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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Abstract

Effects of an aerosol layer on warm cumulus clouds in the Korean Peninsula when the layer is above or around the cloud tops in the free atmosphere are compared to those effects when the layer is around or below the cloud bases in the planetary boundary layer (PBL). For this comparison, simulations are performed using the large-eddy simulation framework. When the aerosol layer is in the PBL, aerosols absorb solar radiation and radiatively heat up air enough to induce greater instability, stronger updrafts and more cloud mass than when the layer is in the free atmosphere. Hence, there is a variation of cloud mass with the location (or altitude) of the aerosol layer. It is found that this variation of cloud mass reduces, as aerosol concentrations in the layer decrease or aerosol impacts on radiation are absent. The transportation of aerosols by updrafts reduces aerosol concentrations in the PBL. This in turn reduces the aerosol radiative heating, updraft intensity and cloud mass.

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). Aerosols act as radiation absorbers, and they absorb solar radiation and heat up the atmosphere to change atmospheric stability. This in turn affects thermodynamics in cumulus clouds (Hansen et al., 1997). When these aerosols act as cloud condensation nuclei (CCN), they have an impact on aerosol activation and subsequent microphysical processes in cumulus clouds (Albrecht, 1989). However, these aerosol effects on warm cumulus clouds are highly uncertain and thus cause the highest uncertainty in the prediction of future climate (Ramaswamy et al., 2001; Forster et al., 2007).

In recent years, people have started to take interest in how aerosol layers affect clouds when these layers are above or around the tops of 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 (Mari al., 2008; Menut et al., 2018; Haslett et al., 2019; Denjean et al., 2020). These layers are lifted and transported to the southeast Atlantic (SEA) region and located above or around the top of a large layer or deck of warm cumulus and stratocumulus clouds (Roberts et al., 2009; van der Werf et al., 2010; Che et al., 2022). Note that aerosols in the transported aerosol layers contain organic and black carbon, and these aerosols act as radiation absorbers as well as CCN (Wilcox, 2010; Deaconu et al., 2019; Chaboureau et al., 2022). Reflecting the interest, to better understand roles of aerosol layers above or around cloud tops in cloud development, there were international field campaigns in the SEA such as the National Aeronautics and Space Administration ObseRvations of Aerosols above CLouds and their intEractionS (ORACLES; https://espo.nasa.gov/oracles/content/ORACLES), the United Kingdom Clouds and Aerosol Radiative Impacts and Forcing (CLARIFY; Redemann et al., 2021) and the French Aerosol, Radiation and Clouds in southern Africa (AEROCLO-sA; Formenti et al., 2019) campaigns.

Despite above-mentioned field campaigns, effects of aerosols above or around tops of warm cumulus clouds, which are induced by shallow convection, have not been examined as much as those of aerosols around or below bottoms of those clouds (Haywood and Shine,

114 1997; Johnson et al., 2004; McFarquhar and Wang, 2006). Motivated by this, this study

115 delves into effects of not only aerosols around or below bottoms of warm cumulus clouds

116 but also those above or around tops of those clouds. Through this, this study aims to

117 contribute to the more comprehensive understanding of aerosol-radiation-cloud

118 interactions. This more comprehensive understanding in turn contributes to more general

119 parameterizations of those interactions for climate and weather-forecast models. To fulfill

120 the aim, this study adopts the large-eddy simulation (LES) framework and an idealized

121 setup for the aerosol layer.

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

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125 **2.1 LES model**

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

128 simulations in this study. The ARW adopts a 50-m resolution for the horizontal domain. In

129 the vertical domain, the resolution coarsens with height. The resolution in the vertical

130 domain is 20 m just above the surface and 100 m at the model top. The ARW model is a

131 compressible model with a nonhydrostatic status. A 5th-order monotonic advection scheme

132 is used to advect microphysical variables (Wang et al., 2009). The ARW adopts a bin

133 scheme, which is detailed in Khain et al. (2011), to parameterize microphysics. A set of

134 kinetic equations is solved by the bin scheme to represent size distribution functions for

135 each class of hydrometeors and aerosols acting as cloud condensation nuclei (CCN). The

136 hydrometeor classes are water drops, ice crystals (plate, columnar and branch types), snow

137 aggregates, graupel and hail. There are 33 bins for each size distribution in a way that the

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

139   Aerosol sinks and sources, which include aerosol advection and activation, control

140 the evolution of aerosol size distribution at each grid point. For example, activated particles

141 are emptied in the corresponding bins of the aerosol spectra. Aerosol mass included in

142 hydrometeors, after activation, is moved to different classes and sizes of hydrometeors

143 through collision-coalescence and removed from the atmosphere once hydrometeors that

144 contain aerosols reach the surface.

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

146   to the bin microphysics scheme. Aerosols before their activation can affect radiation by

147   changing the reflection, scattering, and absorption of radiation. This radiative effect of

148   aerosol is represented following Feingold et al. (2005). The internal aerosol mixture and

149   the ARW relative humidity are used to calculate the hygroscopic growth of the aerosol

150   particles as well as their optical properties. In practice, optical property calculations with

151   the consideration of the hygroscopic growth are performed offline prior to simulation and

152   stored in lookup tables. Calculations are done for the prescribed aerosol size distribution

153   and composition, and unit concentration. During model runtime, grid-point number

154   concentration and relative humidity determine the look-up table entries that specify the

155   grid-point aerosol optical properties and are fed into the RRTM to simulate the radiative

156   effect of aerosol. The effective sizes of hydrometeors are calculated in the bin scheme and

157   the calculated sizes are transferred to the RRTM to consider effects of the effective sizes

158   on radiation.

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

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

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

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163   **2.2 Case and simulations**

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165   **2.2.1 Case and standard simulations**

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167 As a case study, we simulate an observed system of warm cumulus clouds in a domain in

168 the Korean Peninsula on April 13th, 2016. The domain is marked in Figure 1a. Figure 2

169 shows the field of the cloud reflectivity observed by the Communication, Ocean, and

170 Meteorological Satellite (COMS). This field is at 14:00 LST on April 13th, 2016 when the

171 system is around the mature stage in the domain. The ratio of the reflected radiative flux

172 by an object to the incident radiative flux on it is the reflectivity (Liou, 2002) and thus

173 unitless. In Figure 2, we see cloud cells that are elongated in the southwest-northeast

174 direction due to the southwesterly wind.

175   The simulation is performed for a period between 10:00 and 18:00 LST on April 13th,

176   2016. This period includes a time span over which the system exists. For the simulation

177   (i.e., the control run), the length of the domain in both the east-west and north-south

178   directions is 20 km and the model top is at ~4.5 km in altitude. The time step or temporal

179   resolution is set at 0.1 second. Initial and boundary conditions of potential temperature,

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

181   represent the synoptic-scale environment and are produced by the Met Office Unified

182   Model (Brown et al., 2012) every 6 hours on a 0.11° × 0.11° grid. Figure 3 depicts the

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

184   on April 13th, 2016 in radiosonde sounding that is obtained near the domain as marked in

185   Figure 1a. This vertical distribution represents initial environmental conditions for the

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

187   development of warm cumulus clouds. An open lateral boundary condition is employed

188   for the run.

189   Not only a site of the aerosol robotic network (AERONET; Holben et al., 2001) but

190   also ground stations that measure $PM_{2.5}$ are in the domain as marked in Figure 1b. The mass

191   of aerosols with diameter smaller than 2.5 μm per unit volume of the air is $PM_{2.5}$. Around

192   07:00 LST on April 13th, 2016, an aerosol layer advected from East Asia starts to be present

193   in the domain. This advection of aerosols is monitored and identified by $PM_{2.5}$ which is

194   measured by stations in the Yellow sea and domain (Eun et al., 2016; Ha et al., 2019; Lee

195   et al., 2021). The station in the Yellow sea is marked in Figure 1a. Figure 4 shows the

196   evolution of $PM_{2.5}$ at the station in the Yellow sea and the average $PM_{2.5}$ over stations in

197   the domain from 03:00 LST to 18:00 LST on April 13th, 2016. Due to the aerosol-layer

198   advection from East Asia, aerosol mass starts to increase around 04:00 LST and reaches its

199   peak around 08:00 LST at the station in the sea. Then, in the domain, aerosol mass starts

200   to increase around 07:00 LST, and the mass attains its peak around 11:00 LST. This depicts

201   a situation where aerosols or an aerosol layer advected from East Asia first arrives at the

202   station in the Yellow sea around 04:00 LST and then further advected to the east to reach

203   the domain and to start the increase in aerosol mass there around 07:00 LST.

