

Aerosol radiative effects on mesoscale cloud–precipitation variables over Northeast Asia during the MAPS-Seoul 2015 campaign

Shin-Young Park¹, Hyo-Jung Lee¹, Jeong-Eon Kang¹, Taehyoung Lee², Cheol-Hee Kim^{1*}

¹Department of Atmospheric Sciences, Pusan National University, Busan 46241, Korea

²Department of Environmental Science, Hankuk University of Foreign Studies, Yongin, Korea

* Corresponding author: C.-H. Kim

Department of Atmospheric Sciences, Pusan National University

2, Busandaehak-ro 63beon-gil, Geumjeong-gu, Busan, 46241, Rep. of KOREA

Tel.: 82-51-510-3687

Fax: 82-51-515-1689

E-mail address: chkim2@pusan.ac.kr (C.-H. Kim)

1 **(Abstract)** The online model, Weather Research and Forecasting Model with Chemistry (WRF-Chem)
2 is employed to interpret the effects of aerosol-cloud-precipitation interaction on mesoscale
3 meteorological fields over Northeast Asia during the Megacity Air Pollution Study-Seoul (MAPS-
4 Seoul) 2015 campaign. The MAPS-Seoul campaign is a pre-campaign of the Korea-United States Air
5 Quality (KORUS-AQ) campaign conducted over the Korean Peninsula. We validated the WRF-Chem
6 simulations during the campaign period, and analyzed aerosol-warm cloud interactions by diagnosing
7 both aerosol direct, indirect, and total effects. The results demonstrated that aerosol directly decreased
8 downward shortwave radiation up to -44% (-282 W m^{-2}) for this period and subsequently increased
9 downward longwave radiation up to +15% ($\sim 52 \text{ W m}^{-2}$) in the presence of low-level clouds along the
10 thematic area. Aerosol increased cloud fraction indirectly up to ~24% with the increases of both liquid
11 water path and the droplet number mixing ratio. Precipitation properties were altered both directly and
12 indirectly: Direct effects simply changed cloud-precipitation quantities via simple updraft process
13 associated with perturbed radiation and temperature, while indirect effects mainly suppressed
14 precipitation, but sometimes increased precipitation in the higher relative humidity atmosphere or near
15 vapor-saturated condition. The total aerosol effects caused a time lag of the precipitation rate with the
16 delayed onset time of up to 9 hours. This implies the importance of aerosol effects in improving
17 mesoscale precipitation rate prediction in the online approach in the presence of non-linear warm cloud.

18

19 **Keywords:** WRF-Chem, Aerosol-cloud-radiation interaction, Aerosol effects, MAPS-Seoul 2015

20

21

22

23

24

25 **1. Introduction**

26 Aerosol particles directly absorb and scatter solar and thermal infrared radiation, altering the
27 radiative balance of the Earth's atmospheric system. This effect is referred to as the direct effect
28 (Haywood and Boucher, 2000). Aerosol indirectly affects both cloud albedo and cloud lifetime,
29 referred to as first and second indirect effects (Twomey, 1977; Albrecht, 1989; Pincus and Baker, 1994;
30 Haywood and Boucher, 2000; Rosenfeld et al., 2008). The first and second indirect effects control the
31 cloud droplet number, cloud albedo, and cloud lifetime and heights, either suppressing or enhancing
32 precipitation. In addition to these two processes, substantially more complicated aerosol-cloud
33 interactions have been revealed (Rosenfeld et al., 2014). As a result, a better quantification of the ever-
34 increasing effects of aerosol-cloud-radiation interaction and its application of feedback to the daily
35 weather prediction are also open questions, particularly in urban and polluted areas.

36 Northeast Asia emits large amounts of organic aerosols (OA), anthropogenic volatile organic
37 compounds (VOCs), and particulate matter including PM_{2.5} and PM₁₀ (Piccot et al., 1992; Saikawa et
38 al., 2009; Jiang et al., 2012). Specifically, the eastern regions of China, including Northeast, East, and
39 Southeast China generate pollution composed of various emission components (Ohara et al., 2007; Li
40 et al., 2014; Li et al., 2017). Thus, characteristics associated with the development of cloud water vary
41 depending on aerosol components and polluted regions. For example, Northeast China with
42 temperatures below freezing is not favorable for warm cloud formation, and the North China Plain
43 (NCP) has favorable conditions for long-lasting haze formation but not for cloud and precipitation
44 formation, because of its relatively low amount of cloud water (Zhou et al., 2016). While East and
45 Southeast China, with abundant cloud water, are one of the most favorable areas for cloud and
46 precipitation formation, with increased aerosol activation and warm cloud development (Fan et al.,
47 2012; Zhang et al., 2015). Strong air pollution characteristics over the upstream areas greatly affect
48 downstream areas in terms of sources of emissions through the long-range transport of air masses over

49 Northeast Asia (Carmichael et al., 2002; Park et al., 2005; Kim et al., 2012; Wang et al., 2016; Kim et
50 al., 2017a). Thus, the downstream area located east of the Chinese industrial area, which includes the
51 Korean Peninsula, is expected to be significantly influenced by aerosol-cloud-radiation interactions.

52 To account for these aerosol effects, it is desirable to use the online-coupled meteorology-
53 chemistry-aerosol model, which has been applied in regional meteorology and air quality simulations
54 (Rieger et al., 2014; Tao et al., 2015; Zhou et al., 2016; Hong et al., 2017). The Weather Research and
55 Forecasting model with Chemistry (WRF-Chem) is one such fully coupled online model (Grell et al.,
56 2005; Peckham et al., 2011). The WRF-Chem is capable of simulating feedback between aerosols and
57 meteorology for various atmospheric processes by linking aerosol optical properties to the radiation
58 scheme and cloud condensation nuclei (CCN) potential to the microphysics scheme (Gustafson et al.,
59 2007; Chapman et al., 2009). Several studies revealed that meteorological fields and air quality
60 significantly improve model performance when aerosol feedback is considered in the WRF-Chem
61 simulation (Chapman et al., 2009; Wang et al., 2015b; Wu et al., 2017). Previous studies using WRF-
62 Chem for indirect effects focused on marine stratocumulus clouds with relatively clean air quality
63 conditions, which can be resolved at coarser resolutions (Yang et al., 2011; Saide et al., 2012;
64 Grosvenor et al., 2017). Furthermore, other studies point out that aerosol feedback focuses on either
65 global or seasonal scales and ignores shorter time scales (Gao et al., 2017; Song et al., 2017).

66 Previous analyses that employed satellite data also stressed the importance of aerosol-cloud-
67 radiation interaction over Northeast Asia (Wang et al., 2014; Kourtidis et al., 2015; Wang et al., 2015a).
68 For example, some studies highlight the negative relationship between cloud droplet radius and aerosol
69 optical depth (AOD) over the East China Sea, which is in agreement with the first indirect effect known
70 as Twomey's effect (Rosenfeld, 2000; Kim et al., 2003). Other researches (e.g., Wang et al., 2014;
71 Kourtidis et al., 2015) exploring the relationship between AOD and cloud cover overestimate
72 (underestimate) the AOD impact on cloud cover in regions where AOD and water vapor have similar

73 (opposite) seasonal variations. However, these findings also have many limitations in quantitatively
74 determining the mechanism of aerosol effect for two main reasons. The first is the inability to
75 distinguish the contribution of aerosols to the observed cloud variation, and the second is the difficulty
76 in obtaining region-specific observation data such as the vertical profile and absorption of aerosols.

77 The present modeling study was conducted as part of the Megacity Air Pollution Study-Seoul
78 (MAPS-Seoul) 2015 campaign from 18 May 2015 to 13 June 2015 and investigated the manner in
79 which regional aerosol-cloud-radiation interactions are captured in WRF-Chem. The study period was
80 the MAPS-Seoul 2015 campaign, which is a pre-campaign of the Korea-United States Air Quality
81 (KORUS-AQ) study. KORUS-AQ is an international, multi-organizational mission created to observe
82 air quality across the Korean Peninsula and its surroundings (Lee et al, 2016; Kim et al., 2017b).

83 The objectives of this study are to evaluate the complex interactions between aerosol and
84 meteorological fields using the coupled online WRF-Chem model over Northeast Asia, where sources
85 of abundant aerosols create favorable conditions for developing warm cloud processes. Furthermore,
86 this study characterizes the influences of aerosol feedback on meteorological fields during the MAPS-
87 Seoul 2015 campaign. And we diagnose aerosol effects to improve meteorological and chemical
88 forecasting for shorter time scales such as diurnal or weekly cycles.

89

90 **2. Data and methodology**

91 **2.1 MAPS-Seoul 2015 campaign and aircraft data**

92 During the MAPS-Seoul 2015 campaign period, seven research flights (RFs), RF1 to RF7, were
93 carried out near the Seoul Metropolitan Area (SMA) in Korea. Our modeling domain and flight tracks
94 are presented in Fig. S1 and the flight numbers and take-off/landing times are reported in Table S1.
95 RF1 was test flight without spiral flight over SMA and RF2 flew to different two heights over the
96 Yellow. RF3 and RF6 included spiral flights over the Yellow Sea to analyze the vertical structures of

97 aerosols. RF5 travelled to the metropolitan cities of Gwangju and Busan, and returned to base (Fig.
98 S1b). A High Resolution Time-of-Flight Aerosol Mass Spectrometer (HR-ToF-AMS, Aerodyne
99 Research Inc., Billerica, MA, USA) and an Ultra-High Sensitivity Aerosol Spectrometer (UHSAS,
100 Droplet Measurement Technologies Inc., Boulder, CO, USA) were installed on the Hanseo KingAir-
101 C90GT aircraft (Textron Aviation Inc., Wichita, KS, USA). In this study, RF4 and RF7 were used to
102 evaluate the results of the WRF-Chem model, because both flights show the meteorological signal of
103 the long-range transport process from China. RF4 and RF7 flew from Taean (base) to Seoul and Wonju,
104 took samples below an altitude of 2,500 m, and then returned along a similar track.

105 All seven RF excursions were equipped with the HR-ToF-AMS onboard during the campaign.
106 The operation of the HR-ToF-AMS is based on ambient air pulled through a cyclone (URG
107 Corporation, Chapel Hill, NC, USA) at a flow rate of 3 L min⁻¹ to remove particles with aerodynamic
108 diameters larger than 2.5 μm in front of the HR-ToF-AMS. Ambient air enters the HR-ToF-AMS
109 through a pressure drop critical orifice 110 μm in diameter with an average inlet flow rate of 0.105 L
110 min⁻¹. An aerodynamic lens focuses ambient particles of ~35 nm to ~1 μm into a narrow beam to create
111 the particle beam, which then flows into the Particle Time-of-Flight (PToF) region. Non-refractory
112 particles, with the aerodynamic diameter of $d < 1 \mu\text{m}$, are vaporized on impact with a resistively heated
113 surface at ~600 °C and ionized by electron ionization at 70 eV. The operation of the HR-ToF-AMS is
114 described in detail elsewhere (Jayne et al., 2000; Jimenez et al., 2003; Drewnick et al., 2005; DeCarlo
115 et al., 2006; Canagaratna et al., 2007; Lee et al., 2015).

