Aerosol impacts on radiative and microphysical properties of clouds and precipitation formation

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Abstract

Through modifying the number concentration and size of cloud droplets, aerosols have intricate impacts on radiative and microphysical properties of clouds, which together influence precipitation processes. Aerosol-cloud interactions for a mid-latitude convective cloud system are investigated using a two-moment aerosol-aware bulk microphysical scheme implemented into the Weather Research and Forecasting (WRF) model. Three sensitivity experiments with initial identical dynamic and thermodynamic conditions, but different cloud-nucleating aerosol concentrations were conducted. Increased aerosol number concentration has resulted in more numerous cloud droplets of overall smaller size, through which the optical properties of clouds have been changed. While the shortwave cloud forcing is significantly increased in more polluted experiments, changes in the aerosol number concentration have negligible impacts on the longwave cloud forcing. For the first time, it is found that polluted clouds have higher cloud base heights, the feature that is caused by more surface cooling due to a higher shortwave cloud forcing, as well as a drier boundary layer in the polluted experiment compared to

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the clean. The polluted experiment was also associated with a higher liquid water content (LWC) caused by an increase in the number of condensation of water vapor due to higher concentration of hygroscopic aerosols acting as condensation nuclei. The domain-averaged accumulated precipitation is little changed under both polluted and clean atmosphere. Nevertheless, changes in the rate of precipitation are identified, such that under polluted atmosphere light rain is reduced, while both moderate and heavy rain are intensified, confirming the fact that if an ample influx of water vapor exists, an increment of hygroscopic aerosols can increase the amount of precipitation.

Keywords: Aerosol-cloud interactions, Shortwave cloud forcing, Liquid water content, Cloud base height

1. Introduction

Atmospheric aerosols have intricate impacts on cloud microphysics by modifying the number concentration and size of cloud droplets (Andreae et al., 2004; Khain et al., 2004), which change the albedo of clouds (Twomey, 1977; Menon et al., 2002; Quaas et al., 2004; Fan et al., 2013), the coalescence efficiency of droplets (Albrecht, 1989; Rosenfeld and Lensky, 1998; Rosenfeld et al., 2001), the release of latent heat and the strength of cloud convection (Andreae et al., 2004; Khain et al., 2004; Koren et al., 2005), the cloud top height (Andreae et al., 2004; Koren et al., 2005), the glaciation height (Rosenfeld and Lensky, 1998), the strength of the cold pool via changes in evaporation (Lebo, 2014; Lebo and Morrison, 2014; Grant and van den Heever, 2015) and consequently precipitation formation (Albrecht, 1989; Rosenfeld, 1999; Rosenfeld and Woodley, 2000; Khain et al., 2004; Zhang et al., 2005;

14 Seifert and Beheng, 2006; Thompson and Eidhammer, 2014; Wang, 2015).
15 All these changes are referred to as (or the result of) the aerosol indirect
16 effects, which are considered one of the most uncertain forcing in climate
17 (Zhang et al., 2015). The effects of aerosols on clouds include the "first in-
18 direct" or "Twomey" effect (Twomey, 1977) on cloud droplet size and thus
19 reflectivity for a fixed liquid water content (LWC), and the "second indirect"
20 effect (Ackerman et al., 2000) on lifetime and extent of clouds, as well as on
21 precipitation processes (Zhang et al., 2005; Seifert and Beheng, 2006)

22 Twomey (1977) first stated that for a constant liquid water path, en-
23 hancement of aerosol concentration can increase the droplet concentration
24 and decrease the mean size of cloud droplets, which increases the total droplet
25 cross-sectional area, and consequently the cloud albedo, corresponding to the
26 cooling of the climate system. Evidence of enhanced cloud reflectivity can
27 be seen in clouds polluted by ship exhaust (Coakley et al., 1987), and in
28 polluted continental compared to clean maritime clouds (Stefan and Iorga,
29 2004). However, it should be noted that the increase in cloud albedo in re-
30 sponse to a decrease in droplet sizes is the greatest in shallow clouds because
31 regardless of their droplet sizes, deep and water-rich clouds reflect most of
32 the solar radiation (Platnick and Twomey, 1994).

33 The reduction of droplet sizes in polluted clouds inhibits collision and co-
34 alescence processes (Albrecht, 1989; Rosenfeld and Lensky, 1998; Rosenfeld
35 et al., 2001); thereby slows the conversion of cloud droplets into rainfall, to
36 the extent of completely suppressing warm-rain processes (Rosenfeld, 1999).
37 The reduced warm-rain processes due to enhanced aerosols are evident in
38 more precipitable shallow maritime clouds compared to their counterpart

39 continental clouds (Ludlam, 1980; Mülmenstädt et al., 2015). This is also
40 evident in the inverse relation identified between the droplet concentration
41 and frequency of drizzle in stratocumulus clouds, both by observations (Al-
42 brecht et al., 1988; Rosenfeld et al., 2001) and simulations (Saleeby et al.,
43 2015). The decreased rainfall in shallow clouds leads to an increase in the
44 lifetime and coverage of clouds (Albrecht, 1989), to the extent that the re-
45 sultant increase in the global albedo is believed to be much greater than the
46 cloud albedo or "Twomey" effect (Sekiguchi et al., 2003; Kaufman et al.,
47 2005).

