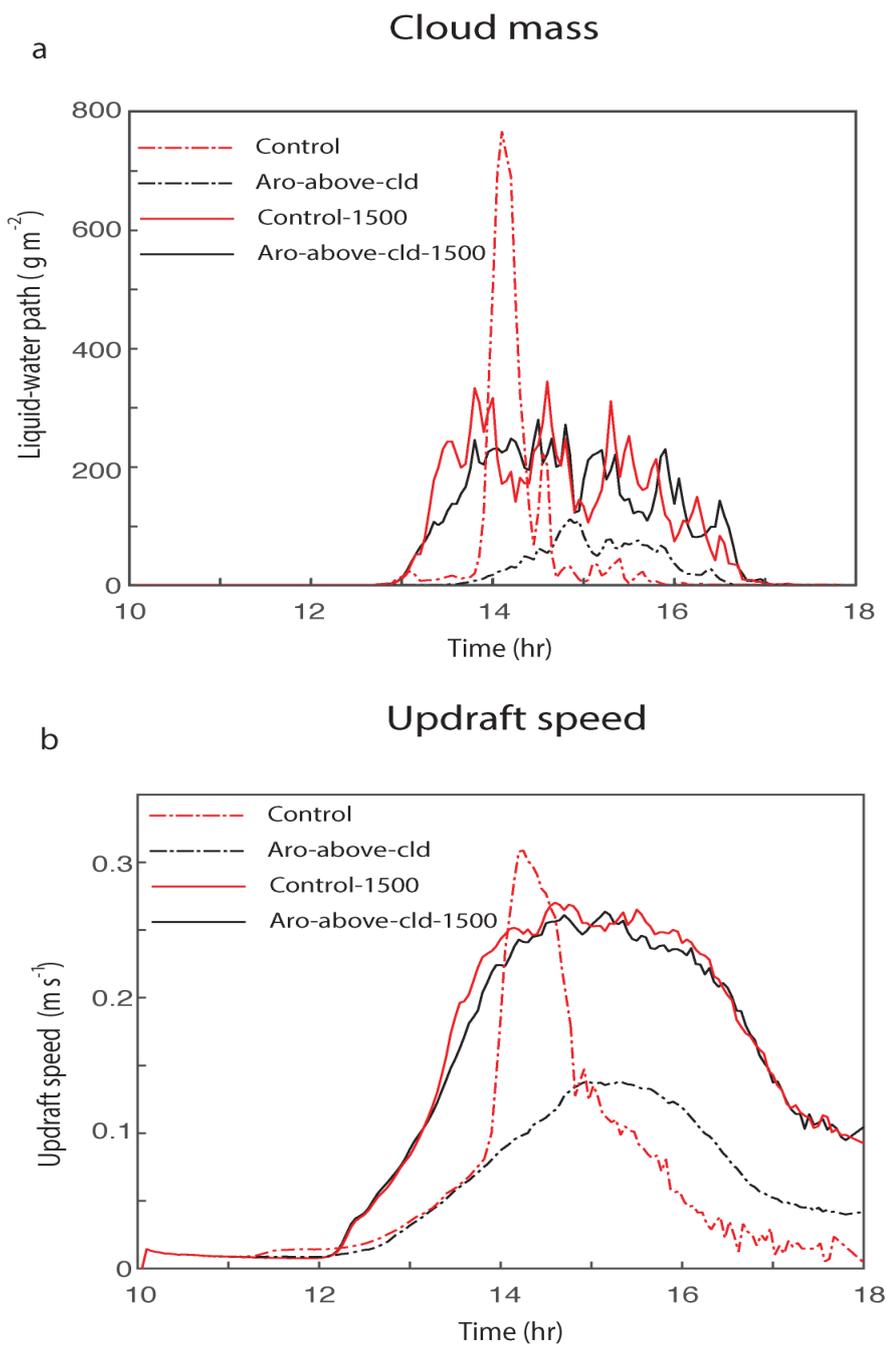
Figure 5

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Figure 5

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Figure 6

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Vertical distributions of aerosol concentrations

Figure 7

c

d

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928   **Figure 7**
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Cloud mass

a

b

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Figure 8