116 In this study, the HR-ToF-AMS was operated under V-mode, and an MS mode of 20 s time
117 resolution was used to determine the compositions of non-refractory particles including the
118 concentrations of OA, nitrate, sulfate, ammonium and chloride. The HR-ToF-AMS was calibrated for
119 ionization efficiency (IE) by using 350 nm ammonium nitrate particles at a number density of ~300

120 cm^{-3} . The values of the relative IE for other aerosol types including OA, sulfate, nitrate, ammonium
121 and chloride were derived from default values.

122

123 **2.2 Other observational data used in this study**

124 The WRF-Chem model results were compared with meteorological observations. The
125 meteorological observation data of 91 stations located over the southern part of the Korean Peninsula
126 were obtained from the Korea Meteorological Administration (KMA) (Fig. S1b). The Tropical Rainfall
127 Measuring Mission (TRMM) was also employed in this study to validate the simulated precipitation
128 results (Huffman et al., 2007). TRMM is an integral part of the National Aeronautics and Space
129 Administration (NASA) Earth Science Enterprise, and provides satellite-derived estimates of
130 tropical/subtropical precipitation across the globe and estimates the associated latent heating. The
131 3B42 product produces three-hourly merged high-quality infrared and microwave precipitation
132 estimates at a spatial resolution of $0.25^\circ \times 0.25^\circ$. The spatial coverage extends from 50°S to 50°N
133 latitude, covering our study domain over Northeast Asia.

134

135 **2.3 Model, configuration, and data used**

136 The employed meteorological model, WRF is a fully compressible and non-hydrostatic mesoscale
137 numerical weather prediction model (Skamarock et al., 2008). WRF-Chem is an online-coupled
138 meteorology-chemistry-aerosol model that can simultaneously simulate trace gases and aerosols with
139 WRF meteorological fields. The air quality and meteorological components use the same advection
140 scheme, grid, and physics schemes for sub-grid scale transport (Grell et al., 2005; Fast et al., 2006).
141 Version 3.7.1 of WRF-Chem was employed in this study. Table S2 summarizes the model configuration.

142 WRF-Chem was designed to simulate aerosol-cloud-radiation interaction with existing model
143 treatments for various feedback processes of direct and indirect aerosol effects. To calculate the direct

144 effect, aerosol optical properties such as extinction, single scattering albedo, and the asymmetry factor
145 for scattering were calculated as functions of wavelength and three-dimensional position. The current
146 study employed the Rapid Radiative Transfer Model for the General Circulation Models (RRTMG)
147 scheme to generate these radiative properties. Each aerosol chemical component has its own associated
148 complex refractive index calculated according to the volume averaging approximation for each size
149 mode. The Mie theory was applied to estimate extinction and scattering efficiencies. For efficient
150 computation, the methodology described by Ghan et al. (2001) was employed. The net radiative
151 impacts were calculated by passing the bulk optical properties of the aerosol layer to the radiative
152 transfer parameterization. Fast et al. (2006) and Barnard et al. (2010) describe the aerosol optical
153 property calculation in detail.

154 For aerosol indirect effects, aerosol-cloud interaction is a complicated process that involves
155 activation and resuspension of aerosol particles, aqueous chemistry, and wet removal. The activation
156 process in WRF-Chem is based on the maximum supersaturation, which is determined from a Gaussian
157 spectrum of updraft velocities and the bulk hygroscopicity of each aerosol compound for all log-normal
158 modes of particles (Abdul-Razzak and Ghan, 2000, 2002). Wet removal of aerosols and trace gases is
159 treated both in and below clouds. Within clouds, aerosols and trace gases dissolved in the water are
160 collected by precipitation such as rain, snow, and graupel. The wet scavenging module by precipitation
161 below clouds was parameterized following the method of Easter et al. (2004). However, because of
162 the irreversible removal process of aerosols by rain, resuspension of aerosol particles from evaporated
163 rain is not possible in the current version of the model.

164 To simulate the indirect effects, the scheme of activation, resuspension, and wet scavenging for
165 aerosol particles needs to be coupled with a double-moment microphysical scheme that predicts the
166 number and mass concentrations for hydrometeors. The Morrison microphysics scheme used in this
167 study includes the prognostic treatment of the cloud droplet number concentration, which considers

168 the losses caused by collision, coalescence, collection, and evaporation, and the sources caused by
169 nucleation. In addition, the Morrison scheme accounts for the autoconversion of cloud water to rain
170 water depending on the cloud droplet number (Liu et al., 2005). Therefore, aerosol activation affects
171 both the rain rate and liquid water content. The couplings were implemented for warm cloud processes
172 only, and direct links of aerosols with ice nuclei (IN) were not considered in this study.

173 In this study, a 27 km \times 27 km horizontal resolution was employed in the study domain (Fig. S1),
174 which covers Northeast Asia (100°E–150°E longitude, 20°N–50°N latitude) with 174 (W–E) \times 128
175 (S–N) grid points and 15 vertical layers. The horizontal resolution was settled considering domain and
176 objective of this study. According to previous studies (e.g., Gao et al., 2015; Gao et al., 2017), the
177 aerosol feedback on meteorological fields was simulated well by WRF-Chem with horizontal
178 resolution of 27km. Initial and boundary conditions for the meteorological variables were obtained
179 from 6-hourly 1° \times 1° National Centers for Environmental Prediction (NCEP) Final Analysis (FNL)
180 data (NCEP, 2000). The chemical boundary conditions for the main trace gases and particulate matter
181 concentrations were provided by simulations by the Model for Ozone and Related Chemical Tracers
182 (MOZART) (Emmons et al., 2010). In the chemical initial condition process, this study employed the
183 model containing re-initialized NCEP meteorological fields for each day of a five-day run, recognized
184 as sufficient for the initial conditions of chemical variables to reach equilibrium.

185 The anthropogenic emissions of sulfur dioxide (SO₂), nitrogen oxides (NO_x), carbon monoxide
186 (CO), VOCs, black carbon (BC), organic carbon (OC), PM₁₀, and PM_{2.5} were based on MAPS-Seoul
187 2015 campaign emission data (Woo et al, 2014). This emission inventory includes the Multi-resolution
188 Emission Inventory of China (MEIC) (<http://www.meicmodel.org/>), Clean Air Policy Support System
189 (CAPSS) (<http://airemiss.nier.go.kr>), and Regional Emission Inventory in Asia (REAS) version 2
190 (<http://www.jamstec.go.jp/frsgc/research/d4/emission.htm>). Biogenic emissions were calculated
191 online using a model based on the Model of Emissions of Gases and Aerosols from Nature (MEGAN)

192 inventory (Guenther et al., 2006). MEGAN has been fully coupled into WRF-Chem to enable the
193 online calculation of biogenic precursor emissions subject to vegetation cover and meteorological
194 conditions such as temperature and solar radiation at the time of the calculation (Grell et al., 2005).
195 Dust, sea salt, and dimethylsulfide (DMS) emissions were not included in this study.

196

197 **2.4 Model experiments and case description**

198 As a case study of the MAPS-Seoul 2015 campaign, two cases with warm cloud were selected.
199 The simulations of these cases were run for 120 h with the first 24 h discarded as model spin-up for
200 meteorology-chemistry interactions. Case A began at 00 UTC on 5 June 2015, and Case B started at
201 the same time on 10 June 2015 (local time: UTC+9 h).

202 To investigate the impact of aerosol effects on mesoscale meteorological fields, three experiments
203 for the two cases were conducted: a control experiment (**CTL**) with no aerosol effects, an aerosol-
204 radiation interaction experiment (**ARI**), and an aerosol-cloud-radiation interaction experiment (**ACR**).
205 The CTL did not include any explicit aerosol-meteorology interactions and the ARI considered only
206 radiative effects by including the aerosol direct effect on radiation without cloud effects. The ACR
207 reflected the radiation effects of ARI plus additional aerosol-cloud interactions for grid-scale clouds.
208 Therefore, the total (= direct + indirect) aerosol effects on meteorological fields were included in the
209 ACR experiment. The differences in each experiment represent the direct effect, indirect effect, and
210 total effect. For example, the difference between ACR and CTL (i.e., ACR - CTL) was to investigate
211 the total aerosol effects, whereas the difference between ARI and CTL (i.e., ARI - CTL) and that
212 between ACR and ARI (i.e., ACR - ARI) represented the aerosol direct and indirect effects, respectively.
213 Table S3 summarizes the experiments.

214 Fig. 1 shows the meteorological and aerosol fields over specific 24-h periods for both case study
215 periods. Figs. 1a and 1b show the results of the ACR experiments from 12 UTC on 7 June (Case A),

216 and Figs. 1c and 1d show those from 00 UTC on 11 June (Case B). Figs. 1a and 1c show the
217 accumulated precipitation together with wind fields at 850 hPa, and Figs. 1b and 1d show the total
218 AOD at 550 nm and the liquid water path (LWP) at 500 hPa for both cases. Case A shows a high-
219 pressure system over the eastern (China) and western (Japan) parts of the domain as well as a low-
220 pressure system over the northern part (Fig. 1a). These pressure distributions led to a strong streamline
221 through the East China Sea and East Sea of Korea with a southwesterly wind. In the modeled
222 meteorological conditions of Case A, widespread precipitation of less than 15 mm day⁻¹ occurred near
223 the Korean Peninsula. In Fig. 1b, a high simulated AOD value of more than 0.7 with clouds (LWP >
224 0.1 g m⁻²) was discovered over the southern region of the Korean Peninsula. Even though the region
225 with clouds was excluded, the AOD was higher than 0.9 over the Yellow Sea. This indicates that the
226 aerosol loading was transported to the Korean Peninsula via the Yellow Sea during the period of Case
227 A. Case B is characterized by continuous westerly winds from China to the downwind regions under
228 the influence of a cyclone system centered on the Northeast China region in Manchuria (Fig. 1c).
229 Compared with Case A, minimal precipitation of <10 mm day⁻¹ was simulated over South Korea. As
230 the transport process of air mass from China progressed, high aerosol loadings were clearly detected
231 over the Korean Peninsula (Fig. 1d).