48 Suppressing warm-rain processes delays the onset of precipitation (An-
49 dreae et al., 2004), such that in mixed-phase clouds enables the unprecipi-
50 tated cloud droplets to reach greater heights, which results in more freezing
51 of cloud droplets and the release of latent heat of freezing, higher cloud top
52 heights, enhancement of cold-rain processes (Rosenfeld and Woodley, 2000; Li
53 et al., 2011) and occasionally more intensive thunderstorms (Andreae et al.,
54 2004). The greater release of latent heat prolongs the growth of convective
55 clouds; thereby further invigorates the cloud development and formation of
56 thunderstorms and large hails (Andreae et al., 2004; Rosenfeld, 2006) that
57 eventually might affect the water cycle. In addition, an increase in the num-
58 ber of aerosol concentrations that can act as ice nuclei is associated with
59 more frequent glaciation of supercooled clouds (referred to as the "glaciation
60 indirect effect"), and an increase of precipitation via the ice phase (Lohmann
61 and Lesins, 2002).

62 The aerosol indirect effects have been also extensively discussed in several
63 recent observational-based studies. Using in situ measurements, Andreae

et al. (2004) indicated a shift in the onset of precipitation to larger heights in smoky polluted clouds, which allows invigoration of updrafts and higher cloud tops, formation of large ice crystals, intense thunderstorms and large hails. They noted that microphysical effects of smoke aerosols on the development of clouds overcome on their radiative cooling effects which tend to suppress the boundary-layer clouds. Koren et al. (2005) found that an increase in the aerosol number concentration is associated with a delay in freezing (known as "thermodynamic effect") and an enhancement of cloud convection, which consequently leads to an increase in cloud coverage and the cloud top height. The increase of cloud convection in response to the increase of the aerosol number concentration was also found in a numerical study by Fan et al. (2007), although they noted that when the air is already heavily polluted, further increase of aerosols has less impact on convection processes. Using the Tropical Rainfall Measuring Mission (TRMM) precipitation measurements, Bell et al. (2008) found a significant increase in the summertime afternoon thunderstorms over the Southeastern United States, which was attributed to the mid-week increase in aerosol concentration.

Although the above discussed aerosol effects on cloud microphysics are well accepted, recent studies indicate that to fully understand such effects, environmental conditions such as humidity or convective available potential energy (CAPE) (Khain et al., 2008; Storer et al., 2010), wind shear (Fan et al., 2009; Lee et al., 2012; Lebo and Morrison, 2014) and the altitude of the dry layer (Grant and van den Heever, 2015) should be considered too. For example, it was found that an increase in aerosol concentration is associated with an increase in precipitation under tropical maritime conditions,

89 but a decrease in precipitation for clouds developing under dry continental
90 conditions (Khain et al., 2008). The decrease of precipitation in dry con-
91 ditions was attributed to the sublimation of ice and evaporation of smaller
92 drops when they are falling into a deep layer of dry air. Analysis of Fan
93 et al. (2007) did not indicate such suppression effect, but they noted that the
94 aerosol indirect effects are negligible in relatively dry air. Lebo and Morrison
95 (2014) investigated the dynamical effects of increasing the aerosol loading on
96 the strength and structure of squall lines. Their results indicated that under
97 a weak wind shear, an increase in aerosol loading leads to the weakening of
98 the cold pool, an intensification of the convective mass flux and an enhance-
99 ment of precipitation, while the overall reduction of precipitation was found
100 under a strong wind shear condition.

101 The explicit nucleation of cloud droplets and ice crystals by aerosols
102 has been recently applied in the Weather Research and Forecasting (WRF)
103 model. Li et al. (2008) first implemented a two-moment microphysics scheme
104 with prognostic aerosol number and mass concentrations into the WRF
105 model, and carried out simulations under different aerosol conditions. They
106 demonstrated that precipitation increases from clean marine to continen-
107 tal background conditions, but it is considerably reduced and completely
108 suppressed under highly polluted cases. Suppression of precipitation at ex-
109 tremely high aerosol concentrations was attributed to the fact that ice pro-
110 duction is minimal in such conditions; thus hydrometeors cannot grow to
111 sufficiently large sizes to survive evaporation. Recently, Thompson and Eid-
112 hammer (2014) implemented a two-moment aerosol-aware bulk microphysi-
113 cal scheme into the WRF model to discuss the impacts of aerosols on clouds

114 development and precipitation in a large winter cyclone. They found that
115 aerosol effects on precipitation are much greater over the regions with light
116 precipitation rates. They also noted a shift in the location of precipitation
117 due to aerosol effects. More recently, Wang (2015) applied the cloud resolv-
118 ing WRF model with a modified Morrison two-moment bulk microphysical
119 scheme. He identified an intensified convection and more efficient mixed-
120 phase processes under a polluted aerosol condition.

121 In spite of the existing knowledge, it is not yet clear how spatial distribu-
122 tion and the rate of precipitation changes in response to the combined effects
123 of aerosols on radiative and microphysical properties of clouds. Here it has
124 been shown that the aerosol effect on precipitation processes is a function of
125 precipitation intensity, such that light rain has been reduced or completely
126 suppressed under the polluted atmosphere, while moderate and heavy rainfall
127 have been intensified.