204   According to the AERONET measurement at 12:00 LST, which is ~1 hour before

205   the observed cumulus clouds start to form, aerosol particles in the advected aerosol layer,

206 on average, are an internal mixture of 70 % ammonium sulfate, 22 % organic compound

207 and 8% black carbon. Aerosol chemical composition in this study is assumed to be

208 represented by this mixture in the whole domain during the whole simulation period. Based

209 on the AERONET observation, the shape of the initial size distribution of aerosols acting

210 as CCN is assumed to follow a bi-modal log-normal distribution as shown in Figure 5 in

211 all parts of the domain. Modal radius of this distribution is 0.11 and 1.20 μm and standard

212 deviation of this distribution is 1.71 and 1.92, while the partition of aerosol number, which

213 is normalized by the total aerosol number of the size distribution, is 0.999 and 0.001 for

214 accumulation and coarse modes, respectively. The total aerosol number concentration in

215 the advected aerosol layer based on the AERONET-observed size distribution is ~15000

216 cm^{-3}. This concentration is applied to all grid points in the aerosol layer at the first time

217 step of the control run. This aerosol layer is idealized to be located around or below cloud

218 bases between the surface and 1.0 km in the planetary boundary layer (PBL). Cloud bases

219 are located around 1.0 km. At 06:00 LST, ~1 hour before the advected aerosol layer starts

220 to be present, the AERONET-measured aerosol concentration is ~150 cm^{-3} in the domain.

221 This aerosol concentration is assumed to be a background aerosol concentration that is not

222 affected by the advected aerosol layer. Based on this assumption, the initial aerosol

223 concentration is set at 150 cm^{-3} outside the layer.

224 This study compares aerosol effects on warm cumulus clouds when the aerosol layer

225 is above or around the cloud tops to those effects when the layer is around or below the

226 cloud bases. For this, we repeat the control run by moving the aerosol layer upward to

227 altitudes between 2.5 and 3.5 km in the free atmosphere which is above the PBL. Here,

228 initial aerosol concentrations in and outside the aerosol layer are 15000 cm^{-3} and 150 cm^{-}

229 ^{3}, respectively, in both of the runs. Altitudes between 2.5 and 3.5 km are places where cloud

230 tops are located frequently and the simulated maximum cloud-top height is 3.3 km. This

231 repeated run is referred to as the aro-above-cld run.

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

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

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

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

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

237   outside the layer. In these repeated runs, the aerosol concentration in the aerosol layer at

238   the first time step is 1500 cm^{-3}. Henceforth, the repeated control and aro-above-cld runs

239   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 through 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. Motivated

246   by this, we take interest in impacts of clouds on aerosols and how these impacts in turn

247   change the influence of aerosols on clouds. To examine this aspect of aerosol-cloud

248   interactions, the above-mentioned four standard simulations (i.e., the control, aro-above-

249   cld, control-1500 and aro-above-cld-1500 runs) are repeated. In these repeated runs,

250   aerosol concentrations at each grid point, which are set at the first time step, do not vary

251   with time or are not affected by cloud processes. These repeated runs are referred to as the

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

253   novary runs. By comparing the standard simulations to these repeated ones, we aim to

254   identify how cloud processes affect the aerosol layer and then the impacts of the layer on

255   clouds.

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

257   scattering and absorption) of radiation by aerosols in impacts of the aerosol layer on clouds.

258   This interception of radiation by aerosols, which is referred to as aerosol radiative effects,

259   results in phenomena such as radiative heating of air by aerosols. To better understand roles

260   of aerosol radiative effects, the above four standard simulations are repeated again by

261   turning off aerosol radiative effects. These repeated runs are the control-norad, aro-above-

262   cld-norad, control-1500-norad, aro-above-cld-1500-norad runs. The summary of

263   simulations in this study is given in Table 1.

264

265   **3. Results**

266

267   **3.1 The control and aro-above-cld runs**

268

269   Figure 6 depicts the simulated field of the cloud reflectivity at 14:00 LST on April 13th,

270   2016 in the control run. Similar to the observed counterpart in Figure 2, simulated cloud

271   cells are elongated in the southwest-northeast direction. Also, there is a good consistency

272   in the overall cell size and population and the overall pattern of the spatial distribution of

273   cloud cells between the observed and simulated fields. Table 3 shows comparisons of cloud

274   and environmental variables between observation and the control run. Observation is

275   performed by ground stations and satellites. Note that ground stations which measure $PM_{2.5}$

276   as marked in Figure 1b also measure cloud and environmental variables. Table 3 shows

277   that differences in those variables between observation and the control run are ~10%. This

278   and Figure 6 indicate that the control run can be considered performed reasonably well.

279   Figure 7 shows the time- and area-averaged vertical distributions of cloud-liquid mass

280   density for the standard simulations. In Figure 7, the cloud layer is between 1.0 and 3.3 km

281   in the control run and between 0.8 and 2.6 km in the aro-above-cld run. The time- and

282   domain-averaged cloud-liquid mass density is 0.7 and 1.3×10^{-3} g m^{-3} in the control run

283   and in the aro-above-cld run, respectively. Hence, we see that clouds are thicker with their

284   higher tops and have greater mass in the control run than in the aro-above-cld run.

285   Figure 8a shows the time series of the domain-averaged liquid-water path, which is

286   the vertical integral of cloud-liquid mass density, for the standard simulations. During the

287   initial stage of the cloud development between 12:50 and 13:50 LST, the average cloud

288   mass is slightly higher in the control run than in the aro-above-cld run. Also, the average

289   non-zero cloud mass starts to appear earlier in the control run. Over the period between

290   13:50 and 14:10 LST, there is a jump (or rapid increase or surge) in the average cloud mass

291   in the control run but not in the aro-above-cld run. During this period with the jump, at

292   some specific time points, the average mass is ~one order of magnitude higher in the

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

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

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

296   mainly attributed to the jump. Figures 8b and 8c show the time series of the domain-

297   averaged updraft speed and condensation rates, respectively. These figures indicate that the

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

299   slightly higher than in the aro-above-cld run for the period between 12:50 and 13:50 LST.

300   The average updraft speed and associated condensation rates jump and thus are much

301   higher in the control run during the period between ~13:50 and ~14:10 LST (Figures 8b

302   and 8c). After the jump, the speed and rates decrease rapidly and become lower in the

303   control run (Figures 8b and 8c). Condensation is the only source of cloud mass in warm

304   cumulus clouds. Also, updrafts with higher speeds tend to produce higher condensation

305   rates for a given environmental condition. Hence, cloud mass, condensation rate and the

306   updraft speed are closely linked to each other. This enables cloud mass, condensation rate

307   and the updraft speed to be similar in terms of their temporal evolution in each of the

308   control and aro-above-cld runs (Figures 8a, 8b and 8c).

309   Figure 8d shows the time series of the domain-averaged convective available potential

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

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

312   of the buoyancy energy in the vertical domain. Hence, the evolution of CAPE is similar to

313   that of the updraft speed, associated condensation rates and cloud mass (Figure 8). This

314   involves the jump not only in CAPE but also in those speed, rates and mass in the control

315   run.

316   In Figure 8, the peaks (or the maximum values) of the domain-averaged CAPE, the

317   updraft speed, condensation rates and cloud mass in the control run occur around 14:10

318   LST and this occurrence is earlier than that which occurs around 14:50 LST in the aro-

319   above-cld run. This means that the cloud system in the control run reaches its mature stage

320   earlier. Immediately after the peak around 14:10 LST, the system enters its dissipating

321   stage in the control run. However, the system enters its dissipating stage after 14:50 LST

322   in the aro-above-cld run. Hence, the cloud system in the control run matures and demises

323   faster. Stated differently, the cloud system in the control run has a shorter life cycle.