232

233 **3. Results**

234 **3.1 Model evaluation**

235 To verify the model's representation of our aerosol-cloud-radiation feedback experiment, model
236 performance was evaluated by comparing the simulated meteorological variables and aerosol loadings
237 against surface, aircraft measurements, and satellite data. Table 1 provides a statistical summary of the
238 comparisons for meteorological variables including temperature, relative humidity (RH), and wind
239 speed for the ACR experiments for both cases. The model depicted the temperature and its temporal

240 variation with a Corr. R of 0.93 to 0.94 and an MB of -1.38 to -0.95 °C. RH was also reasonably
241 reproduced with Corr. R values of 0.86 to 0.93 and MB of 8.95 to 10.47. The simulated wind speed
242 agreed well with the observation of Corr. R = 0.70 to 0.77, but was systematically overestimated for
243 Case A with an NMB of 112%. Noted that in previous studies, a high positive bias (e.g., NMB of 78.1
244 to 105%) in wind speed was often obtained in WRF-Chem simulations (Matsui et al., 2009; Zhang et
245 al., 2010; Molders et al., 2012; Tuccella et al., 2012; Gao et al., 2015; Zhang et al., 2015). This high
246 bias is partly attributed to unresolved topographical features in surface drag parameterization and the
247 coarse resolution which were all previously studied (Cheng and Steenburgh, 2005; Yahya et al., 2014;
248 Zhang et al., 2015).

249 Observed and simulated precipitation rates (PR) were estimated at three sites, namely Gosan,
250 Jinju, and Incheon, where precipitation was observed during the study periods (Fig. 2). The model
251 results overestimated the PR for all experiments. However, the results of the model led to higher threat
252 score values: 0.25~0.29 for ACR experiment with an NMB reduction of about 25% compared to the
253 CTL and ARI experiments. Furthermore, the onset detection of PR demonstrated a time lag for the
254 ACR experiment compared to other experiments, with a delay of onset time of nine and two hours at
255 Gosan and Jinju, respectively. In addition, the peak time of PR also demonstrated a time lag with a
256 delay of nine and four hours at Gosan and Jinju, respectively (Figs. 2a and 2b). This suggests that the
257 ACR experiment, with full aerosol feedback effects, simulated a higher probability of precipitation
258 than the other experiments. Fig. 3 indicates the spatial distributions of the PR, showing a stronger
259 similarity to the distribution of the TRMM product in the ACR. In the ACR experiment, the PR was
260 less intense and covered a smaller area over the Korean Peninsula at 21 UTC on 7 June. And the shape
261 of the precipitation band had similar patterns to the TRMM observation at 21 UTC on 11 June (Fig.
262 3). The analysis above indicates that the ability to simulate precipitation, including the presence and
263 rate of precipitation improved in the ACR experiment.

264 To investigate the chemical compositions of the aerosols in detail, the aircraft measurements of
265 RF7 for Case B were compared with those of RF4 for Case A. Here, noted that the flight paths of RF4
266 and RF7 were similar, with the aircraft traveling to Seoul (Fig. S1). Fig. S2 shows that PM_{2.5}, sulfate,
267 and ammonium for RF4 were simulated well, whereas those for RF7 were slightly underestimated at
268 about 15, 5, and 1 $\mu\text{g m}^{-3}$ for each of the three chemical components, respectively. This suggests that
269 the lack of formation mechanisms of secondary inorganic aerosols, such as the heterogeneous reaction
270 of SO₂ on the surface (Harris et al., 2013), is a possible reason for the underprediction. The modeled
271 OA was significantly underestimated for both RF4 and RF7 flights, although OA had a larger fraction
272 of total mass for the aircraft measurements. The capability of OA forecasting of the WRF-Chem is
273 suggested as a reason for underestimation. Therefore, with the exception of OA, the overall simulated
274 aerosol mass could be reasonably compared quantitatively to observed values.

275

276 **3.2 Aerosol effects on radiative variables**

277 To evaluate how well the WRF-Chem model simulated the impact of aerosol feedback on
278 mesoscale meteorological fields, the radiation-cloud variables were analyzed. The three experiments
279 were investigated by two different sky conditions: clear-sky and all-sky as in a previous study (Archer-
280 Nicholls et al., 2016). The clear-sky variables were calculated for the grid points with no clouds,
281 yielding results without the influence of clouds in the model. The all-sky condition denotes a clear sky
282 plus cloudy sky conditions. To compare the radiation-cloud variables with the corresponding modeled
283 aerosol loading, the resultant vertical profiles of the extinction coefficient at 550 nm over South Korea
284 for the two cases in the ACR experiment were examined.

285 Fig. 4 shows a time series of various aerosol effects on downward shortwave (SW) radiation in
286 clear- and all-sky conditions for both cases at ground level averaged across South Korea. In the case
287 of clear-sky conditions, as indicated in Figs. 4a and 4b, the total effects on the changes in downward

288 SW radiation at the ground (denoted as $\Delta SW_{\text{sfc,clr}}^{\downarrow}$), with maximum values of -112.2 and -144.6 W m⁻²
289 ² for Case A and Case B, originated mostly from direct effects. As expected, only small indirect effects
290 of ~26.2 W m⁻² at 07 UTC on 8 June (Fig. 4a) and ~50.1 W m⁻² at 06 UTC on 11 June (Fig. 4b) were
291 found for Case A and Case B, respectively. However, in the all-sky condition, the radiative variables
292 with cloud fields tended to change more dramatically than in the clear-sky condition. In Figs. 4c and
293 4d, the more sharply reduced downward SW radiation shown in the all-sky condition (denoted as
294 $SW_{\text{sfc}}^{\downarrow}$) is mostly attributed to the indirect effects, especially for Case B. This difference between direct
295 and indirect effects was more obvious at 03 UTC on 11 June for Case B with -11.4 W m⁻² (-1.8 %
296 against CTL) for direct effects, but -271.0 W m⁻² (-42.6 % against CTL) for indirect effects, where
297 cloud fraction (CF) of up to 40% was simulated.

298 Whereas almost sinusoidal daily variations in direct effects contribute for both clear- and all-sky
299 conditions, indirect effects demonstrated more intermittent patterns with sharply increased
300 contributions depending on CF for all-sky conditions. For example, the maximum decrease of $SW_{\text{sfc}}^{\downarrow}$
301 in the indirect effects including cloud impact were simulated as -125.5 to -271.0 W m⁻² (-23.1 to -
302 42.6 %) for Case A and Case B, respectively (Figs. 4c and 4d). At the same time, the gaps between
303 $\Delta SW_{\text{sfc,clr}}^{\downarrow}$ and $\Delta SW_{\text{sfc}}^{\downarrow}$ reached 99.3 W m⁻² (20.1 %) and 220.1 W m⁻² (36.7 %) for the two cases,
304 respectively.

305 Fig. 5 illustrates the vertical profiles of the extinction coefficient for both cases, showing some
306 agreement compared to the variation in $SW_{\text{sfc,clr}}^{\downarrow}$ and $SW_{\text{sfc}}^{\downarrow}$. The radiative variables are attributed
307 mainly to the scattering and absorption of solar radiation. The reduction of downward SW radiation by
308 aerosol varies as amount of solar radiation begin to deviate after sunrise, and in turn tends to change
309 more dramatically by the presence of high aerosol loadings. The results changed drastically in $SW_{\text{sfc}}^{\downarrow}$
310 reaching up to -271.0 W m⁻² when the extinction coefficient was more than 0.22 km⁻¹ near the surface.

311 This indicates that the reduction of downward SW radiation by aerosols is associated with the
312 extinction coefficient profile directly and indirectly, particularly in conditions of high aerosol loading.

313 Regarding upward SW radiation at the ground (denoted as SW_{sfc}^{\uparrow}), it is known that the portion of
314 incoming radiation is reflected at the surface and strongly dependent on the surface albedo. This study
315 obtained the same results. However, we also determined that the simulated $\Delta SW_{sfc,clr}^{\uparrow}$ and $\Delta SW_{sfc}^{\uparrow}$
316 are approximately 15% (or less) of $\Delta SW_{sfc,clr}^{\downarrow}$ and $\Delta SW_{sfc}^{\downarrow}$ with the same temporal variations.

317 In this study, we excluded the change in upward longwave (LW) radiation by aerosol, because no
318 significant biases were found. In general, upward LW radiation in the clear-sky condition (denoted as
319 $LW_{sfc,clr}^{\uparrow}$) at the ground depends mostly on the changing skin temperature based on the Stefan-
320 Boltzmann law as a function of the variations in solar radiation (Stefan, 1879; Boltzmann, 1884). The
321 modeled $LW_{sfc,clr}^{\uparrow}$ was in phase with the surface skin temperature. In our WRF-Chem simulations, the
322 LW radiation change between the all-sky condition (denoted as LW_{sfc}^{\uparrow}) and $LW_{sfc,clr}^{\uparrow}$ was small and
323 negligible (not shown).

324 Figs. 6a and 6b show the time series of the differences in downward LW radiation at the ground
325 averaged across South Korea for both cases. As shown in Figs. 6a and 6b, the total effects on downward
326 LW radiation at the ground (denoted as LW_{sfc}^{\downarrow}) were similar to the indirect effects, whereas the direct
327 effects were small and negligible with a difference of -6.5 to 8.1 $W m^{-2}$ (-1.8 to 2.6%) for Cases A and
328 B. The total aerosol effects can lead to large increases in $\Delta LW_{sfc}^{\downarrow}$ with maximum values of 30.8 to
329 51.8 $W m^{-2}$ (9.7 to 15.0 %) for both cases, whereas the common averaged values across South Korea
330 ranged from -7.2 to 10.5 $W m^{-2}$ (-1.9 to 2.7 %).

331 To analyze the total effects in interpreting the changes in LW_{sfc}^{\downarrow} , cloud parameters were examined
332 in this study. Figs. 6c and 6d illustrate the cloud water mixing ratio (QCLOUD) for both cases. In Figs.
333 6c and 6d, the variation of QCLOUD was seen extensively by the high aerosol loading through the

334 process of aerosol-cloud interaction. Upon the condition of all large increases of $\Delta LW_{sfc}^{\downarrow}$ with a
335 maximum value of 51.8 W m^{-2} (15.0 %), significant growth in cloud formation was found particularly
336 in the condition of $\Delta QCLOUD$ exceeding 110 mg kg^{-1} , particularly at altitudes below 950 hPa (Figs.
337 6c and 6d). This is suggesting the increased $QCLOUD$ reflecting the LW radiation, especially at near
338 the surface. Therefore, a portion of LW radiation emitted from the surface is trapped by low-level
339 clouds, resulting in radiative warming at the ground. However, the magnitude of $\Delta LW_{sfc}^{\downarrow}$ was
340 determined as relatively small compared to $\Delta SW_{sfc}^{\downarrow}$.

341

342 **3.3 Aerosol effects on clouds and precipitation**

343 To investigate the manner in which aerosol-cloud-radiation interaction affects the cloud
344 microphysics of the atmosphere, the associated CF and PR were analyzed. Fig. 7 displays the time
345 series of CF and PR averaged over South Korea for the three experiments. Of the three experiments,
346 the simulated CF showed the highest values in the ACR experiment for both cases. The direct effects
347 contributed to CF values of -6.7% to 3.8%, whereas the indirect effects significantly increased in CF
348 difference (ΔCF) up to 22.7% (Figs. 7b and 7f), controlling the cloud lasting time. This indicates the
349 importance of the second indirect effects associated with changing the cloud cover and cloud lifetime.