128 The main objective of the present study is to investigate if and to the
129 extent that the following features are related to the aerosol indirect effects:
130 (1) the optical properties of clouds; (2) the cloud base height; (3) total accu-
131 mulated precipitation; and (4) the rate of precipitation. To this end, a two-
132 moment aerosol-aware bulk microphysical scheme, developed by Thompson
133 and Eidhammer (2014), is applied.

134 **2. Model description**

135 *2.1. The WRF model*

136 The WRF mesoscale model version 3.7 (Janjic et al., 2003) was applied to
137 investigate responses of cloud properties to changes in aerosol concentration.

138 2.2. Two-moment aerosol-aware bulk microphysical scheme

139 A two-moment aerosol-aware bulk microphysical scheme, developed and
140 discussed by Thompson and Eidhammer (2014), is used in the present study.
141 This is an update of Thompson et al. (2008) bulk microphysical scheme
142 which explicitly incorporates nucleation of cloud droplets and ice crystals.
143 In the two-moment bulk microphysical schemes, both the mass and total
144 number concentration of cloud droplets are predicted, while one-moment
145 microphysical schemes only consider the mass concentration of hydrometeors.
146 The Thompson aerosol-aware scheme calculates the mass mixing ratio of
147 water vapor, cloud droplets, rain drops, ice crystals, snow, and graupel, as
148 well as the number concentration of cloud droplets, ice crystals and rain
149 drops. The Köhler activation theory (Köhler, 1936) is used for nucleation
150 of aerosols to form cloud droplets, and a generalized gamma distribution is
151 assumed for cloud droplets.

152 Ice nucleation is based on the number concentration of mineral dust
153 aerosols as dust is the most abundant natural ice nuclei in the atmosphere
154 (Hoose et al., 2010; Murray et al., 2012), while other ice nuclei are not in-
155 corporated in the current version of the WRF model. Nucleation of dust
156 particles into ice crystals is based on the parameterization of DeMott et al.
157 (2010) for condensation and immersion freezing at relative humidity above
158 water saturation and the parameterization of Phillips et al. (2008) for de-
159 position nucleation at relative humidity less than water saturation. More
160 descriptions of the two-moment aerosol-aware bulk microphysical scheme,
161 and the approach adopted for nucleation of cloud droplets and ice crystals
162 can be found in Thompson and Eidhammer (2014).

163 2.3. *Input aerosol dataset*

164 The initial aerosol number concentrations were obtained from 7-yr (2001-
165 2007) global simulations (Colarco et al., 2010) of the Goddard Chemistry
166 Aerosol Radiation and Transport (GOCART) model (Ginoux et al., 2001)
167 with 0.5° longitude by 1.25° latitude resolution. The aerosol input data
168 include mass mixing ratios of sulfate, dust, black carbon (BC), organic
169 carbon (OC), and sea salt, although black carbon is ignored in the cloud
170 condensation nuclei (CCN) activation. As noted by Thompson and Eid-
171 hammer (2014), dust aerosols larger than $0.5 \mu\text{m}$ are accumulated into the
172 ice-nucleating mode, while all other aerosol species are combined into the
173 cloud-droplet-nucleating mode. By assuming lognormal distributions and
174 applying the approach of Chin et al. (2002) for characteristic diameters and
175 geometric standard deviations, the input mass mixing ratios were converted
176 to number concentrations. For simplicity, a variable lower boundary con-
177 dition was implemented to represent surface aerosol emissions based on the
178 initial near-surface aerosol concentration and mean surface wind. The fol-
179 lowing formula then has been applied in lowest level of the model to calculate
180 the flux of aerosols: $dN_{wfa}/dt = 10^{[\log(N_{wfa}) - 3.69897]}$, where N_{wfa} denotes hy-
181 groscopic aerosol number concentration. More descriptions are presented in
182 Thompson and Eidhammer (2014).

183 2.4. *Numerical experiments*

184 Three sensitivity experiments with different initial and boundary hygro-
185 scopic aerosol number concentrations were conducted for an observed case of
186 convective cloud development and rainfall over Tehran on April 14, 2012. The
187 aim is to investigate the response of cloud properties to changes in aerosol

188 number concentrations, while the number of non-hygroscopic ice-nucleating
 189 aerosol concentrations (that has been considered to be dust) was remained
 190 identical for the three experiments. Hygroscopicity parameter was assumed
 191 to be 0.4, while aerosol mean radius considered to be $0.04\ \mu\text{m}$, although they
 192 can be both altered to represent different aerosol populations. The aerosol
 193 number concentration in the first simulation represents conditions of the cur-
 194 rent era, and this simulation is referred to as the control experiment. In the
 195 second simulation, hygroscopic aerosol number concentration was reduced to
 196 one-fifth of the control experiment, and it is referred to as the clean exper-
 197 iment. Finally, to investigate the impacts of high aerosol concentration on
 198 cloud microphysics and precipitation, the aerosol number concentration at
 199 all model grid points was increased by a factor of 5, and this simulation is
 200 referred to as the polluted experiment. As shown in Table 1, the innermost
 201 domain-averaged hygroscopic aerosol number concentrations averaged over
 202 the simulation period and in all vertical levels were 109.8, 509.1 and 2656.6
 203 cm^{-3} in the clean, control and polluted experiments, respectively. Spatial
 204 distribution of initial hygroscopic aerosol number concentration at the sur-
 205 face in the control simulation is also shown in Figure 1, while vertical profiles
 206 of initial domain-averaged hygroscopic and ice-friendly aerosol number con-
 207 centrations for the three experiments are depicted in Figure 2. Apart from
 208 differences in the initial and boundary hygroscopic aerosol number concentra-
 209 tions, the model setups and meteorological initial and boundary conditions
 210 for the three experiments are identical.