324   To find mechanisms controlling the jump in CAPE which is a main cause of the greater

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

326   10:00 LST and 13:50 LST which is immediately before the jump starts to occur. The

327   average net shortwave fluxes at the surface are shown in Table 2 for the initial period in

328   the control and aro-above-cld runs. Table 2 shows that during the initial period, there is a

329   smaller amount of the surface-reaching shortwave radiation in the control run than in the

330 aro-above-cld run. The aerosol layer intercepts solar radiation and reduces the surface-
331 reaching solar radiation. In spite of the fact that the initial depth of the aerosol layer and
332 aerosol concentrations in the layer are identical between the runs, results here indicate that
333 the aerosol layer in the atmosphere around or below cloud bases is more efficient in the
334 interception of solar radiation than that in the atmosphere around or above cloud tops. Due
335 to the less solar radiation reaching the surface, the time- and area-averaged net surface heat
336 fluxes, which are the sum of the surface sensible and latent-heat fluxes, become lower in
337 the control run during the initial period (Table 2). Hence, the surface fluxes favor more
338 instability or higher CAPE and associated subsequent more intense updrafts and more
339 cloud mass in the aro-above-cld run.

340   The vertical distributions of the time- and domain-averaged radiative heating rates are
341 obtained for the initial period. For the initial period, the average radiative heating rate is
342 much higher in the control run than in the aro-above-cld run particularly at altitudes
343 between 0.0 and ~1.0 km where cloud bases are located (Figure 9a). This is associated with
344 the fact that the aerosol layer is located at altitudes between 0.0 and 1.0 km in the control
345 run. This more radiative heating in the PBL during the initial period results in the
346 subsequent jump in CAPE, associated higher CAPE, more intense updrafts and more cloud
347 mass after the initial period by outweighing the lower surface heat fluxes in the control run.
348 The aerosol layer is located at altitudes between 2.5 and 3.5 km, hence, the average
349 radiative heating rate is higher around those altitudes in the aro-above-cld run (Figures 9a
350 and 9b). However, this higher radiative heating rate is in the upper part of the domain and
351 tends to induce more stabilization of the atmosphere in the aro-above-cld run. Thus, the
352 higher radiative heating rate in the aro-above-cld run contributes to lower CAPE, less
353 intense updrafts and less cloud mass in the aro-above-cld run especially for the period when
354 the jumps occur in the control run.

355

356 **3.2 Comparisons between simulations with different aerosol concentrations**

357

358 With the lower concentration of aerosols in the aerosol layer, there are the much more
359 surface-reaching solar radiation and resultant higher surface fluxes in the control-1500 run
360 than in the control run and in the aro-above-cld-1500 run than in the aro-above-cld run

361   (Table 2). This induces higher CAPE, stronger updrafts and more condensation and cloud

362   mass in the control-1500 run than in the control run over most of the simulation period

363   except for the period with the jump in CAPE in the control run, and in the aro-above-cld-

364   1500 run than in the aro-above-cld run throughout the simulation period (Figure 8). This

365   leads to the greater time- and domain-averaged cloud mass in the control-1500 run than in

366   the control run and in the aro-above-cld-1500 run than in the aro-above-cld run (Figure 7).

367   Regarding the control and control-1500 runs, this is despite the fact that aerosol radiative

368   heating in the PBL is higher due to higher aerosol concentrations there in the control run

369   than in the control-1500 run (Figure 9). Regarding the aro-above-cld-1500 and the aro-

370   above-cld runs, the greater time- and domain-averaged cloud mass is contributed by lower

371   aerosol concentrations and less aerosol radiative heating in the free atmosphere in the aro-

372   above-cld-1500 run than in the aro-above-cld run (Figure 9). Figure 7 shows that the time-

373   and domain-averaged cloud mass in the aro-above-cld-1500 run is higher than in the

374   control run. This is due to more solar radiation reaching the surface in the aro-above-cld-

375   1500 run (Table 2). The higher average cloud mass in the aro-above-cld-1500 run is despite

376   higher aerosol concentrations and more aerosol radiative heating not only in the PBL in the

377   control run, but also in the free atmosphere in the aro-above-cld-1500 run (Figure 9). Figure

378   7 also shows that the time- and domain-averaged cloud mass in the control-1500 run is

379   higher than in the aro-above-cld run. This is associated with the fact that more solar

380   radiation reaches the surface in the control-1500 run than in the aro-above-cld run (Table

381   2). The higher average cloud mass in the control-1500 run is also associated with higher

382   aerosol concentrations and more aerosol radiative heating not only in the PBL in the

383   control-1500 run, but also in the free atmosphere in the aro-above-cld run (Figure 9).

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

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

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

387   run. However, overall, CAPE is higher and cloud mass is greater in the control-1500 run

388   than in the aro-above-cld-1500 run (Figures 7, 8a and 8d). This is because similar to the

389   situation between the control and aro-above-cld runs, aerosols heat up the PBL more in the

390   control-1500 run and the free atmosphere more in the aro-above-cld-1500 run (Figure 9c).

391   The CAPE evolution shows that there is no jump in CAPE and thus updrafts in the control-

392   1500 run (Figures 8b and 8d). This mainly contributes to smaller differences in CAPE,

393   updrafts, condensation and cloud mass between the control-1500 and aro-above-cld-1500

394   runs than between the control and aro-above-cld runs (Figures 7 and 8).

395   In the control run, the instability or CAPE accumulates or increases rapidly to reach

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

397   increases gradually to reach its peak from ~12:00 LST to ~14:30 LST (Figure 8d). For a

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

399   value around ~13:50 LST in the control run. However, CAPE decreases gradually and

400   never drops back to the CAPE value at ~12:00 LST until the end of the simulation period

401   in the control-1500 run. This leads to the shorter life cycle or lifetime of the system in the

402   control run than in the control-1500 run as well as in the aro-above-cld run. Accompanying

403   this is the similar life cycle between the control-1500 and aro-above-cld-1500 runs. Here,

404   we see that as aerosol concentration increases in the aerosol layer in the atmosphere around

405   or below cloud bases, the time scale of the accumulation and consumption of the instability

406   or convective energy gets shorter, leading to the shorter lifetime of the cloud system.

407

408   **3.3 Comparisons between simulations with predicted and prescribed aerosol**

409   **concentrations**

410

411   Figure 10 shows the vertical distributions of aerosol concentrations, which are averaged

412   over the horizontal domain and simulation period, for the standard and repeated runs with

413   no temporal variation of aerosols. Comparisons between the control and control-novary

414   runs and between the control-1500 and control-1500-novary runs show that due to the

415   upward transportation of aerosols by updrafts, aerosol concentrations in the aerosol layer

416   in the PBL reduces and those in the air above the layer increases (Figures 10a and 10c).

417   Note that the PBL is where cloud-induced updrafts develop and grow, hence, the upward

418   transportation of aerosols by them is dominant. This leads to the more PBL radiative

419   heating of air by aerosols in the control-novary run than in the control run and in the

420   control-1500-novary run than in the control-1500 run.

421   Comparisons between the aro-above-cld and aro-above-cld-novary runs and between

422   the aro-above-cld-1500 and aro-above-cld-1500-novary runs show that due to the

transportation of aerosols by downdrafts, aerosol concentrations in the aerosol layer in the free atmosphere reduces and those in the air below the layer increases (Figures 10b and 10d). Note that the free atmosphere, which includes the above-PBL atmosphere around or above cloud tops, 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 aerosol layer mainly occur between ~1.5 and ~2.5 km, and aerosol concentrations and the associated instability in the PBL do not change significantly (Figures 10b and 10d). This leads to similar instability in the PBL and CAPE, which in turn leads to similar updrafts and cloud mass between the aro-above-cld and aro-above-cld-novary runs and between the aro-above-cld-1500 and aro-above-cld-1500-novary runs (Figure 11a).