350 Our study also correlated the difference in CF by indirect effects with that of low-level $QCLOUD$,
351 as shown in Figs. 6c and 6d. Comparing Figs. 4c and 4d to Figs. 7b and 7f strongly associated the
352 phase of indirect effects on CF and other cloud variables such as $QCLOUD$ with the radiative variables.
353 This implies that the perturbation in radiation properties yields cloud formation involving indirect
354 effects and vice versa; therefore, the interaction of clouds and radiation properties is undoubtedly
355 significant in the prediction of cloud cover and amount.

356 In Figs. 7d and 7h, the fluctuation in PR by aerosol effects was shown to occur at a similar period
357 when CF was increased by the total effects, but showed a relatively low correlation with cloud

358 variations, likely because of nonlinear cloud to precipitation conversion processes. As shown in Figs.
359 7c and 9g, the ACR experiment decreased precipitation more than other experiments. This result is
360 consistent with those from observed PR, as illustrated in Fig. 2. Fig. 2 indicates that the ACR
361 experiment better agreed with observations, especially at Gosan., implying that aerosol-cloud-radiation
362 interaction is an important factor for improving daily weather prediction of precipitation. Similar
363 results are reported by numerous studies that performed an inter-comparison of coupled chemistry-
364 meteorology models over Europe, such as Forkel et al. (2012), Brunner et al. (2015), and Kong et al.
365 (2015). Their studies considered the importance of aerosol, cloud, and radiation interaction, validating
366 that the simulation with aerosol feedback more accurately reproduced microphysical variables and
367 incoming downward SW radiation.

368 Fig. S3 illustrates the mean spatial distributions of LWP simulated from the ACR experiment and
369 its attribution fraction of aerosol total effects over specific 24-h periods of the two cases. Here, Figs.
370 S3a and S3c represent LWP (g m^{-2}), and Figs. S3b and S3d illustrate the contribution fraction of total
371 effects to LWP, respectively. As shown in Fig. S3, cloud formation occurred mainly over Southeast
372 and East China, the Korean Peninsula, and East China Sea (Figs. S3a and S3c). In Figs. S3b and S3d,
373 the LWP showed a spatially increased pattern nearly proportional to the LWP by the total aerosol
374 effects, mostly due to indirect effects. The LWP was greatly augmented to 614.8 and 518.9 g m^{-2} for
375 Cases A and B, respectively. This implies that the simulated LWP was influenced by aerosol indirect
376 effects, as shown in Figs. 7b and 7f, which alter cloud properties such as liquid water content, CF and
377 cloud lifetime.

378 These augmentations in LWP can be attributed to the increase in the droplet number mixing ratio
379 (QNDROP), which was derived from the higher number of particle concentrations in the ACR
380 experiment. The CTL and ARI experiments did not consider the activation of aerosol-originated cloud
381 droplet number source; therefore, the QNDROP without aerosol-cloud interaction was significantly

382 lower than that in ACR experiment. However, previous studies such as by Zhang et al. (2015) and
383 Forkel et al. (2015) showed that aerosol-cloud interaction reduces QNDROP, because of almost
384 horizontally homogeneous particle numbers and the QNDROP calculated for unpolluted conditions.
385 The process described above yields different results for cloud-precipitation than those determined in
386 the present study. The discrepancy between previous studies and this research is attributed to the
387 different environment of aerosol characteristics. Our domain, which includes highly polluted areas,
388 represents distinctly different physical and chemical aerosol characteristics under abundant cloud
389 water content conditions. For example, Forkel et al. (2015) showed the result of indirect effects in
390 unpolluted marine or coastal area, while Zhang et al. (2015) focused on a low cloud cover region and
391 a season dominated by indirect effects. That is, a more adequate quantity of data on aerosol
392 characteristics is a prerequisite to diagnose the relationship between aerosol effects and cloud variables.
393 Furthermore, it can partly be explained by the different version of the aerosol-cloud feedback
394 mechanism of WRF-Chem. In the detailed WRF-Chem model (i.e., version 3.6), better performance
395 becomes possible for more realistic aerosol fields and aerosol-cloud interaction under the various and
396 different conditions of aerosol concentrations.

397 Last, we examined the manner in which the increased LWP affects precipitation in and around the
398 Korean Peninsula. Fig. 8 displays the spatial distributions of total, direct, and indirect effects on
399 precipitation for both cases. Here, total precipitation is indicated in Figs. 1a and 1c. In Fig. 8, the
400 changes of aerosol effects on precipitation occur predominantly in regions such as the southeastern
401 and central parts of Korean Peninsula for Case A and Case B, respectively, where the LWP is distinctly
402 increased by aerosol effects. The spatial distribution of changes in precipitation demonstrated
403 continuously iterated patterns of decrease and increase, which were also reported by Mashayekhi and
404 Sloan (2014).

405 Considering the overall spatial distribution of aerosol effects, a distinct difference is evident
406 between Case A and Case B. Direct and indirect effects equally contributed to the total effects on the
407 variation of precipitation for Case A. Whereas, in Case B, a more dominant contribution of indirect
408 effects than direct effects was evident. For example, for Case A, the direct effects on changes in
409 precipitation over South Korea, ranging from -54.6 to 24.1 mm, were comparable to those of indirect
410 effects, ranging from -23.8 to 24.0 mm, and almost equally contributed to the total effects with values
411 ranging from -63.2 to 27.1 mm (Figs. 8a–c). In contrast, the direct effects for Case B, with a minimum
412 value of -7.0 mm, had little influence on the changes in precipitation compared to the indirect effects,
413 with a minimum value of -36.6 mm (Figs. 8d–f). The characteristics of each case correspond to the
414 averaged Δ PR over South Korea, as shown in Figs. 7d and 7h. For Case A, the time series of Δ PR by
415 direct effects was more dominant, followed by total effects. For Case B, the phase of Δ PR derived
416 from indirect effects with relatively little direct effects.

417 To further explain and obtain better insight into the differences between the two cases, both
418 (=direct + indirect) effects are dominant, and only indirect effects are dominant on precipitation for the
419 conversion of cloud water to rain. We analyzed the longitude-height cross section of the difference in
420 the tendency for the conversion of cloud water to rain (Fig. 9). The differences in wind fields averaged
421 over specific 24-h periods for both cases are indicated in Fig. 9. For both cases, the precipitation system
422 propagated from the southwest (or west) toward the northeast, passing through the Korean Peninsula.
423 In Figs. 9a and 9e, over the upstream (West) area, total effects on the cloud water to rain conversion
424 showed a negative (–) sign for both cases, indicating that aerosol effects suppress precipitation.
425 However, a positive sign (+) for Case A was simulated over the downstream (East) area (Figs. 9a and
426 9e).

427 The variations in direct effects shown in Fig. 9b can be explained by vertical wind fields, which
428 vary according to meteorological variables such as upward motion due to radiation, temperature, and

429 stability/convection of the atmosphere in Case A. That is, radiative direct cooling over the near-surface
430 high aerosol concentration area induces a sinking motion (see wind vector perturbations in Fig. 9b)
431 due to the decreased surface temperature (Fig. 4c), enhancing stable stratification of the suppression
432 process of the cloud water into rain conversion. As illustrated in Fig. 4c, the $\Delta SW_{sfc}^{\downarrow}$ for direct effects
433 significantly influences on the reduction in SW_{sfc}^{\downarrow} , because of the total effects for Case A. The decrease
434 of radiation reaching the ground is reflected in the perturbation of temperature, which results in
435 variation of atmospheric stability and convection processes. Following Zhao et al. (2012), because
436 aerosol-induced warming or cooling accumulates over time, the temperature pattern does not
437 accurately match the anomalies of vertical motion. Hence, it should be note that the change of vertical
438 velocity is not directly comparable to that of temperature, although the wind field varies with the
439 temperature. In Case B, however, the radiative flux convergence was little simulated with no
440 significant changes of vertical motions over the study domain.

441 The indirect effects on change in precipitation with the alteration of cloud properties by acting as
442 the CCN were also investigated, as shown in Figs. 9c and 9g. The indirect effects suppressed
443 precipitation over the upstream areas for both cases, and the results correspond to the distributions of
444 cloud variables (Figs. S3b and S3d). This suggests that indirect effects over Northeast Asia can
445 suppress the PR and resultantly delay the onset of precipitation. Interesting here is the opposite
446 tendency over the downstream area for Case A (Fig. 9c), although no significant sign was found in
447 Case B (Fig. 9g). In Case A, the time-mean RH was predominantly high ($RH > 85\%$ at altitude below
448 1.5km), as shown in Fig. 9d, where abundant water vapor was favorable for saturation. Therefore,
449 vapor condensation invokes adiabatic warming, directly inducing an enhanced updraft motion, as
450 indicated in Figs. 9c and 9d. This process ultimately enhances the fluctuations of precipitation in these
451 conditions. In contrast, the RH in Case B is lower than 75%, except for near the surface (Fig. 9h),

452 where there was little saturation-warming thermodynamic processes and therefore a relatively smaller
453 (or no) increase in cloud to precipitation conversions over the downstream (East) area in Case B.

454 Obviously, clouds are highly nonlinear systems and cloud-radiation environments can yield
455 unpredictable results that create challenges in the interpretation of simulated and observed data.
456 Furthermore, the current analysis is not sufficient to completely describe aerosol effects, because the
457 aerosol activation process employed here cannot consider IN for the aerosol scheme (i.e.,
458 MADE/SORGAM) used in the current version of the WRF-Chem study. This indicates that the
459 tendency towards possible underestimation in quantifying the role of indirect effects should be
460 explored in the modeling study. To represent aerosol effects more accurately and quantitatively, it is
461 therefore necessary to perform further modeling and employ more comprehensive measurements for ice-
462 borne aerosols and IN treatment.

463

464 **4. Summary and conclusions**

465 The main objectives of this study were to characterize and diagnose the influences of aerosol
466 effects on mesoscale cloud-radiation variables over Northeast Asia during the MAPS-Seoul 2015
467 campaign period. We employed the fully coupled online WRF-Chem model and three different
468 experiments with and/or without aerosol effects were carried out for the investigation of aerosol-cloud-
469 radiation interaction effects.

470 The numerical results were first evaluated against available measurements obtained from MAPS-
471 Seoul 2015 campaign, and we clearly found out that a case with full aerosol with feedback effects
472 yielded better prediction capability than other two experiments. It is also noted that the aerosol effects
473 played a dominant role in altering radiative variables and cloud properties over downstream areas. The
474 largest difference among the radiative variables due to aerosol effects was in downward SW radiation.
475 The resultant perturbation in radiation properties was highly likely to be associated with the aerosol

476 loading profile, and in turn involved in the cloud response. This is suggesting that the interaction
477 between cloud and radiation was significant in the prediction of cloud cover/amount. In addition,
478 compared with the respective aerosol effects on cloud properties, the total effects are attributed to
479 indirect effects. Also, the variations of cloud properties and precipitation by total effects occurred at
480 almost the same time over South Korea. However, contribution fraction of respective aerosol effects
481 on precipitation varied according to meteorological conditions. We realized that the nonlinear cloud to
482 precipitation conversion process worked on the downstream area.