211 Simulations were configured with a mother domain and two two-way in-
 212 teractive nested domains (nesting ratios: 1:3:3), using 120×120 , $121 \times$

213 100 and 151×121 horizontal grid points, respectively. The WRF domain
214 configuration is shown in Figure 1, while topographic features of Iran are
215 overlapped. In the vertical, 45 σ -levels up to 50 hPa were used, spaced from
216 68 m near the surface to 500 m near the midtroposphere. The mother do-
217 main was configured with the horizontal resolution of 21000 m, while the
218 nested domains were configured to have horizontal resolutions of 7000 and
219 2333 m, respectively. Simulations were conducted for 30 hours, starting at
220 1800 UTC 13 April 2012. To minimize the impact from the initial condi-
221 tions, only results from 0000 UTC 14 to 0000 UTC 15 April (referred to
222 as the simulation period hereafter) are analyzed. The meteorological initial
223 and lateral boundary conditions were derived from the National Center for
224 Environmental Prediction final analysis (NCEP/FNL) data at 1° horizontal
225 resolution and 6 h temporal intervals.

226 The geographical data for the land-use and topography were obtained
227 from the standard USGS (United States Geological Survey) dataset, with the
228 resolution of 30-arc second (0.9 km) for the innermost domain. The model
229 physics that have been applied in the experiments are as follows. Convection
230 processes for the mother and first nested domains were parameterized using
231 Kain-Fritsch scheme (Kain, 2004), while modeled explicitly in the inner-
232 most domain. Dudhia (Dudhia, 1989) and Rapid Radiative Transfer Model
233 (RRTM; Mlawer et al., 1997) were used for shortwave and longwave radia-
234 tion, respectively. Five layer thermal diffusion land-surface scheme and the
235 revised MM5 similarity theory surface layer scheme (Jiménez et al., 2012)
236 were used. This revised version removes previously identified limits, and
237 uses updated stability functions. Boundary-layer processes were parameter-

238 ized using the non-local Yonsei University (YSU) scheme, with an explicit
239 entrainment layer and a parabolic K profile in an unstable mixed layer (Hong
240 et al., 2006).

241 **3. Model evaluation**

242 Accumulated rainfall from the state-of-the-art WRF model for the control
243 experiment is compared against the TRMM product (3B42 V7) with the 0.25°
244 $\times 0.25^\circ$ resolution (Figure 3a-b). The WRF model correctly represented
245 the rainfall occurrence in the northern half of Iran. However, the model
246 overestimated the accumulated rainfall over the complex topographic regions.

247 Vertical profiles of air temperature and wind speed simulated at 0000
248 UTC 14 April 2012 are also compared against the corresponding atmospheric
249 radiosondes at a synoptic station located at 35.7° N, 51.3° E and 1190.8 m
250 above mean sea level (Figure 3c-d). Apart from layers very close to the
251 surface, the vertical profile of air temperature simulated by the model agrees
252 very well with the radiosonde profile. However, a temperature inversion
253 near the surface that is simulated at 0000 UTC was not observed. The
254 vertical profile of the observed wind speed is also well simulated by the model,
255 including the observed maximum wind speed located between 600 and 500
256 hPa.

257 **4. Synoptic overview**

258 The pattern of the convective system triggered by a mid-level trough
259 forcing is shown in Figure 4. It is clear that the arrival of a cooler air from
260 the north and the mid-level trough from the southwest of Tehran invigorates

261 pre-existing convective processes, which are typical in the early spring season.
 262 At 0000 UTC 14 April 2012, the trough appeared at both 500 and 300 hPa
 263 in southwest of Iran, while a ridge was dominated in the east parts. At this
 264 time, a closed low was formed at 500 hPa in north of the Persian Gulf, and a
 265 strong jet stream at 300 hPa was extended from eastern Turkey through Iraq
 266 to the southern Persian Gulf. This mid-level trough was associated with a
 267 tongue of surface low pressure on the border of Iran and Iraq, with the value
 268 of 1008 hPa. Twelve hours later, the low-pressure at 500 hPa was weakened
 269 and moved a bit to the east, while the jet at 300 hPa was intensified. At
 270 the surface, a closed low pressure was formed, with the center of 1008 hPa.
 271 At 0000 UTC 15 April 2012, the 500-hPa trough moved further eastward
 272 by the westerlies, and the 300-hPa jet was significantly weakened. As a
 273 consequence of the influence of this convective system, a total rainfall of 14.4
 274 mm was reported at Tehran Mehrabad airport synoptic station from 0000
 275 UTC 14 April to 0000 UTC 15 April 2012.