Due to more radiative heating of air in the PBL, there are higher CAPE, stronger updrafts and higher cloud mass in the control-novary run than in the control run and in the control-1500-novary run than in the control-1500 run (Figure 11a). It is notable that cloud mass in the control-novary run is so large that its maximum value in the vertical profile exceeds that even in the control-1500-novary run (Figure 11a). Associated with this, there are only ~20 % changes in cloud mass between the control-1500 and control-1500-novary runs, while there are as much as ~200 % changes in cloud mass between the control and control-novary runs. This indicates that with higher aerosol concentrations in the PBL, changes in cloud mass due to the wind-induced variation of those concentrations are much larger.

3.4 Comparisons between simulations with and without aerosol radiative effects

Figure 11b shows that with no aerosol radiative effects, differences in cloud mass due to the altitude of the aerosol layer are smaller. However, even with no aerosol radiative effects, there is higher cloud mass when the aerosol layer is in the PBL than in the free atmosphere as in the standard runs. Also, cloud mass increases when aerosol radiative effects are turned off and this increase enhances as aerosol concentrations increase (Figure 11b). Here, we see that aerosol radiative effects suppress clouds and reduce cloud mass by reducing the surface-reaching solar radiation and the surface heat fluxes. The suppression of clouds and

454 reduction in cloud mass are greater with higher aerosol concentrations, since more aerosols
455 reduce the surface-reaching solar radiation more.

456 Note that aerosol activation mainly occurs around cloud bases in the PBL and more
457 aerosols induce more activation for a given thermodynamic condition. Hence, there are
458 more aerosol activation (or nucleation of droplets) and higher cloud droplet number
459 concentration (CDNC) when the aerosol layer is in the PBL than in the free atmosphere.
460 The averaged CDNC over grid points with non-zero CDNC and the whole simulation
461 period is 532, 57, 131 and 53 cm^{-3} in the control-norad, aro-above-cld-norad, control-1500-
462 norad and the aro-above-cld-1500-norad runs, respectively. Droplets act as a source of
463 condensation, since individual droplets provide their surface areas onto which water vapor
464 condenses. Hence, higher CDNC induces more condensation and this in turn induces
465 stronger updrafts and more cloud mass with the aerosol layer in the PBL than in the free
466 atmosphere. These effects of more aerosols, which induce more condensation and stronger
467 updrafts, are generally referred to as aerosol microphysical effects (Lee et al., 2016). The
468 differences in CDNC due to the altitude of the aerosol layer increase with increasing
469 aerosol concentrations. This leads to greater differences in condensation, associated
470 updrafts and cloud mass due to the altitude of the aerosol layer with higher aerosol
471 concentrations when there are no aerosol radiative effects (Figure 11b).

472 Here, we see that differences in cloud mass due to the altitude of the aerosol layer are
473 greater when aerosol microphysical and radiative effects work together than when aerosol
474 microphysical effects work alone (Figure 11b). Also, remember that the initial
475 concentration of aerosols in the aro-above-cld-norad run is identical to that in the aro-
476 above-cld-1500-norad run in the PBL. Due to this, CDNC, condensation and cloud mass
477 in the aro-above-cld-norad run are similar to those in the aro-above-cld-1500-norad run
478 (Figure 11b).

479

480 **4. Summary and conclusions**

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482 This study examined how impacts of aerosols on warm cumulus clouds in the Korean
483 Peninsula vary with the altitude of an aerosol layer. It is found that the aerosol layer
484 intercepts the surface-reaching solar radiation more when the layer is in the PBL, which

485   corresponds to the atmosphere around or below cloud bases, than in the free atmosphere

486   which includes the above-PBL atmosphere around or above cloud tops. With the aerosol

487   layer in the PBL, this makes the surface heat fluxes and associated CAPE lower, which

488   tend to make updrafts weaker and cloud mass lower. However, the layer in the PBL heats

489   up the air there more to produce the higher CAPE and cloud mass.

490   With decreasing concentrations of aerosols in the aerosol layer, there are decreases in

491   the interception of the surface-reaching solar radiation, increases in surface heat fluxes,

492   CAPE and cloud mass. However, the decreasing concentrations of aerosols cause the jump

493   in CAPE to disappear when the layer is in the PBL. This makes differences in cloud mass

494   due to the altitude of the layer reduce. When the aerosol layer is in the PBL, with increasing

495   aerosol concentrations in the layer, the lifetime of cloud system reduces and becomes

496   shorter than when the layer is in the free atmosphere.

497   Updrafts and downdrafts in clouds transport aerosols. In particular, for the aerosol layer

498   in the PBL, updrafts transport aerosols in the layer to places above it. This reduces aerosol

499   concentrations in the layer, leading to reduction in radiative heating of air by aerosols,

500   CAPE, updrafts and cloud mass. This reduction enhances with increasing aerosol

501   concentrations in the layer. For the aerosol layer in the free atmosphere, downdrafts

502   transport aerosols in the layer to places below it. However, this does not affect aerosol

503   concentrations and radiative heating of air in the PBL significantly. This in turn has

504   negligible effects on CAPE and cloud mass.

505   Aerosol radiative effects suppress clouds and reduce cloud mass by cutting down the

506   surface-reaching solar radiation. This suppression of clouds increases with increasing

507   aerosol concentrations in the aerosol layer. Aerosol microphysical effects enhance cloud

508   mass and these effects are stronger with higher aerosol concentrations. Differences in cloud

509   mass due to the altitude of the aerosol layer are enhanced when aerosol radiative effects

510   and aerosol microphysical effects work together as compared to when only aerosol

511   microphysical effects are present.

512   This study shows that aerosol-induced changes in the surface fluxes and those in

513   radiative heating of air interact with each other in terms of responses of convection and

514   clouds to aerosols. This interaction varies with the altitude of aerosols and cloud-induced

515   wind. In general, traditional parameterizations for warm cumulus clouds in climate and

516   weather-forecast models have not been able to consider this dependence of the interaction

517   on the altitude of aerosols, since those parameterizations do not differentiate aerosol layers

518   based on their vertical locations. In addition, the cloud-induced wind at cloud scales has

519   not been represented by those parameterizations with good confidence. So, impacts of

520   aerosol transportation by cloud-induced wind on the interaction have not been properly

521   considered in those traditional parameterizations. This suggests that the vertical locations

522   of aerosols and cloud-induced wind should be added to factors that need to be considered

523   or improved to better parameterize warm cumulus clouds and their interactions with

524   aerosols.

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547 **Code/Data source and availability**

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

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

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

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

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

554
555 **Author contributions**

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

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

558 and contribute to their improvement.

559
560 **Competing interests**

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

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744 **FIGURE CAPTIONS**
745
746 Figure 1. (a) An inner rectangle in the map of the Korean Peninsula represents the
747 simulation domain. The green represents the land area and the light blue the ocean area in
748 the map. A black dot marks the location of a site where the radiosonde sounding is obtained
749 and a red dot the location of the $PM_{2.5}$ station in the Yellow sea. (b) The simulation domain
750 is shown. The black dots mark the locations of the $PM_{2.5}$ stations and the red dot the location
751 of the AERONET site in the domain.

752
753 Figure 2. Spatial distribution of cloud reflectivity which is unitless and observed by the
754 COMS at 14:00 LST April 13th, 2016 in the simulation domain. Contours are at 0.11, 0.15,
755 0.19 and 0.25.

756
757 Figure 3. Vertical distributions of potential temperature and water-vapor mixing ratio at
758 09:00 LST on April 13th, 2016. These distributions are obtained from radiosonde sounding
759 near the simulation domain in Figure 1a.

760
761 Figure 4. Time series of $PM_{2.5}$ observed at the station in the Yellow sea (blue line) and of
762 the average $PM_{2.5}$ over stations in the simulation domain (red line) between 03:00 LST and
763 18:00 LST on April 13th in 2016.

764
765 Figure 5. Aerosol size distribution at the surface. N represents aerosol number
766 concentration per unit volume of air and D represents aerosol diameter.

767
768 Figure 6. Same as Figure 2 but in the control run.

769
770 Figure 7. Vertical distributions of the time- and area-averaged cloud-liquid mass density
771 that represents cloud mass for the standard simulations (i.e., the control, aro-above-cld,
772 control-1500 and aro-above-cld-1500 runs).

773
774 Figure 8. Time series of the domain-averaged (a) liquid-water path, (b) updraft speed, (c)
775 condensation rate and (d) CAPE in the standard simulations.