483 In addition, we pointed out that, under abundant cloud water and aerosol conditions, the direct
484 and indirect effects locally suppressed the precipitation and resultantly delayed the onset of
485 precipitation with the time lag of up to 9 hours over Northeast Asia. This implies the importance of
486 aerosol effects in improving mesoscale precipitation predictabilities in the high polluted area based on
487 the online approach considering aqueous phase chemistry. In particular, the change in onset time and
488 intensity of precipitation hours can have a severe and important impact on the regional daily
489 meteorological fields, particularly over the Korean Peninsula. The results through complex aerosol-
490 cloud-radiation interaction examined in this study over Northeast Asia can have profound and serious
491 implications to the regional cloud and precipitation, and therefore immediate follow-up studies would
492 be needed.

493 However, we also found out here some results with big discrepancies in comparison with the
494 previous results, particularly regarding the aerosol-cloud relationship with regards to the aerosol
495 indirect effects. Previous studies mostly showed the indirect effects were dominated only in unpolluted
496 marine/coastal areas with low-cloud cover. Therefore, as a future study, multiple sensitivity
497 experiments that apply more comprehensive aerosol treatments should be conducted against well-
498 designed field studies considering various cloud and precipitation conditions over Northeast Asia.

499 Those are to better understand the aerosol feedback mechanism in the online-coupled WRF-Chem
500 model.

501

502 **Acknowledgments**

503 We thank National Institute of Environmental Research (NIER) for providing the MAPS-Seoul 2015
504 campaign measurements, and also thank KMA for providing meteorological observation data and
505 computer resources. We also thank NOAA members for supporting to install WRF-Chem, and this
506 research was partially supported by National Research Foundation of Korea (NRF) funded by the
507 Ministry of Education (NRF-2015R1D1A1A01060088).

508

509 **Appendix A. Supplementary data**

510 Supplementary data related to this article can be found at <http://dx.doi.org/XXX>

511

512 **References**

- 513 Abdul-Razzak, H., Ghan, S. J., 2000. A parameterization of aerosol activation 2. Multiple aerosol types.
514 J. Geophys. Res. 105, 6837-6844, doi:10.1029/1999JD901161.
- 515 Abdul-Razzak, H., Ghan, S. J., 2002. A parameterization of aerosol activation 3. Sectional
516 representation. J. Geophys. Res. 107, 4026, doi:10.1029/2001JD000483.
- 517 Ackermann, I. J., Hass, H., Memmsheimer, M., Ebel, A., Binkowski, F. S., Shankar, U., 1998. Modal
518 aerosol dynamics model for Europe: development and first applications. Atmos. Environ. 32,
519 2981-2999.
- 520 Albrecht, B. A., 1989. Aerosols, cloud microphysics, and fractional cloudiness. Science 245, 1227-
521 1230.

522 Archer-Nicholls, S., Lowe, D., Schultz, D. M., McFiggans, G., 2016. Aerosol-radiation-cloud
523 interactions in a regional coupled model: the effects of convective parameterisation and
524 resolution. *Atmos. Chem. Phys.* 16, 5573-5594.

525 Barnard, J. C., Fast, J. D., Paredes-Miranda, G., Arnott, W. P., Laskin, A., 2010. Technical Note:
526 Evaluation of the WRF-Chem “Aerosol Chemical to Aerosol Optical Properties” Module using
527 data from the MILAGRO campaign. *Atmos. Chem. Phys.* 10, 7325-7340, doi:10.5194/acp-10-
528 7325-2010.

529 Boltzmann, L., 1884. Ableitung des Stefan'schen Gesetzes, betreffend die Abhängigkeit der
530 Wärmestrahlung von der Temperatur aus der electromagnetischen Lichttheorie. *Annalen der*
531 *Physik und Chemie (in German)* 258 (6), 291-294, doi:10.1002/andp.18842580616.

532 Brunner, D., Savage, N., Jorba, O., Eder, B., Giordano, L., Badia, A., Balzarini, A., Baró, R., Bianconi,
533 R., Chemel, C., Curci, G., Forkel, R., Jiménez-Guerrero, P., Hirtl, M., Hodzic, A., Honzak, L.,
534 Im, U., Knote, C., Makar, P., Manders-Groot, A., van Meijgaard, E., Neal, L., Pérez, J. L.,
535 Pirovano, G., San Jose, R., Schröder, W., Sokhi, R. S., Syrakov, D., Torian, A., Tuccella, P.,
536 Werhahn, J., Wolke, R., Yahya, K., Zabkar, R., Zhang, Y., Hogrefe, C., Galmarini, S., 2015.
537 Comparative analysis of meteorological performance of coupled chemistry-meteorology
538 models in the context of AQMEII phase 2. *Atmos. Environ.* 115, 470-498.

539 Canagaratna, M., Jayne, J., Jimenez, J., Allan, J., Alfarra, M., Zhang, Q., Onasch, T., Drewnick, F.,
540 Coe, H., Middlebrook, A., 2007. Chemical and microphysical characterization of ambient
541 aerosols with the aerodyne aerosol mass spectrometer. *Mass Spectrom. Rev.* 26, 185-222.

542 Carmichael, G.R., Calori, G., Hayami, H., Uno, I., Cho, S.Y., Engardt, M., Kim, S.B., Ichikawa, Y.,
543 Ikeda, Y., Woo, J.H., Ueda, H., Amann, M., 2002. The MICS-Asia study: model
544 intercomparison of long-range transport and sulfur deposition in East Asia. *Atmos. Environ.*
545 36, 175-199.

546 Chapman, E. G., Gustafson Jr., W. I., Easter, R. C., Barnard, J. C., Ghan, S. J., Pekour, M. S., Fast, J.
547 D., 2009. Coupling aerosol-cloud-radiative processes in the WRF-Chem model: Investigating
548 the radiative impact of elevated point sources. *Atmos. Chem. Phys.* 9, 945-964,
549 doi:10.5194/acp-9-945-2009.

550 Chen, F., Dudhia, J., 2001. Coupling an advanced land-surface/ hydrology model with the Penn State/
551 NCAR MM5 modeling system. Part I: Model description and implementation. *Mon. Wea. Rev.*
552 129, 569-585.

553 Cheng, W. Y. Y., Steenburgh, W. J., 2005. Evaluation of Surface Sensible Weather Forecasts by the
554 WRF and the Eta Models over the Western United States. *Wea. Forecast.* 20, 812-821.

555 DeCarlo, P.F., Kimmel, J.R., Trimborn, A., Northway, M.J., Jayne, J.T., Aiken, A.C., Gonin, M., Fuhrer,
556 K., Horvath, T., Docherty, K.S., 2006. Field-deployable, high-resolution, time-of-flight aerosol
557 mass spectrometer. *Anal. Chem.* 78, 8281-8289.

558 Drewnick, F., Hings, S.S., DeCarlo, P., Jayne, J.T., Gonin, M., Fuhrer, K., Weimer, S., Jimenez, J.L.,
559 Demerjian, K.L., Borrmann, S., 2005. A new time-of-flight aerosol mass spectrometer (TOF-
560 AMS) - Instrument description and first field deployment. *Aerosol Sci. Tech.* 39, 637-658.

561 Easter, R. C., Ghan, S. J., Zhang, Y., Saylor, R. D., Chapman, E. G., Laulainen, N. S., Abdul-Razzak,
562 H., Leung, L. R., Bian, X., Zaveri, R. A., 2004. MIRAGE: Model description and evaluation
563 of aerosols and trace gases. *J. Geophys. Res.* 109, D20210, doi:10.1029/2004JD004571.

564 Emmons, L. K., Walters, S., Hess, P. G., Lamarque, J.-F., Pfister, G. G., Fillmore, D., Granier, C.,
565 Guenther, A., Kinnison, D., Laepple, T., Orlando, J., Tie, X., Tyndall, G., Wiedinmyer, C.,
566 Baughcum, S. L., Kloster, S., 2010. Description and evaluation of the Model for Ozone and
567 Related chemical Tracers, version 4 (MOZART-4). *Geosci. Model Dev.* 3, 43-67,
568 doi:10.5194/gmd-3-43-2010.

569 Fan, J., Leung, L. R., Li, Z., Morrison, H., Chen H., Zhou, Y., Qian, Y., Wang, Y., 2012. Aerosol impacts
570 on clouds and precipitation in eastern China: Results from bin and bulk microphysics. *J.*
571 *Geophys. Res.* 117, D00K36, doi:10.1029/2011JD016537.

572 Fast, J. D., Gustafson Jr., W. I., Easter, R. C., Zaveri, R. A., Barnard, J. C., Chapman, E. G., Grell, G.
573 A., Peckham, S. E., 2006. Evolution of ozone, particulates, and aerosol direct radiative forcing
574 in the vicinity of Houston using a fully coupled meteorology-chemistry-aerosol model. *J.*
575 *Geophys. Res.* 111, D21305, doi:10.1029/2005JD006721.

576 Forkel, R., Balzarini, A., Baró, R., Bianconi, R., Curci, G., Jiménez-Guerrero, P., Hirtl, M., Honzak,
577 L., Lorenz, C., Im, U., Pérez, J. L., Pirovano, G., San José, R., Tuccella, P., Werhahn, J., Žabkar,
578 R., 2015. Analysis of the WRF-Chem contributions to AQMEII phase2 with respect to aerosol
579 radiative feedbacks on meteorology and pollutant distributions. *Atmos. Environ.* 115, 630-645.

580 Forkel, R., Werhahn, J., Hansen, A. B., McKeen, S., Peckham, S., Grell, G., Suppan, P., 2012. Effect
581 of aerosol-radiation feedback on regional air quality - A case study with WRF/Chem. *Atmos.*
582 *Environ.* 53, 202-211.

583 Gao, M., Liu, Z., Wang, Y., Lu, X., Ji, D., Wang, L., Li, M., Wang, Z., Zhang, Q., Carmichael, G.R.,
584 2017. Distinguishing the roles of meteorology, emission control measures, regional transport,
585 and co-benefits of reduced aerosol feedbacks in “APEC Blue”. *Atmos. Environ.* doi:
586 10.1016/j.atmosenv.2017.08.054.

587 Gao, Y., Zhang, M., Liu, Z., Wang., L., Wang, P., Xia, X., Tao, M., Zhu, L., 2015. Modeling the
588 feedback between aerosol and meteorological variables in the atmospheric boundary layer
589 during a severe fog-haze event over the North China Plain. *Atmos. Chem. Phys.* 15, 4279-4295.

590 Ghan, S., Laulainen, N., Easter, R., Wagener, R., Nemesure, S., Chapman, E., Zhang, Y., Leung, R.,
591 2001. Evaluation of aerosol direct radiative forcing in MIRAGE. *J. Geophys. Res.* 106, 5295-
592 5316.

593 Grell, G. A., Devenyi, D., 2002. A generalized approach to parameterizing convection combining
594 ensemble and data assimilation techniques. *Geophys. Res. Lett.* 29(14), Article 1693.