276 **5. Results**

277 *5.1. Changes in the number and size of cloud droplets*

278 Microphysical properties of clouds are analyzed under different hygro-
 279 scopic aerosol number concentrations to understand the effects of aerosols on
 280 clouds development and precipitation processes. Aerosol number concentra-
 281 tions averaged over the innermost domain and in all vertical levels for the
 282 three experiments are presented in Table 1. They include 105.5 cm^{-3} in the
 283 clean, 574.7 cm^{-3} in the control and 2670.8 cm^{-3} in the polluted experiments.
 284 Changes in the initial and boundary hygroscopic aerosol number concentra-

285 tions were associated with significant changes in the number concentrations
 286 of cloud droplets. On average, the number concentrations of cloud droplets
 287 in cloudy grid points of the innermost domain were found to be 45.9, 158.3
 288 and 416.5 cm^{-3} in the clean, control and polluted experiments, respectively.
 289 This indicates that an increase in the number of hygroscopic aerosol con-
 290 centrations was associated with more activation of aerosols to form cloud
 291 droplets, consistent with previous observations (e.g. Andreae et al., 2004)
 292 and numerical studies (e.g. Fan et al., 2007; Li et al., 2008). Reduction of
 293 supersaturation is expected when a larger number of cloud droplets are com-
 294 peting for a fixed amount of available water vapor. With the increase in the
 295 number of hygroscopic aerosol concentrations, the mean cloud droplet size
 296 has been decreased. The mean effective radii of cloud droplets in cloudy grid
 297 points of the innermost domain averaged during the simulation period have
 298 been 8.53, 6.10 and $5.22 \mu\text{m}$ for the clean, control and polluted experiments,
 299 respectively, confirming the reduction in the mean size of cloud droplets with
 300 an increase in aerosol loading.

301 *5.2. Changes in cloud radiative forcing*

302 An increase in the number concentration, but a decrease in the mean size
 303 of cloud droplets in the polluted experiment is associated with an increase in
 304 the total droplet cross-sectional area, and consequently the shortwave cloud
 305 forcing, as shown in Figure 5 and Table 2, which is expected according to the
 306 "Twomey" effect. In the polluted experiment, reflection of solar radiation
 307 back to space by clouds is increased by 4.8% (5.1 W m^{-2}) in the innermost
 308 domain, implying that the first indirect effect of aerosols plays a major role
 309 in the atmospheric energy budget. In contrary, the shortwave cloud forcing

310 is decreased by 10.5% (11.1 W m^{-2}) in the clean compared to the control
 311 experiment. Higher reflection of solar radiation in the polluted experiment is
 312 also clearly evident in Figure 5, which is attributed to the discussed changes
 313 in the mean size of cloud droplets. The obtained values of shortwave cloud
 314 forcing are comparable to the previous estimates. For example, observational
 315 analysis by Kaufman et al. (2005) indicated that on the global scale com-
 316 bination of aerosol-cloud albedo and lifetime effects are associated with the
 317 forcing of $-11 \pm 3 \text{ W m}^{-2}$.

318 In contrast to the impacts on shortwave radiation, cloud forcing on long-
 319 wave radiation at the top of the atmosphere (TOA) is positive because clouds
 320 block longwave radiation emitted to space from the Earth's surface. Never-
 321 theless, as Table 2 indicates, in all experiments, the positive longwave cloud
 322 forcing is nearly half of the negative shortwave cloud forcing, suggesting that
 323 clouds have an overall cooling effect on the climate system. Nearly similar
 324 simulated values of the longwave cloud forcing in the three experiments sug-
 325 gest that changes in the CCN concentration have little effect on the longwave
 326 radiation, as previously noted by Albrecht (1989).

327 The identified net (shortwave + longwave) cooling effect when hygro-
 328 scopic aerosols are more abundant can lead to the stabilization of the daytime
 329 boundary layer, which can weaken or suppress formation of convective clouds.
 330 This thermodynamic impact is in addition to the radiative and microphysical
 331 impacts of aerosols on cloud and precipitation processes, although changes
 332 in cloud microphysics by aerosols have dynamical and thermodynamical con-
 333 sequences too.

334 5.3. *Other effects of aerosols on cloud properties*

335 Comparisons of the three conducted experiments indicate that cloud base
336 heights increase with the increase in the number of hygroscopic aerosol con-
337 centrations, such that the innermost domain- and time-averaged cloud bases
338 are found approximately at 1710.7, 1745.9 and 1773.3 m above the surface
339 for the clean, control and polluted experiments, respectively (Table 2). It
340 should be noted that the obtained cloud base heights in the three exper-
341 iments show only small differences, confirming the fact that variations in
342 aerosol concentrations have little impact on cloud development compared to
343 the changes in environmental conditions (e.g. see Grant and van den Heever,
344 2015). The increase of the cloud base height in the polluted experiment can
345 be related to the identified higher shortwave cloud forcing, which resulted in
346 more cooling effect at the surface. As a results, surface convection processes
347 are weakened, which caused simulation of a drier boundary layer in the pol-
348 luted experiment compared to the clean. Indeed, our analysis indicates that
349 throughout the day, near-surface air temperature is lower in the polluted
350 experiment compared to the other two experiments, while a drier boundary
351 layer is also simulated (not shown). It should be mentioned that the small
352 differences in the cloud base heights of the three experiments could be also
353 affected by the coarse vertical resolution of the model.