776   Figure 9. Vertical distributions of the time- and area-averaged radiative heating rate (a) in

777   the control and aro-above-cld runs over the initial period between 10:00 and 13:50 LST,

778   (b) in the control and aro-above-cld runs and (c) in the control-1500 and aro-above-cld-

779   1500 runs over the whole simulation period.

780

781   Figure 10. Vertical distributions of the time- and area-averaged aerosol concentrations (a)

782   in the control and control-novary runs, (b) aro-above-cld and aro-above-cld-novary runs,

783   (c) control-1500 and control-novary-1500 runs and (d) aro-above-cld-1500 and aro-above-

784   cld-novary-1500 runs.

785

786   Figure 11. Vertical distributions of the time- and area-averaged cloud-liquid mass density.

787   In (a), the control-novary, aro-above-cld-novary, control-1500-novary and aro-above-cld-

788   1500-novary runs and in (b), the control-norad, aro-above-cld-norad, control-1500-norad

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

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Simulations	Altitudes of a aerosol layer (km)	Aerosol concentrations in the 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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807   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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821 Table 2. The time- and area-averaged net solar radiation, latent heat, sensible heat and total
822 heat (sensible plus latent heat) fluxes at the surface over the whole simulation period in the
823 standard simulations. Numbers in the parentheses are averaged over the initial period
824 between 10:00 and 13:50 LST for the control and aro-above-cld runs.

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	Control run	Observations	Observation sources
Cloud fraction (CF)	0.25	0.21	Ground stations
Cloud-top height (CTH) (km)	2.8	2.6	COMS
Cloud-bottom height (CBH) (km)	1.1	1.0	Ground stations
Cloud optical depth (COD)	3.5	3.2	The Moderate Resolution Imaging Spectroradiometer (MODIS)
Droplet effective radius (re) (μm)	7.5	8.0	MODIS
Liquid-water path (LWP) (g m^{-2})	17.3	16.8	MODIS
The surface wind speed (WS) (m s^{-1})	1.8	1.6	Ground stations
The surface wind direction (WD) (Degree; measured clockwise from geographical north)	220	230	Ground stations
The surface air temperature (ST) (Degree Celsius)	16.9	16.7	Ground stations

842

843 Table 3. The simulated and observed values of cloud and environmental variables, and the

844 observation sources that have been used to obtain the observed values. At each observation

845 time (simulation time step), CF is averaged (obtained) over ground stations (grid points) in

846 the domain as shown in Figure 1b and the averaged (obtained) CF is averaged over the

847 simulation period with clouds to calculate the presented and observed (simulated) CF

848 values. To obtain the presented values of CTH, CBH, COD, re and LWP, the observed

849 values at observation spatial points (the simulated values in grid columns for CTH, CBH

850 and LWP and at grid points for COD and re) in the domain are averaged over areas with

851 non-zero values at each observation time (simulation time step) and then over the

852 simulation period with non-zero values. To obtain the presented values of WS, WD and

853 ST, the simulated values at grid points, which correspond to the atmosphere immediately

854 above the surface, and each simulation time step, and the observed values at ground stations

855 and each observation time are averaged over the domain and then over the whole

856 simulation period.

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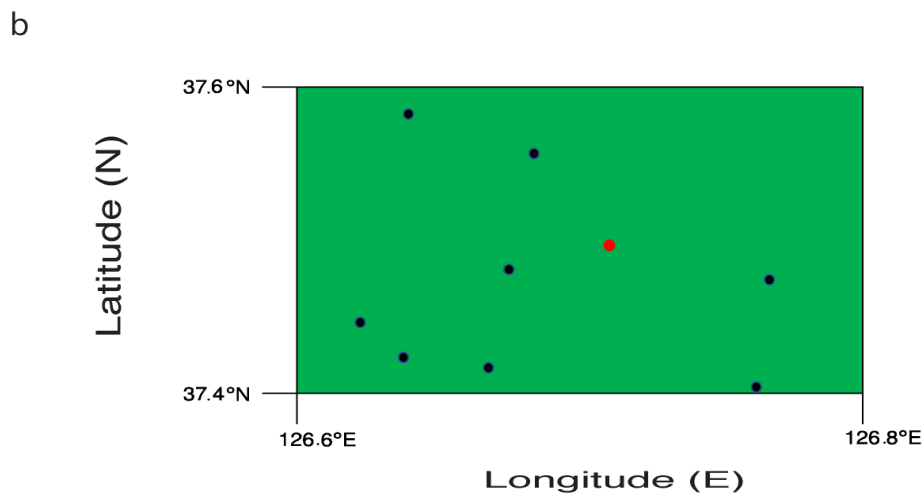
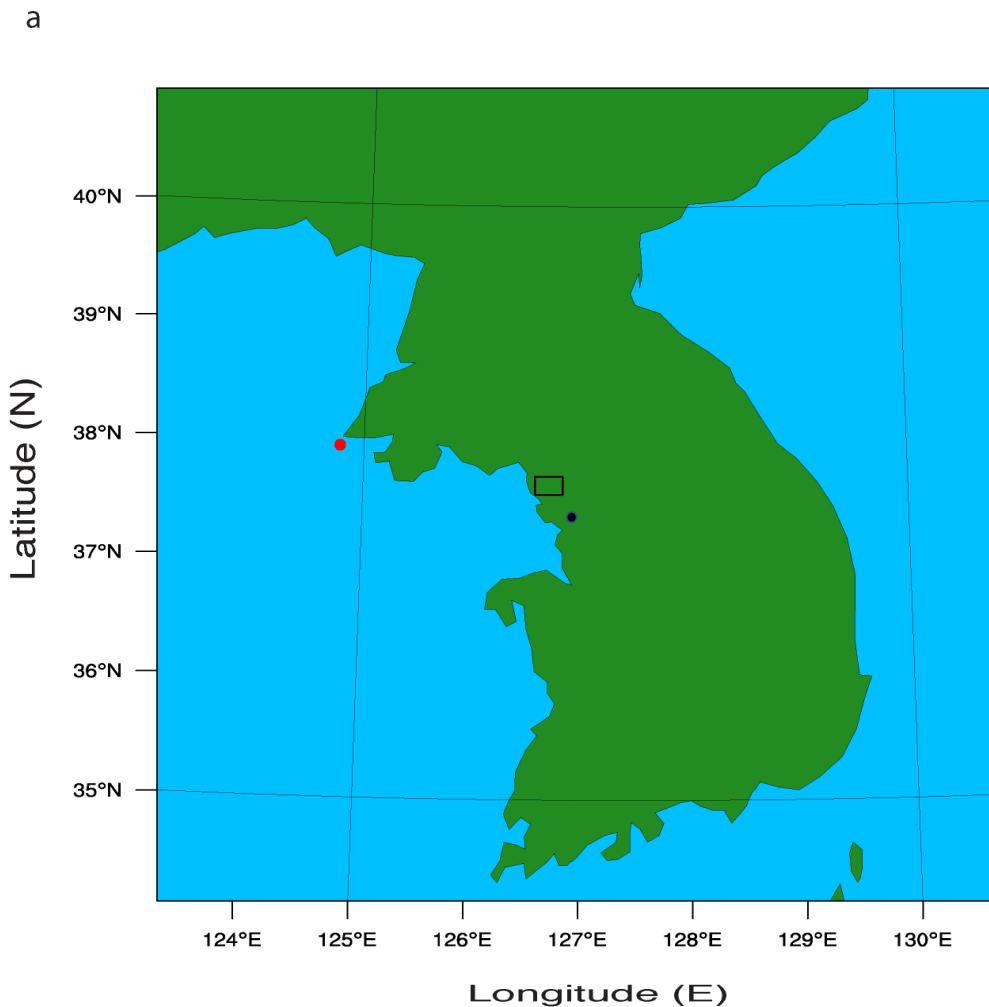
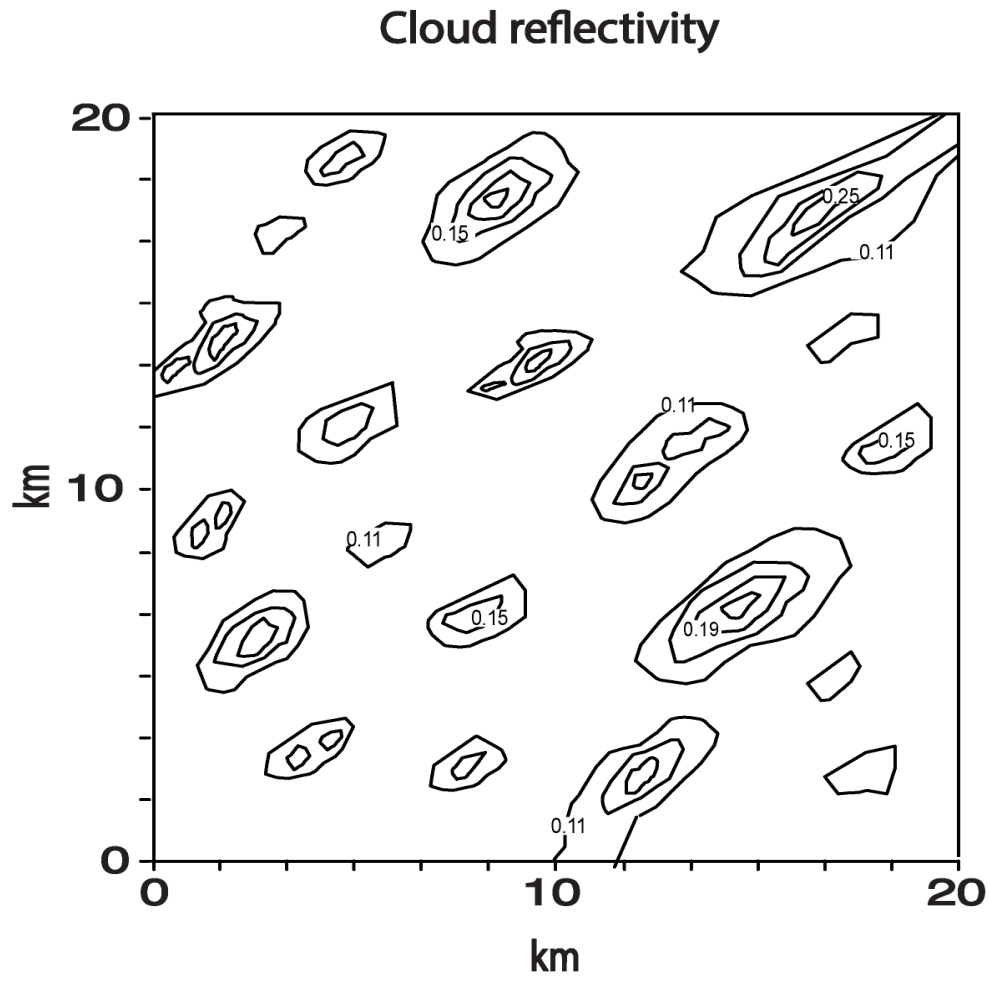
a