595 Grell, G. A., Peckham, S. E., Schmitz, R., McKeen, S. A., Frost, G., Skamarock, W. C., Eder, B., 2005.
596 Fully coupled “online” chemistry within the WRF model. *Atmos. Environ.* 39, 6957-6975.

597 Grosvenor, D. P., Field, P. R., Hill, A. A., Shipway, B. J., 2017. The relative importance of
598 macrophysical and cloud albedo changes for aerosol-induced radiative effects in closed-cell
599 stratocumulus: insight from the modelling of a case study. *Atmos. Chem. Phys.* 17, 5155-5183,
600 doi:10.5194/acp-17-5155-2017.

601 Guenther, A., Karl, T., Harley, P., Wiedinmyer, C., Palmer, P. I., Geron, C., 2006. Estimates of global
602 terrestrial isoprene emissions using MEGAN (Model of Emissions of Gases and Aerosols from
603 Nature). *Atmos. Chem. Phys.* 6, 3181-3210, doi:10.5194/acp-6-3181-2006.

604 Gustafson Jr., W.I., Chapman, E.G., Ghan, S.J., Easter, R. C., Fast, J. D., 2007. Impact on modeled
605 cloud characteristics due to simplified treatment of uniform cloud condensation nuclei during
606 NEAQS 2004. *Geophys. Res. Lett.* 34, L19809, doi:10.1029/2007GL030021.

607 Harris, E., Sinha, B., van Pinxteren, D., Tilgner, A., Fomba, K. W., Schneider, J., Roth, A., Gnauk, T.,
608 Fahlbusch, B., Mertes, S., Lee, T., Collett, J., Foley, S., Borrmann, S., Hoppe, P., Herrmann,
609 H., 2013. Enhanced role of transition metal ion catalysis during in-cloud oxidation of SO₂.
610 *Science* 340, 727-730.

611 Haywood, J., Boucher, O., 2000. Estimates of the direct and indirect radiative forcing due to
612 tropospheric aerosols: A review. *Rev. Geophys.* 38, 513-543.

613 Hong, C., Zhang, Q., Zhang, Y., Tang, Y., Tong, D., He, K., 2017. Multi-year downscaling application
614 of two-way coupled WRF v3.4 and CMAQ v5.0.2 over east Asia for regional climate and air
615 quality modeling: model evaluation and aerosol direct effects. *Geosci. Model Dev.* 10, 2447-
616 2470.

617 Hong, S.-Y., Noh, Y., Dudhia, J., 2006. A new vertical diffusion package with an explicit treatment of
618 entrainment processes. *Mon. Wea. Rev.* 134, 2318-2341.

619 Huffman, G.J., Adler, R. F., Bolvin, D.T., Gu, G., Nelkin, E. J., Bowman, K. P., Hong, Y., Stocker, E.
620 F., Wolff, D. B., 2007. The TRMM Multisatellite Precipitation Analysis (TMPA): Quasi-Global,
621 Multiyear, Combined-Sensor Precipitation Estimates at Fine Scales. *J. Hydrometeorol.* 8(1),
622 38-55.

623 Iacono, M. J., Delamere, J. S., Mlawer, E. J., Shephard, M. W., Clough, S. A., Collins, W. D., 2008.
624 Radiative forcing by long-lived greenhouse gases: calculations with the AER radiative transfer
625 models. *J. Geophys. Res.* 113, D13103.

626 Jayne, J.T., Leard, D.C., Zhang, X., Davidovits, P., Smith, K.A., Kolb, C.E., Worsnop, D.R., 2000.
627 Development of an aerosol mass spectrometer for size and composition analysis of submicron
628 particles. *Aerosol Sci. Tech.* 33, 49-70.

629 Jiang, F., Liu, Q., Huang, X., Wang, T., Zhuang, B., Xie, M., 2012. Regional modeling of secondary
630 organic aerosol over China using WRF/Chem. *J. Aerosol Science.* 43, 57-73.

631 Jimenez, J. L., Jayne, J. T., Shi, Q., Kolb, C. E., Worsnop, D. R., Yourshaw, I., Seinfeld, J. H., Flagan,
632 R. C., Zhang, X., Smith, K. A., Morris, J. W., Davidovits, P., 2003. Ambient aerosol sampling
633 using the Aerodyne Aerosol Mass Spectrometer. *J. Geophys. Res.* 108(D7), doi:
634 10.1029/2001JD001213.

635 Kim, B.-G., Schwartz, S.E., Miller, M.A., Min, Q., 2003. Effective radius of cloud droplets by ground-
636 based remote sensing: Relationship to aerosol. *J. Geophys. Res.* 108(D23), doi:
637 10.1029/2003JD003721.

638 Kim, C.-H., Park, S.-Y., Kim, Y.-J., Chang, L.-S., Song, S.-K., Moon, Y.-S., Song, C.-K., 2012. A
639 numerical study on indicators of long-range transport potential for anthropogenic particulate
640 matters over northeast Asia. *Atmos. Environ.* 58, 35-44.

641 Kim, D.-Y., Kim, J.-Y., Kim, J.-J., 2013. Mesoscale simulations of multi-decadal variability in the
642 wind resource over Korea. *Asia- Pacific J. Atmos. Sci.* 49, 182-192.

643 Kim, B.-U., Bae, C., Kim, H. C., Kim, E., Kim, S., 2017a. Spatially and chemically resolved source
644 apportionment analysis: Case study of high particulate matter event. *Atmos. Environ.* 162, 55-
645 70.

646 Kim, N., Park, M., Yum, S. S., Park, J. S., Song, I. H., Shin H. J., Ahn, J. Y., Kwak K.-H., Kim, H.,
647 Bae, G.-N., Lee, G., 2017b. Hygroscopic properties of urban aerosols and their cloud
648 condensation nuclei activities measured in Seoul during the MAPS-Seoul campaign. *Atmos.*
649 *Environ.* 153, 217-232.

650 Kong, X., Forkel, R., Sokhi, R. S., Suppan, P., Baklanov, A., Gauss, M., Brunner, D., Barò, R.,
651 Balzarini, A., Chemel, C., Curci, G., Jiménez-Guerrero, P., Hirtl, M., Honzak, L., Im, U., Pérez,
652 J. L., Pirovano, G., San Jose, R., Schlünzen, K. H., Tsegas, G., Tuccella, P., Werhahn, J., Žabkar,
653 R., Galmarini, S., 2015. Analysis of meteorology-chemistry interactions during air pollution
654 episodes using online coupled models within AQMEII phase-2. *Atmos. Environ.* 115, 527-540.

655 Kourtidis, K., Stathopoulos, S., Georgoulas, A. K., Alexandri, G., Rapsomanikis, S., 2015. A study of
656 the impact of synoptic weather conditions and water vapor on aerosol-cloud relationships over
657 major urban clusters of China. *Atmos. Chem. Phys.* 15, 10955-10964.

658 Lee, H.-J., Kim, C.-H., 2016. Transport/transformations study during the MAPS (pre-KORUS-AQ)
659 field campaign. 17th Annual WRF Users' Workshop. NCAR, Boulder, CO, USA.

660 Lee, T., Choi, J., Lee, G., Ahn, J., Park, J.S., Atwood, S.A., Schurman, M., Choi, Y., Chung, Y., Collett
661 Jr, J.L., 2015. Characterization of aerosol composition, concentrations, and sources at
662 Baengnyeong Island, Korea using an aerosol mass spectrometer. *Atmos. Environ.* 120, 297-
663 306.

664 Li, M., Zhang, Q., Streets, D. G., He, K. B., Cheng, Y. F., Emmons, L. K., Huo, H., Kang, S. C., Lu,
665 Z., Shao, M., Su, H., Yu, X., Zhang, Y., 2014. Mapping Asian anthropogenic emissions of non-
666 methane volatile organic compounds to multiple chemical mechanisms. *Atmos. Chem. Phys.*
667 14, 5617–5638.

668 Li, M., Zhang, Q., Kurokawa, J., Woo, J.-H., He, K., Lu, Z., Ohara, T., Song, Y., Streets, D. G.,
669 Carmichael, G. R., Cheng, Y., Hong, C., Huo, H., Jiang, X., Kang, S., Liu, F., Su, H., Zheng,
670 B., 2017. MIX: a mosaic Asian anthropogenic emission inventory under the international
671 collaboration framework of the MICS-Asia and HTAP. *Atmos. Chem. Phys.* 17, 935–963.

672 Liu, Y., Daum, P. H., McGraw, R. L., 2005. Size truncation effect, threshold behavior, and a new type
673 of autoconversion parameterization. *Geophys. Res. Lett.* 32, L11811,
674 doi:10.1029/2005GL022636.

675 Madronich, S., 1987. Photodissociation in the atmosphere: 1. actinic flux and the effects of ground
676 reflections and clouds. *J. Geophys. Res.* 92, 9740-9752.

677 Mashayekhi, R., Sloan, J. J., 2014. Effects of aerosols on precipitation in north-eastern North America.
678 *Atmos. Chem. Phys.* 14, 5111-5125.

679 Matsui, H., Koike, M., Kondo, Y., Takegawa, N., Kita, K., Miyazaki, Y., Hu, M., Chang, S. Y., Blake,
680 D. R., Fast, J. D., Zaveri, R. A., Streets, D. G., Zhang, Q., Zhu, T., 2009. Spatial and temporal
681 variations of aerosols around Beijing in summer 2006: Model evaluation and source
682 apportionment. *J. Geophys. Res.* 114, D00G13, doi:10.1029/2008jd010906.

683 Mlawer, E. J., Taubman, S. J., Brown, P. D., Iacono, M. J., Clough, S. A., 1997. Radiative transfer for
684 inhomogeneous atmosphere: RRTM, a validated correlated-k model for the longwave. *J.*
685 *Geophys. Res.* 102 (D14), 16663-16682.

686 Molders, N., Tran, H. N. Q., Cahill, C. F., Leelasakultum, K., Tran, T. T., 2012. Assessment of
687 WRF/Chem PM2.5 forecasts using mobile and fixed location data from the Fairbanks, Alaska
688 winter 2008/09 field campaign. *Atmos. Pollut. Res.* 3, 180-191.

689 Morrison, H., Thompson, G., Tatarskii, V., 2009. Impact of Cloud Microphysics on the Development
690 of Trailing Stratiform Precipitation in a Simulated Squall Line: Comparison of One- and Two-
691 Moment Schemes. *Mon. Wea. Rev.* 137, 991-1007.

692 NCEP, 2000. NCEP FNL Operational Model Global Tropospheric Analyses, Continuing from July
693 1999. <http://dx.doi.org/10.5065/D6M043C6>

694 Ohara, T., Akimoto, H., Kurokawa, J., Horii, N., Yamaji, K., Yan, X., Hayasaka, T., 2007. *Atmos. Chem.*
695 *Phys.* 7, 4419–4444.