354 Figure 6 compares vertical profiles of mass concentrations of some hy-
355 drometeors under the clean, control and polluted aerosol conditions aver-
356 aged during the simulation period. The mass content of cloud droplets is the
357 highest in the polluted experiment because in response to higher concentra-
358 tions of aerosols, more aerosols are activated into cloud droplets; thus more

359 condensation of water vapor is occurred (Figure 6a). In other words, as the
360 number of hygroscopic aerosols is greater in the polluted experiment, more
361 water vapor has the chance to condense on aerosols and form cloud droplets.
362 On the other hand, the lowest content of cloud droplets is found in the clean
363 experiment, which is caused by less available aerosols to activate into cloud
364 droplets. In spite of the discussed differences in the mass concentration, the
365 height of the maximum cloud droplets content is nearly the same in the three
366 experiments because condensation of water vapor occurs mainly below the
367 freezing level, as previously stated by Li et al. (2008).

368 The discussed changes in the number of cloud droplets influence conden-
369 sation and efficiency of the autoconversion process from cloud droplets to rain
370 drops, such that overall, the rain drops content attains the greatest amount
371 in the clean experiment (Figure 6b). Less overall contents of rain drops in
372 the control and polluted experiments compared to the clean are due to the
373 fact that the effective radii of cloud droplets are reduced in more polluted
374 experiments (see Table 1); thus conversion of cloud droplets to rain drops
375 are hindered.

376 Figure 6c indicates that time-averaged ice production is maximum in the
377 clean experiment. As Pruppacher and Klett (1997) argued, this could be due
378 to the fact that immersion freezing increases with the formation of larger
379 droplets in the low CCN case. However, contribution of immersion freez-
380 ing in producing different ice production in the three experiments cannot
381 be examined in the present study because based on Thompson and Eidham-
382 mer (2014), condensation and immersion nucleation are grouped into a single
383 function following DeMott et al. (2010). Some other factors might have also

384 contributed. As previously discussed by Khain et al. (2001), clean clouds pro-
385 duce larger droplets which lead to rapid freezing at warmer temperatures. It
386 is not then surprising that the ice production in the lower atmosphere is
387 simulated to be the highest in the clean experiment. In spite of more mass
388 concentration of ice crystals, the clean experiment corresponds to the least
389 snow formation (Figure 6d), reflecting less conversion of ice to snow due to a
390 smaller number of ice crystals. The polluted experiment, on the other hand,
391 corresponds to the maximum snow formation. Overall, comparisons of rain
392 drops and snow contents in the polluted and clean experiments indicate that
393 polluted clouds produce more snow (due to more efficient mixed-phase pro-
394 cesses), but less rain (due to suppression of warm-rain processes) compared
395 to clean clouds. In fact, as polluted clouds produce less rain, more water
396 vapor is left to ascend from lower levels to the mid and upper troposphere
397 (Teller and Levin, 2006), causing more snow to form.

398 To better understand the impacts of aerosols on cloud microphysics, ver-
399 tical cross sections through the most cloudy region for the number concen-
400 trations of cloud droplets, rain drops and ice crystals for the clean, control
401 and polluted experiments at 0400 UTC 14 April 2012 are presented in Fig-
402 ure 7. Due to more available CCN, significantly higher cloud droplet number
403 concentration is found in the polluted experiment compared to the other two
404 experiments (Figure 7a-c). The least cloud droplet number concentration is
405 simulated for the clean experiment, which is caused primarily by the lack of
406 aerosol particles for the supercooled water to condense. Cloud droplets also
407 reach to greater heights in the polluted experiment, simply because droplets
408 are smaller in the polluted experiment; thus typically freeze at lower temper-

409 atures (higher in the cloud).

410 In contrast to the number concentration of cloud droplets, the number
411 concentration of rain drops is the highest in the clean experiment, while the
412 least is simulated for the control experiment (Figure 7d-f). This implies
413 that as cloud droplets are larger in the clean experiment (see Table 1), their
414 conversion into rain drops is more efficient. In all experiments, ice crystals
415 are primarily distributed above the 7-km level (Figure 7g-i). However, on
416 average, due to the lowest number of rain drops, the number concentration
417 of ice crystals is the highest in the control experiment. Suppression of warm
418 rain results in fewer rain drops and thus more cloud droplets that are lofted
419 into the colder regions of the cloud, allowing for the formation of more ice.

420 Vertical cross sections of the mass concentrations of hydrometeors at 0400
421 UTC 14 April 2012 are also shown in Figure 8. Due to more droplet conden-
422 sation, the highest mass content of cloud droplets is simulated in the polluted
423 experiment (Figure 8a-c), consistent with previous observations (Rosenfeld
424 and Woodley, 2000; Kaufman and Koren, 2006) and numerical simulations
425 (Fan et al., 2007) in cumulus clouds. On the contrary, on average the great-
426 est mass of rain drops is found for the clean experiment (Figure 8d-f), which
427 combined with the identified less cloud droplets, suggest that conversion from
428 cloud droplets to rain drops is more efficient in the clean experiment. The
429 highest concentration of ice crystals at this particular time is found for the
430 polluted case (Figure 8g-i), suggesting a hindered warm rain process, but
431 enhanced mixed-phase processes because of a greater number of elevated
432 aerosols.