b

 Figure 1

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863   **Figure 2**

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Figure 3

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Figure 4

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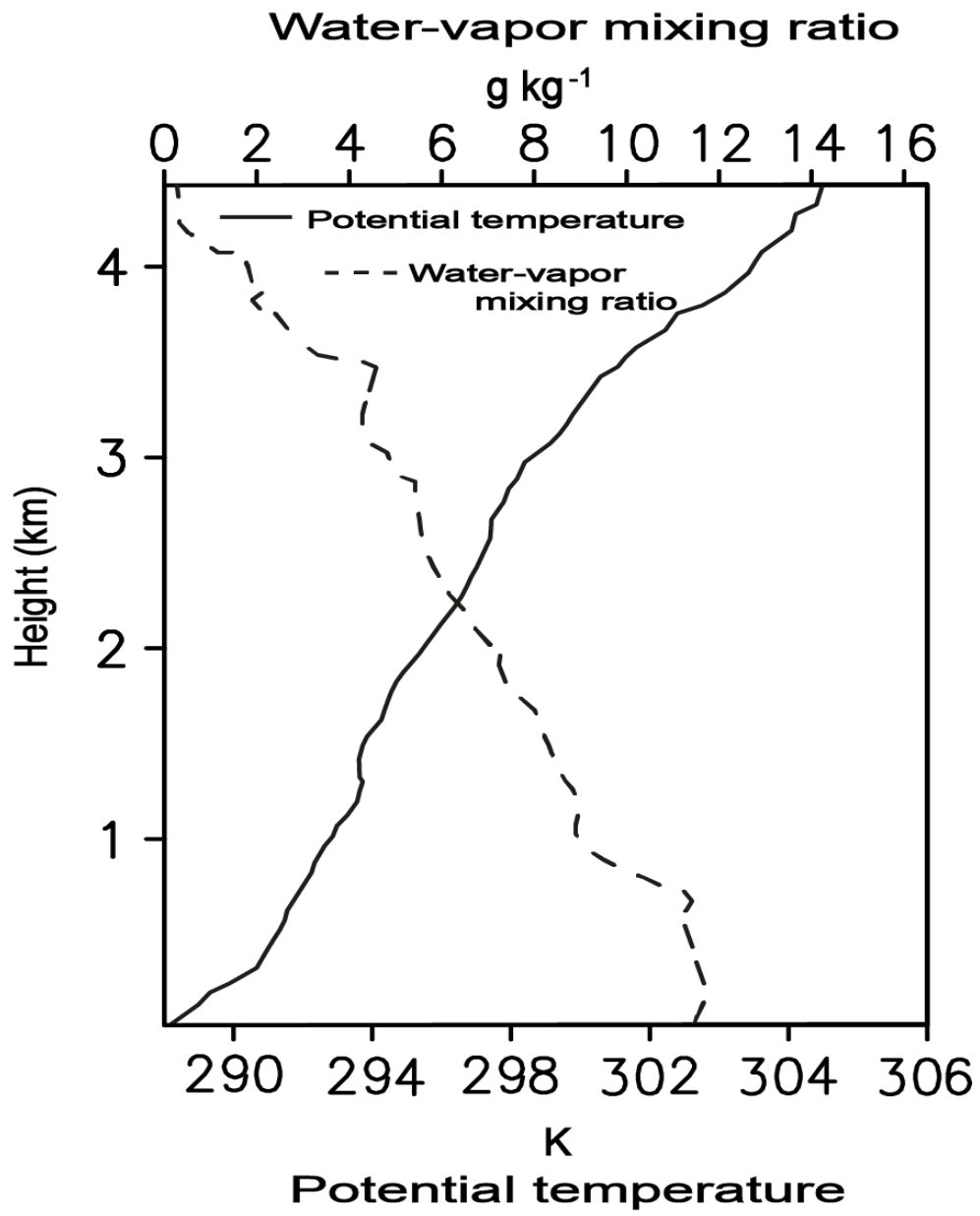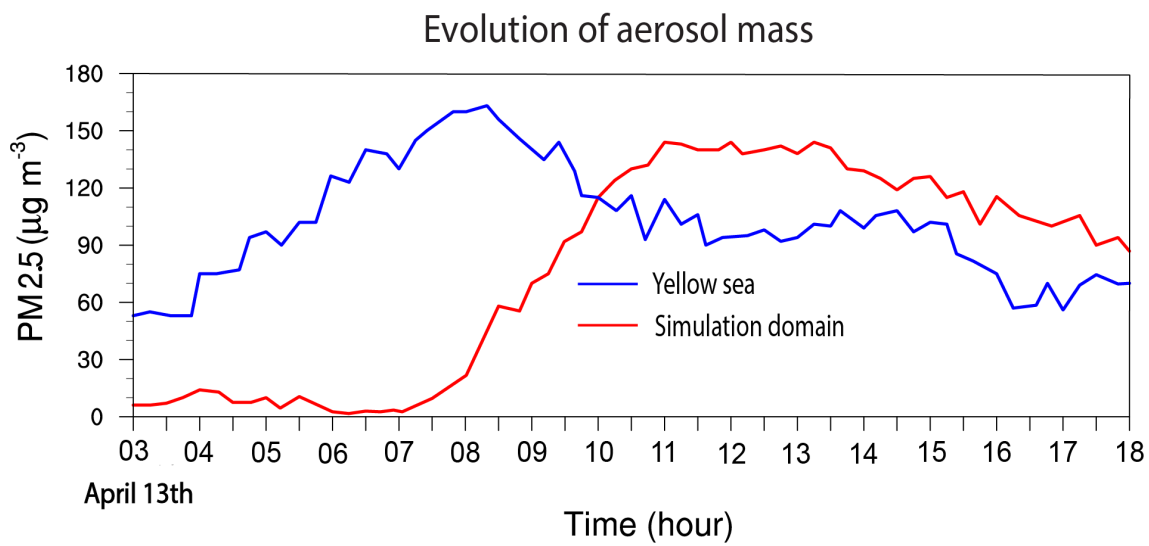
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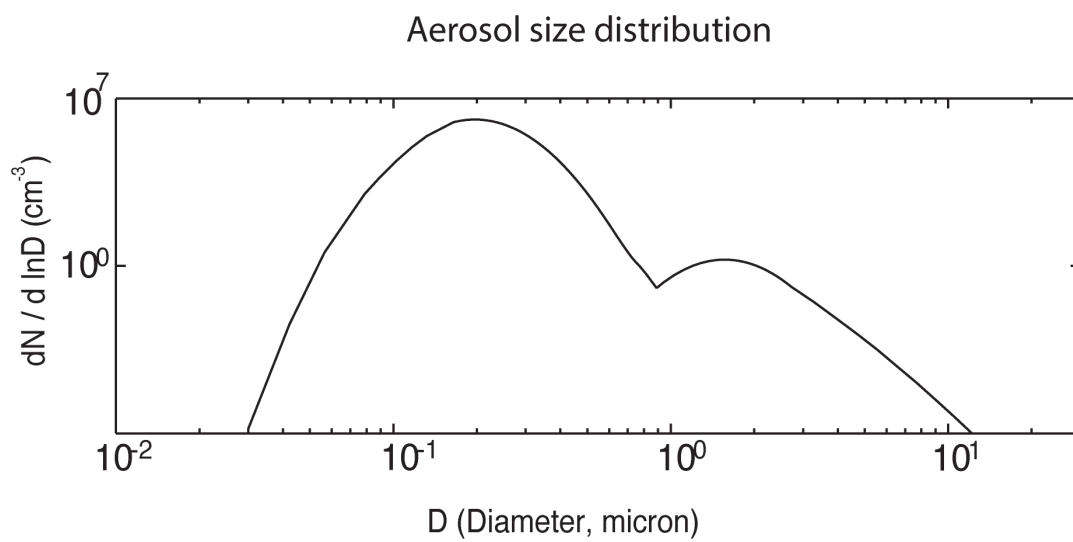
Figure 5