696 Park, I.-S., Choi, W.-J., Lee, T.-Y., Lee, S.-J., Han, J.-S., Kim, C.-H., 2005. Simulation of long-range
697 transport of air pollutants over Northeast Asia using a comprehensive acid deposition model.
698 *Atmos. Environ.* 39, 4075-4085.

699 Peckham, S., Grell, G. A., McKeen, S. A., Barth, M., Pfister, G., Wiedinmyer, C., Fast, J. D., Gustafson,
700 W. I., Zaveri, R., Easter, R. C., Barnard, J., Chapman, E., Hewson, M., Schmitz, R., Salzman,
701 M., Freitas, S., 2011. WRF/Chem Version 3.3 User's Guide. NOAA Technical Memo. 98 pp.

702 Piccot, S. D., Watson, J. J., Jones, J. W., 1992. Global inventory of volatile organic compound
703 emissions from anthropogenic sources. *J. Geophys. Res.* 97, 9897-9912.

704 Pincus, R., Barker, H. W., Morcrette, J.-J., 2003. A fast, flexible, approximate technique for computing
705 radiative transfer in inhomogeneous cloud fields. *J. Geophys. Res.* 108,
706 doi:10.1029/2002JD003322.

707 Pincus, R., Baker, M. B., 1994. Effect of precipitation on the albedo susceptibility of clouds in the
708 marine boundary layer. *Nature* 372, 250-252, doi:10.1038/372250a0.

709 Rieger, D., Bangert, M., Kottmeier, C., Vogel, H., Vogel, B., 2014. Impact of aerosol on post-frontal
710 convective clouds over Germany. *Tellus B* 66, 22528,
711 <http://dx.doi.org/10.3402/tellusb.v66.22528>.

712 Rosenfeld, D., 2000. Suppression of Rain and Snow by Urban and Industrial Air Pollution. *Science*
713 287 (5459), 1793-1796.

714 Rosenfeld, D., Lohmann, U., Raga, G. B., O'Dowd, C. D., Kulmala, M., Fuzzi, S., Reissell, A.,
715 Andreae, M. O., 2008. Flood or drought: how do aerosols affect precipitation? *Science* 321,
716 1309-1313.

717 Rosenfeld, D., Sherwood, S., Wood, R., Donner, L., 2014. Climate effects of aerosol-cloud interactions.
718 *Science* 343, 370-380.

719 Saide, P. E., Spak, S. N., Carmichael, G. R., Mena-Carrasco, M. A., Yang, Q., Howell, S., Leon, D. C.,
720 Snider, J. R., Bandy, A. R., Collett, J. L., Benedict, K. B., de Szoeke, S. P., Hawkins, L. N.,
721 Allen, G., Crawford, I., Crosier, J., Springston, S. R., 2012. Evaluating WRF-Chem aerosol
722 indirect effects in Southeast Pacific marine stratocumulus during VOCALS-REx. *Atmos. Chem.*
723 *Phys.* 12, 3045-3064, doi:10.5194/acp-12-3045-2012.

724 Saikawa, E., Naik, V., Horowitz, L. W., Liu, J. F., Mauzerall, D.L., 2009. Present and potential future
725 contributions of sulfate, black and organic carbon aerosols from China to global air quality,
726 premature mortality and radiative forcing. *Atmos. Environ.* 43, 2814-2822.

727 Schell, B., Ackermann, I. J., Hass, H., Binkowski, F. S., Ebel A., 2001. Modeling the formation of
728 secondary organic aerosol within a comprehensive air quality model system. *J. Geophys. Res.*
729 106, 28275-28293, doi:10.1029/2001JD000384.

730 Skamarock, W. C., Klemp, J. B., Dudhia, J., Gill, D. O., Barker, D. M., Duda, M., Huang, X.-Y., Wang,
731 W., Powers, J. G., 2008. A description of the Advanced Research WRF version 3. Technical
732 Note 475+STR, NCAR, Boulder, CO, USA., 113pp.

733 Song, H., Wang, K., Zhang, Y., Hong, C., Zhou, S., 2017. Simulation and evaluation of dust emissions
734 with WRF-Chem (v3.7.1) and its relationship to the changing climate over East Asia from 1980
735 to 2015. *Atmos. Environ.* doi: 10.1016/j.atmosenv.2017.08.051.

736 Stefan, J., 1879. Über die Beziehung zwischen der Wärmestrahlung und der Temperatur.
737 Sitzungsberichte der mathematisch-naturwissenschaftlichen Classe der kaiserlichen Akademie
738 der Wissenschaften (in German) Vienna, 79, 391-428.

739 Stockwell, W. R., Kirchner, F., Kuhn, M., Seefeld, S., 1997. A new mechanism for regional
740 atmospheric chemistry modeling. *J. Geophys. Res.* 102, 25847-25879.

741 Tao, Z., Yu, H., Chin, M., 2015. The role of aerosol-cloud-radiation interactions in regional air
742 quality—A NU-WRF Study over the United States. *Atmosphere* 6, 1045-1068,
743 doi:10.3390/atmos6081045.

744 Tuccella, P., Curci, G., Visconti, G., Bessagnet, B., Menut, L., Park, R. J., 2012. Modeling of gas and
745 aerosol with WRF/Chem over Europe: Evaluation and sensitivity study. *J. Geophys. Res.* 117,
746 D03303, doi:10.1029/2011jd016302.

747 Twomey, S., 1977. The influence of pollution on the shortwave albedo of clouds. *J. Atmos. Sci.* 34,
748 1149-1152.

749 Wang, F., Guo, J., Wu, Y., Zhang, X., Deng, M., Li, X., Zhang, J., Zhao, J., 2014. Satellite observed
750 aerosol-induced variability in warm cloud properties under different meteorological conditions
751 over eastern China. *Atmos. Environ.* 84, 122-132.

752 Wang, F., Guo, J., Zhang, J., Huang, J., Min, M., Chen, T., Liu, H., Deng, M., Li, X., 2015a. Multi-
753 sensor quantification of aerosol-induced variability in warm clouds over eastern China. *Atmos.*
754 *Environ.* 113, 1-9.

755 Wang, J., Xu, J., He, Y., Chen, Y., Meng, F., 2016. Long range transport of nitrate in the low atmosphere
756 over Northeast Asia. *Atmos. Environ.* 144, 315-324.

757 Wang, K., Zhang, Y., Yahya, K., Wu, S.-Y., Grell, G., 2015b. Implementation and initial application of
758 new chemistry-aerosol options in WRF/Chem for simulating secondary organic aerosols and
759 aerosol indirect effects for regional air quality. *Atmos. Environ.* 115, 716-732.

760 Woo, J.-H., Quan, S., Choi, K.-C., Kim, H., Jin, H., Song, C.-K., Han, J., Lee, S., 2014. Development
761 of the Comprehensive Regional Emissions inventory for Atmospheric Transport Experiment
762 (CREATE) inventory in support of integrated modeling of climate and air quality for East Asia.
763 16th GEIA Conference. NCAR, Boulder, CO, USA.

764 Wu, J., Li, G., Cao, J., Bei, N., Wang, Y., Feng, T., Huang, R., Liu, S., Zhang, Q., Tie, X., 2017.
765 Contributions of trans-boundary transport to summertime air quality in Beijing, China. *Atmos.*
766 *Chem. Phys.* 17, 2035-2051, doi:10.5194/acp-17-2035-2017

767 Yahya, K., Wang, K., Gudoshava, M., Glotfelty, T., Zhang, Y., 2015. Application of WRF/Chem over
768 North America under the AQMEII Phase 2: Part I. Comprehensive evaluation of 2006
769 simulation. *Atmos. Environ.* 115, 733-755.

770 Yang, Q., Gustafson Jr., W. I., Fast, J. D., Wang, H., Easter, R. C., Morrison, H., Lee, Y.-N., Chapman,
771 E. G., Spak, S. N., Mena-Carrasco, M. A., 2011. Assessing regional scale predictions of
772 aerosols, marine stratocumulus, and their interactions during VOCALS-REx using WRF-Chem.
773 *Atmos. Chem. Phys.* 11, 11951-11975, doi:10.5194/acp-11-11951-2011.

774 Zhang, B., Wang, Y., Hao, J., 2015. Simulating aerosol-radiation-cloud feedbacks on meteorology and
775 air quality over eastern China under severe haze conditions in winter. *Atmos. Chem. Phys.* 15,
776 2387-2404.

777 Zhang, D., Anthes, R. A., 1982. A High-Resolution Model of the Planetary Boundary Layer-Sensitivity
778 Tests and Comparisons with SESAME-79 Data. *J. Appl. Meteorol.* 21, 1594-1609.

779 Zhang, Y., Wen, X.-Y., Jang, C. 2010. Simulating chemistry-aerosol-cloud-radiation-climate feedbacks
780 over the continental US using the online-coupled Weather Research Forecasting Model with
781 chemistry (WRF/Chem). *Atmos. Environ.* 44, 3568-3582.

782 Zhou, C., Zhang, X., Gong, S., Wang, Y., Xue, M., 2016. Improving aerosol interaction with clouds
783 and precipitation in a regional chemical weather modeling system. *Atmos. Chem. Phys.* 16,
784 145-160.

785

786

787

788

789

790

791

792

793

794

795

796

797

798

799

800

801

802

803

List of Tables

804 Table 1. Statistics of the comparisons between simulated and observed temperature ($^{\circ}\text{C}$), relative
805 humidity (RH, %), and wind speed (m s^{-1}) for Case A (00 UTC 6 June to 23 UTC 9 June) and
806 Case B (00 UTC 11 June to 23 UTC 14 June), respectively.

807

808

809

810 Table 1

	Case A (00UTC 6 June to 23UTC 9 June)			Case B (00UTC 11 June to 23UTC 14 June)		
	Temp.	RH	WS	Temp.	RH	WS
	(°C)	(%)	(m s ⁻¹)	(°C)	(%)	(m s ⁻¹)
MEAN OBS ^a	20.41	69.46	1.77	22.08	73.64	2.20
MEAN MOD ^a	19.46	79.93	3.75	20.70	82.59	4.34
Corr. R ^b	0.93	0.86	0.70	0.94	0.93	0.77
MB ^c	-0.95	10.47	1.98	-1.38	8.95	2.14
NMB ^d	-4.65%	15.07%	112.28%	-6.26%	12.15%	97.26%
NME ^d	6.52%	15.76%	112.28%	6.58%	12.15%	97.26%
FB ^e	-4.65%	14.65%	74.67%	-6.78%	11.55%	67.69%
FE ^e	6.90%	15.28%	74.67%	7.09%	11.55%	67.69%
RMSE ^f	1.55	13.09	2.09	1.73	10.51	2.24
IOA ^g	0.94	0.82	0.45	0.91	0.88	0.45

811 ^a MEAN OBS and MEAN MOD are hourly mean values of observation and model results, respectively; ^b Corr. R the correlation coefficient
812 between the observation and model results; ^c MB the mean biases; ^d NMB and NME the normalized mean bias and error; ^e FB and FE the
813 fractional bias and error; ^f RMSE the root-mean-square error; ^g IOA the index of agreement.