433 Time-averaged liquid water contents, cloud fractions and surface rainfall

434 in the three experiments are depicted in Figure 9. In earlier studies, LWC
 435 was assumed to be constant as aerosol concentration underwent a change
 436 (Twomey et al., 1984). However, as evidenced by the results of the present
 437 study (Figure 9), and was first noted by Charlson et al. (1987), LWC varies
 438 with CCN concentrations. For example, an increase in LWC was identified
 439 in the ship tracks compared to the surrounding environment (Ferek et al.,
 440 2000). Figure 9 and Table 2 clearly indicate that a decrease in the number
 441 of CCN in the clean experiment is associated with a significant reduction
 442 in LWC. This is expected because when few aerosol particles activate, the
 443 result is few but relatively large drops, which results in a relatively low bulk
 444 condensation rate relative to a situation in which there are numerous small
 445 droplets. Similarly, a decrease in CCN concentrations resulted in an overall
 446 decrease in cloud fraction in the clean experiment (see Table 2 and Figure
 447 9).

448 5.4. *Precipitation response to aerosols*

449 Distributions of accumulated surface precipitation in the three experi-
 450 ments are also depicted in Figure 9. Although Table 1 indicates that the
 451 domain-averaged accumulated precipitation in the three experiments have
 452 little changed (with the highest amount in the clean experiment), significant
 453 changes in the amount of accumulated precipitation in different regions are
 454 detectable in Figure 9. Locally, both increase and decrease in surface pre-
 455 cipitation in response to changes in the number of aerosol concentration are
 456 evident, similar to the results of Thompson and Eidhammer (2014). Nearly
 457 in the center of the domain where the highest accumulated precipitation is
 458 simulated in the three experiments, rainfall has been much stronger in the

459 polluted experiment.

460 To better examine precipitation response to the number of aerosols, rain-
461 fall is categorized into three classes based on its intensity: light ($< 0.6 \text{ mm hr}^{-1}$), moderate ($0.6\text{-}1 \text{ mm hr}^{-1}$) and heavy ($\geq 1 \text{ mm hr}^{-1}$) rain. Figure
462 hr^{-1}), moderate ($0.6\text{-}1 \text{ mm hr}^{-1}$) and heavy ($\geq 1 \text{ mm hr}^{-1}$) rain. Figure
463 10 compares the percentage of precipitation amount for the three categories
464 in the clean and polluted experiments. It is evident that in the polluted
465 atmosphere, light rainfall is weakened, while moderate to heavy rainfall is
466 intensified. This implies that over the areas with an ample influx of water
467 vapor, an increment of hygroscopic aerosols can increase the amount of pre-
468 cipitation. On the other hand, over the areas with insufficient amounts of
469 water vapor, droplets have to compete for water vapor in order to grow; thus
470 further increment of hygroscopic aerosols reduce the precipitation amount.
471 The decrease of light rainfall, and the increase of moderate to heavy rainfall
472 in polluted clouds, i.e. shifting the rain distribution from light to heavy un-
473 der more polluted conditions, was also previously observed (e.g., Qian et al.,
474 2009; Li et al., 2011).

475 6. Conclusions

476 Computationally efficient two-moment aerosol-aware bulk microphysical
477 scheme developed and implemented into the WRF model by Thompson and
478 Eidhammer (2014) is used to investigate the impacts of aerosols on cloud
479 properties and precipitation. Three sensitivity experiments with different hy-
480 groscopic aerosol number concentrations were conducted: the control, clean
481 and polluted experiments. The aerosol number concentration represents the
482 current era conditions for the control experiment, while it is reduced to one-

483 fifth and increased by a factor of 5 in the clean and polluted experiments,
484 respectively.

485 The impacts of changing the aerosol number concentrations on shortwave
486 and longwave cloud forcing were investigated. In the polluted experiment,
487 more numerous cloud droplets of overall smaller size was identified, while
488 the reverse was true for the clean experiment. As a consequence, it was
489 found that an increase in the number of CCN in the polluted experiment
490 was associated with an increase in the amount of solar radiation reflected
491 back to space, while a little impact on the longwave radiation was identified.
492 The higher reflection of shortwave radiation by polluted clouds implies that
493 they have more cooling effect on the Earth-atmosphere system. This day-
494 time cooling aerosol first indirect effect partly compensates the greenhouse
495 warming, suggesting that less surface warming trend in the daytime com-
496 pared to nighttime in urban areas (Karl et al., 1984; Argüeso et al., 2014;
497 Alizadeh-Choobari et al., 2016) could be partly due to the aerosol-cloud ra-
498 diative effects. Due to a larger mean droplet size and less LWC, the least
499 shortwave cloud forcing was simulated in the clean experiment.

500 For the first time, it was found that the cloud base height responses to
501 the aerosol number concentrations, such that a higher cloud base height was
502 found under a polluted atmosphere. The identified higher cloud base height
503 in polluted clouds is caused by more surface cooling and a drier boundary
504 layer compared to clean clouds. In addition, decreases in LWC and cloud
505 fraction were identified in the clean experiment, while they were both in-
506 creased in the polluted experiment. The lower LWC in the clean experiment
507 was caused by less condensation of water vapor due to lower concentration of

508 hygroscopic aerosols. The increased LWC in the polluted experiment which
509 has been identified in the present study is in contrast to the results of Twomey
510 (1977) who assumed a constant LWC, but is similar to the results of Albrecht
511 (1989).