Cloud reflectivity

896
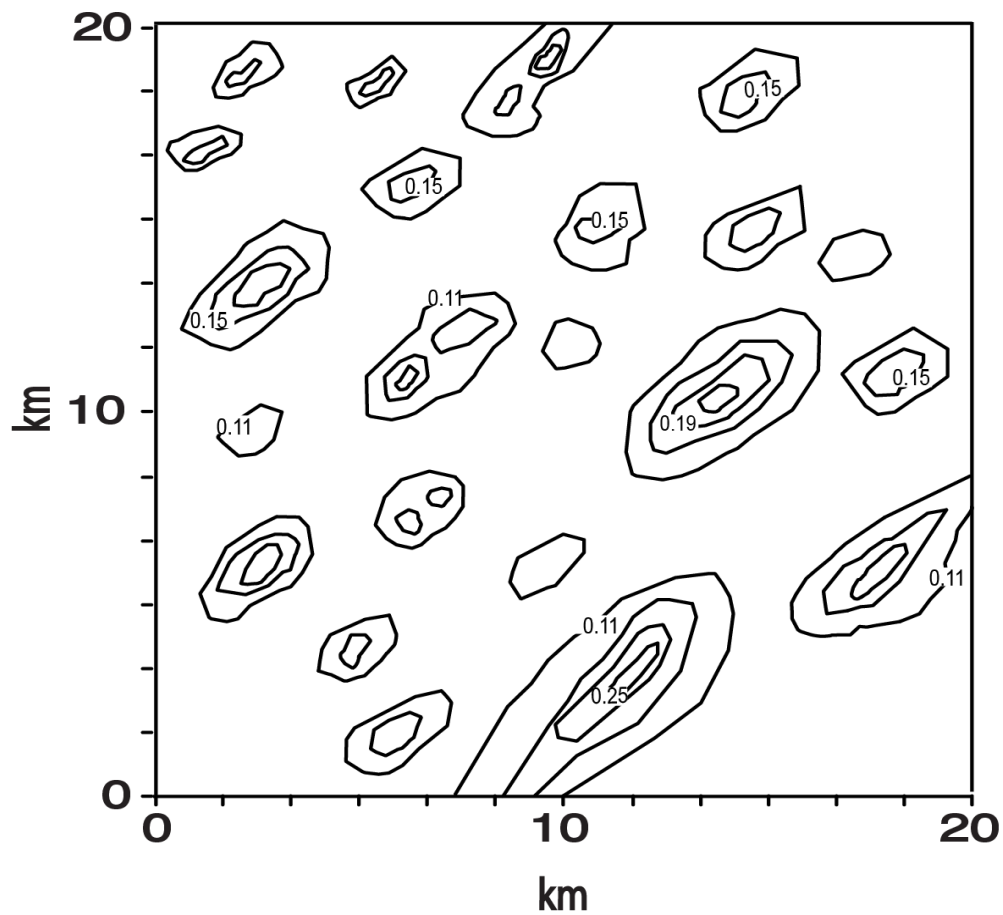
Figure 6

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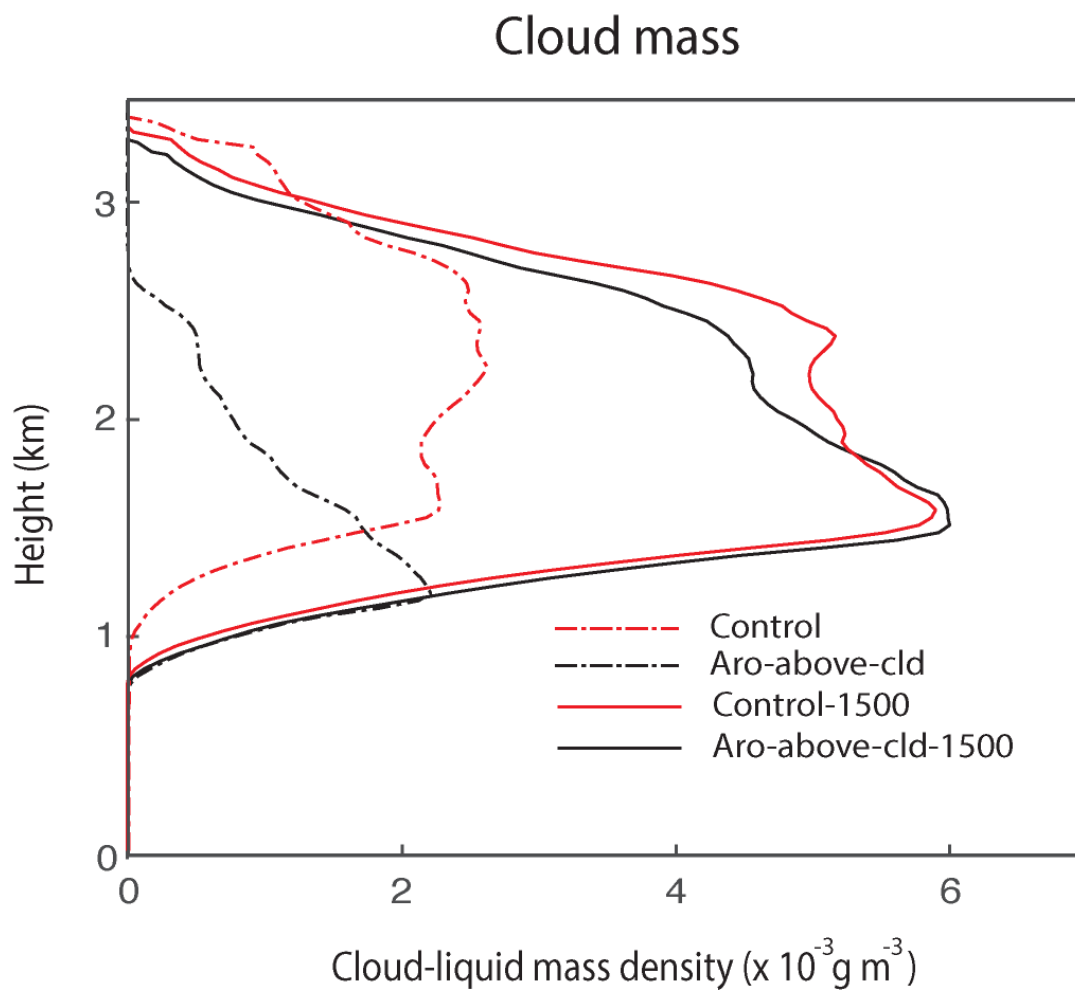
Figure 7

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911   **Figure 8**

Radiative heating rate
Initial period

Whole period
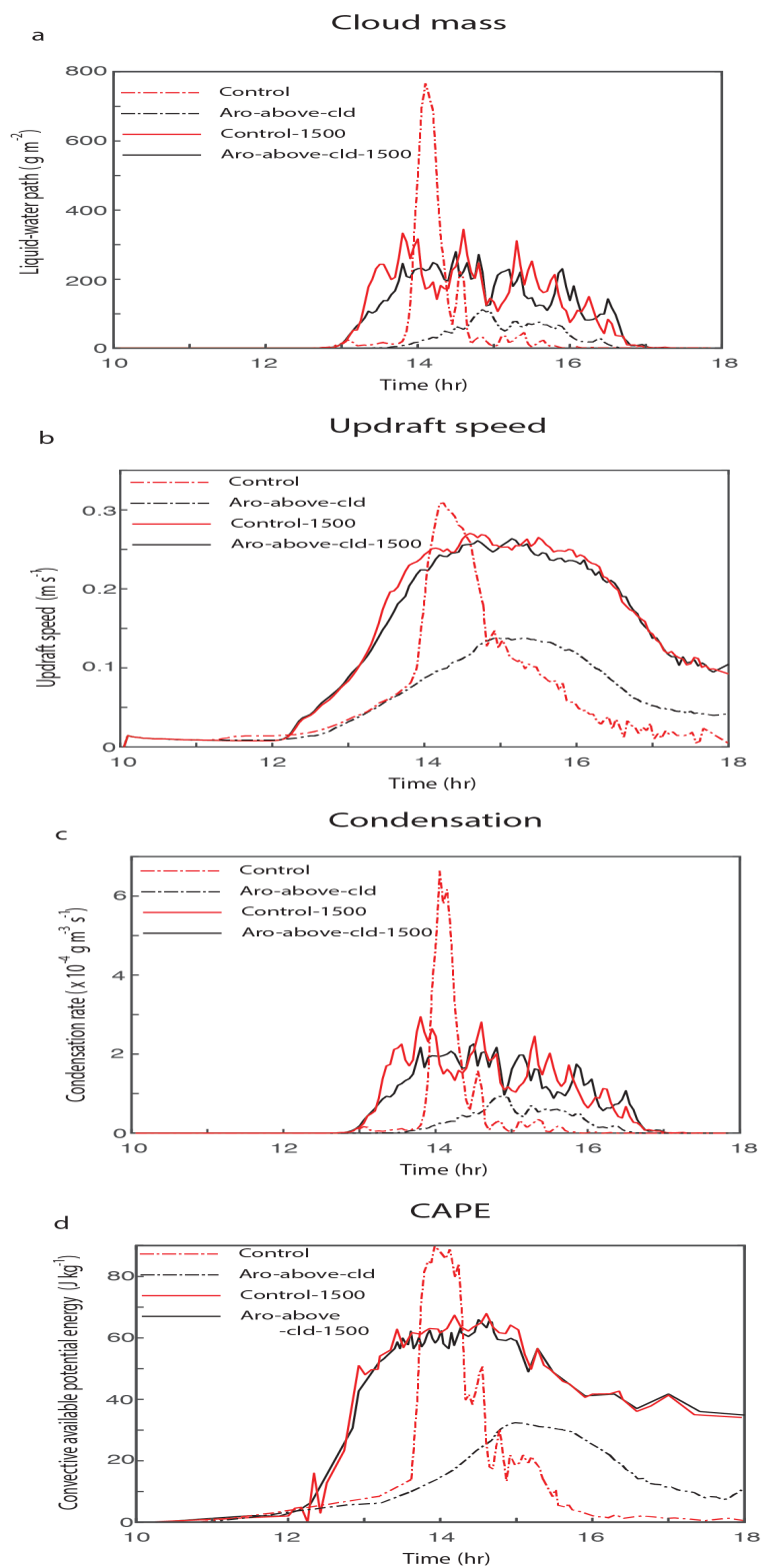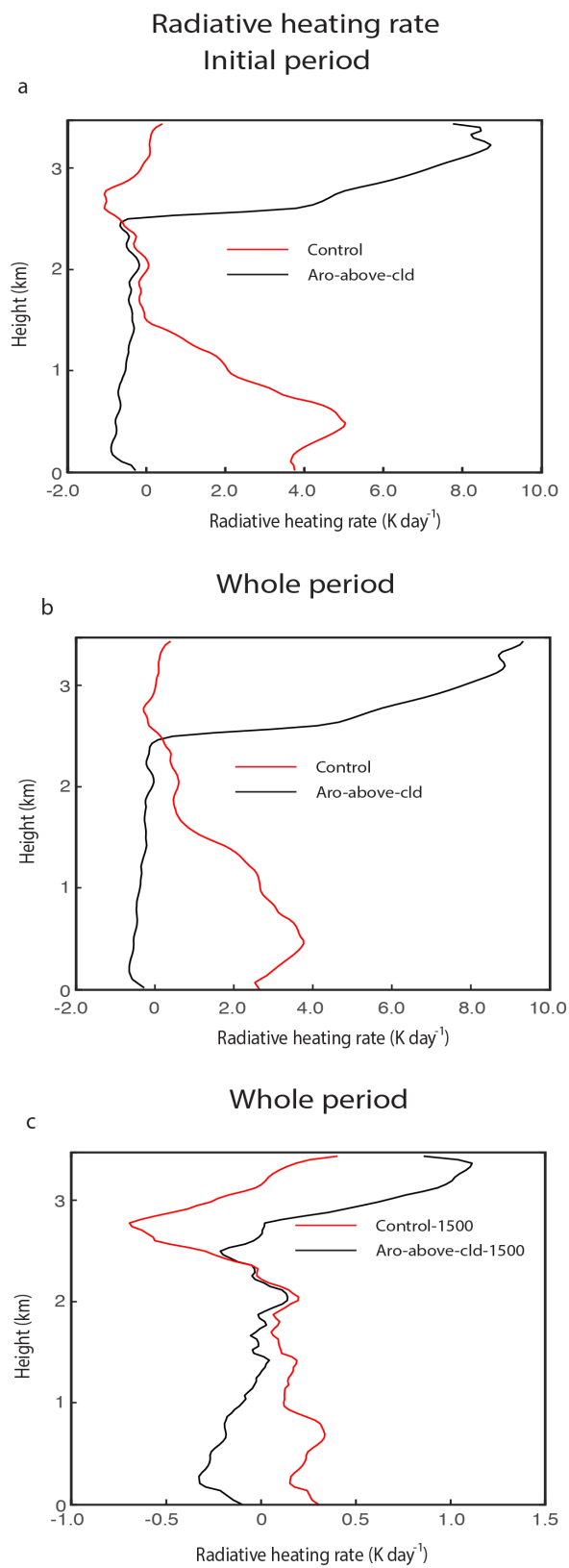
Figure 9

Vertical distributions of aerosol concentrations

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Figure 10

Cloud mass

a

b

918
919
920

Figure 11