512 The impacts of changing the aerosol number concentrations on total pre-
513 cipitation were examined. Although the domain-averaged accumulated pre-
514 cipitation was little changed under both polluted and clean experiments,
515 noticeable changes in the spatial distribution and rate of precipitation were
516 identified. Generally, under polluted atmosphere light rain is reduced, while
517 moderate and heavy rain are both intensified, and the heaviest precipitation
518 changes the most. Our results for a convective cloud system in spring are
519 in contrast to the results of Thompson and Eidhammer (2014) who found
520 that for a cloud system driven by a frontal system in winter, the lightest
521 amount of precipitation changes the most. This implies that the response of
522 precipitation to aerosol loading strongly depends on the fact that whether
523 the region is influenced by a frontal system or a convective cloud system.

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Table 1. The innermost domain-averaged hygroscopic aerosol number concentration (cm^{-3}) averaged over the simulation period and in all vertical levels, as well as the innermost domain-averaged number concentration (cm^{-3}) and effective radius of cloud droplets (μm) in cloudy grid points averaged over the simulation period. The accumulated rainfall (mm) for the simulation period, from 0000 UTC 14 April to 0000 UTC 15 April 2012, for the clean, control and polluted experiments are also provided.

Experiments	Mean hygroscopic aerosol number concentration (cm^{-3})	Mean cloud droplet number concentration (cm^{-3})	Mean effective radius of cloud droplets (μm)	Accumulated rain (mm)
Clean	105.5	45.9	8.53	13.3
Control	574.7	158.3	6.10	12.7
Polluted	2670.8	416.5	5.22	13.1

Table 2. The innermost domain-averaged shortwave and longwave cloud forcing (SWCF and LWCF, W m^{-2}) at the top of the atmosphere (TOA), lifting condensation level (LCL) or cloud base (m) and liquid water content (LWC, mg m^{-3}) at cloud base averaged over the simulation period from 0000 UTC 14 April to 0000 UTC 15 April 2012 for the control, clean and polluted experiments.

Experiments	SWCF (W m^{-2})	LWCF (W m^{-2})	LCL (m)	LWC (mg m^{-3})
Clean	-94.8	50.7	1710.7	16.56
Control	-105.9	49.9	1745.9	16.83
Polluted	-111.0	50.3	1773.3	18.08

Figure 1. Multiple-nested model configuration of the three sensitivity experiments, with the grid spacing of 21000, 7000 and 2333 m for the mother and two nested domains, respectively. The initial hygroscopic aerosol number concentration (blue contours, cm^{-3}) at the surface in the control simulation and topographical features (shaded, m) are overlapped. The color bar indicates elevation above the mean sea level (AMSL).

Figure 2. The initial vertical profiles of (a) hygroscopic aerosol number concentration (cm^{-3}) and (b) ice-friendly aerosol number concentration (cm^{-3}) averaged over the innermost domain for the clean, control and polluted experiments. Note that the initial ice-friendly aerosol number concentration is identical for the three experiments.

Figure 3. Accumulated rainfall from 0000 to 2100 UTC 14 April 2012 a) observed by the Tropical Rainfall Measuring Mission (TRMM) satellite and b) simulated by the Weather Research and Forecasting (WRF) model in the mother domain, and profiles of c) air temperature (°C) and (d) wind speed (m s^{-1}) at 0000 UTC 14 April 2012 observed by a radiosonde (dashed lines) and simulated by the control experiment of the innermost domain of the WRF model (solid lines) at Mehrabad weather station (35.7° N , 51.3° E).

Figure 4. Mean sea level pressure (solid black contour, contour interval 4 hPa), geopotential height at 500 hPa (dashed brown contour, contour interval 4 dm) and wind speed at 300 hPa (shaded, m s⁻¹) in the mother domain at (a) 0000 UTC 14 April (b) 1200 UTC 14 April and (c) 0000 UTC 15 April 2012.

Figure 5. Shortwave (top, W m^{-2}) and longwave (bottom, W m^{-2}) cloud forcing at the top of the atmosphere (TOA) in the innermost domain averaged over the simulation period for the clean (left), control (middle) and polluted (right) experiments.

Figure 6. Vertical profiles of (a) cloud droplets, (b) rain drops, (c) ice crystals and (d) snow contents in mg m^{-3} averaged during the simulation period for the clean, control and polluted experiments. Averages were taken over grid points of the innermost domain with nonzero values.

Figure 7. Vertical cross sections of the number concentration of cloud droplets, rain drops and ice particles in cm^{-3} for the clean (left), control (middle) and polluted (right) experiments in the innermost domain at around 35°N at 0400 UTC 14 April 2012.

Figure 8. As in Figure 7, but for the mass concentration in mg m⁻³.

Figure 9. The liquid water content (g m^{-3}) and cloud fraction (CLDFRA) averaged over the simulation period, as well as accumulated rainfall in the innermost domain from 0000 UTC 14 April to 0000 UTC 15 April 2012 in the clean (left), control (middle) and polluted (right) experiments.

Figure 10. Probability distribution functions of three different precipitation categories under the clean and polluted experiments. Hourly rates of precipitation from 0000 UTC 14 April to 0000 UTC 15 April 2012 were computed through averaging over the innermost domain. The hourly precipitation values then were categorized into three classes based on the intensity, which include light ($< 0.6 \text{ mm hr}^{-1}$), moderate ($0.6\text{-}1 \text{ mm hr}^{-1}$) and heavy ($\geq 1 \text{ mm hr}^{-1}$). Values are percentage of precipitation amount under the three categories over the total precipitation amount.