814

Figure Captions

815

816 Fig. 1 Spatial distributions of (a) total precipitation (color, in mm day^{-1}) and temporally averaged wind
817 vectors (arrows, in m s^{-1}) at 850 hPa and (b) temporally averaged column aerosol optical depth
818 (AOD) at 550 nm (color) and liquid water path (LWP) at 500 hPa (white contours, in g m^{-2}
819 above 0.1 g m^{-2}) from the aerosol-cloud-radiation interaction (ACR) experiment for Case A (12
820 UTC on 7 June 2015 to 11 UTC on 8 June 2015). Panels (c) and (d) are the same as (a) and (b),
821 but for Case B (00 UTC on 11 June 2015 to 23 UTC on 11 June 2015).

822 Fig. 2 Time series of observed (gray bar) and simulated hourly precipitation rate (mm hr^{-1}) at (a) Gosan
823 and (b) Jinju for Case A and (c) Incheon for Case B. The black circles represent the aerosol-
824 cloud-radiation interaction (ACR) experiments, red triangles indicate the aerosol-radiation
825 interaction (ARI) experiments, and blue diamonds show the control (CTL) experiments.

826 Fig. 3 Spatial distributions of precipitation rate (mm hr^{-1}) from (a) Tropical Rainfall Measuring Mission
827 (TRMM) data, (b) the aerosol-cloud-radiation interaction (ACR) experiment, and (c) the
828 control (CTL) experiment at 21 UTC on 7 June 2015 for Case A. Panels (d)–(f) are the same
829 as (a)–(c), respectively, but at 21 UTC on 11 June 2015 for Case B.

830 Fig. 4 Time series of change in downward shortwave (SW) radiation averaged at the ground level over
831 South Korea for (a) Case A and (b) Case B in clear-sky conditions. Panels (c) and (d) are the
832 same as (a) and (b), respectively, but for all-sky conditions.

833 Fig. 5 Simulated vertical profiles of extinction coefficient at 550 nm from the aerosol-cloud-radiation
834 interaction (ACR) experiment averaged over South Korea for (a) Case A and (b) Case B.

835 Fig. 6 Time series of change in downward longwave (LW) radiation averaged at the ground level over
836 South Korea for (a) Case A and (b) Case B in all-sky conditions. Panels (c) and (d) show
837 simulated vertical profiles of total effects on the cloud water mixing ratio (Q_CLOUD) averaged
838 over South Korea for Case A and Case B, respectively.

839 Fig. 7 Time series of cloud fraction (CF) and precipitation rate (PR) averaged over the South Korea.

840 Panel (a) CF for three experiments; panel (b) difference of CF in aerosol effects of total, direct,
841 and indirect effects; panel (c) PR simulated from three experiments, (d) difference of PR from
842 aerosol effects for Case A. Panels (e)–(h) are the same as (a)–(d), respectively, but for Case B.

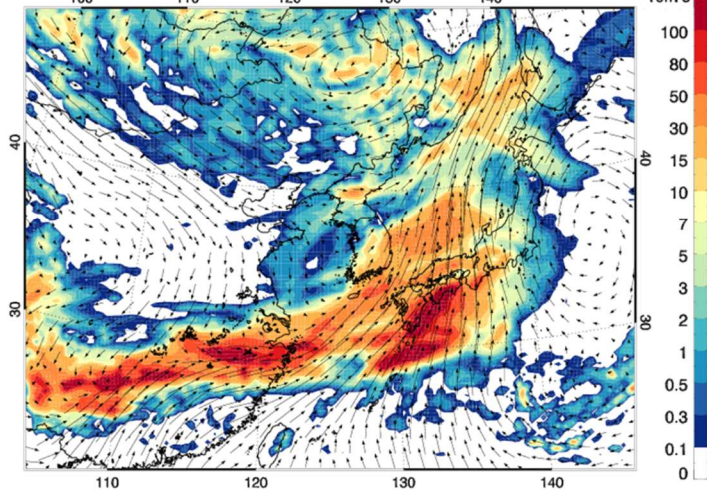
843 Fig. 8 Spatial distributions of temporally averaged (a) total effects, (b) direct effects, and (c) indirect
844 effects on total precipitation for Case A. Panels (d)–(f) are the same as (a)–(c), respectively, but
845 for Case B.

846 Fig. 9 Longitude-height cross section of (a) total effects, (b) direct effects, and (c) indirect effects on
847 tendency for conversion of cloud water to rain (color, in $\mu\text{g kg}^{-1} \text{s}^{-1}$) and wind fields (arrow, in
848 m s^{-1}), and (d) relative humidity (RH) for the aerosol-cloud-radiation interaction (ACR)
849 experiment for Case A. Panels (e)–(h) are the same as (a)–(d), respectively, but for Case B.
850 Note that the vertical wind speed was multiplied by a factor of 100.

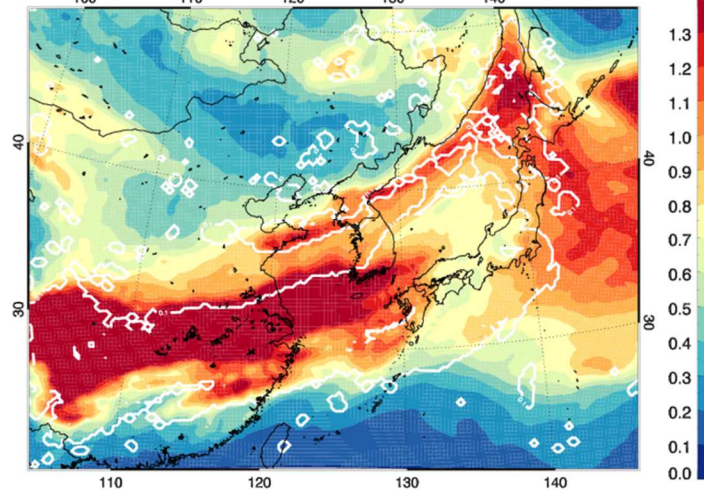
851

852

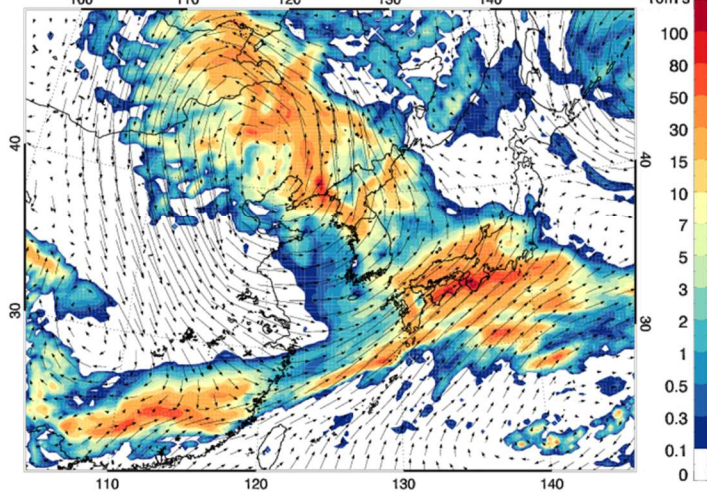
(a) Total precip. and Wind vectors [2015.06.07 12UTC - 2015.06.08 11UTC] unit : [mm] -10m s



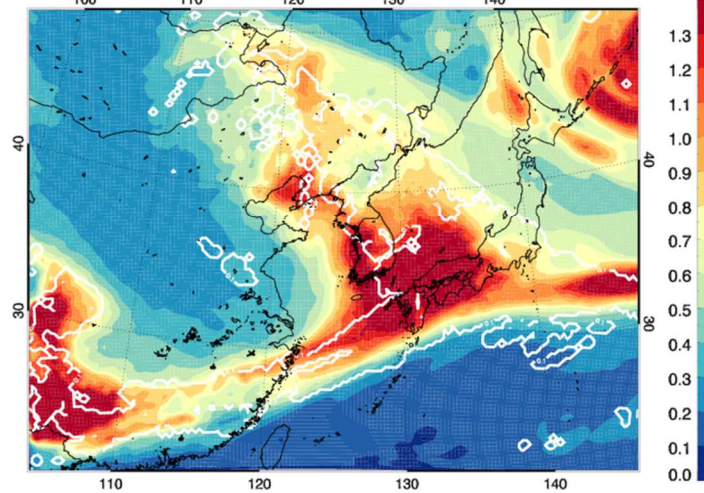
(b) 550nm AOD and LWP at 500hPa [2015.06.07 12UTC - 2015.06.08 11UTC]

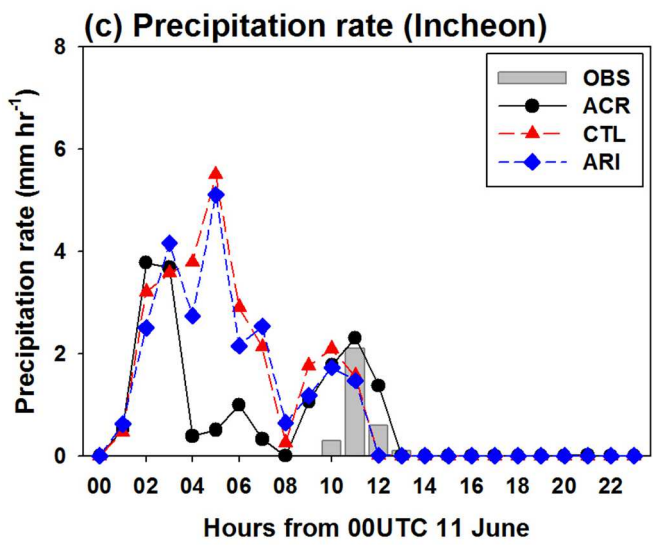
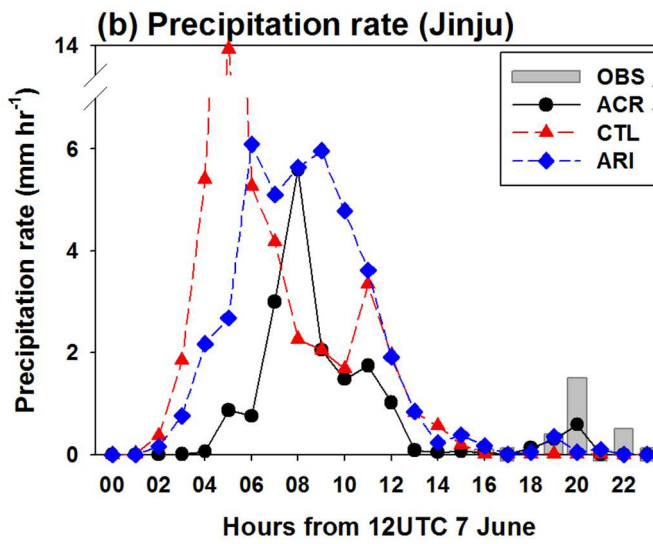
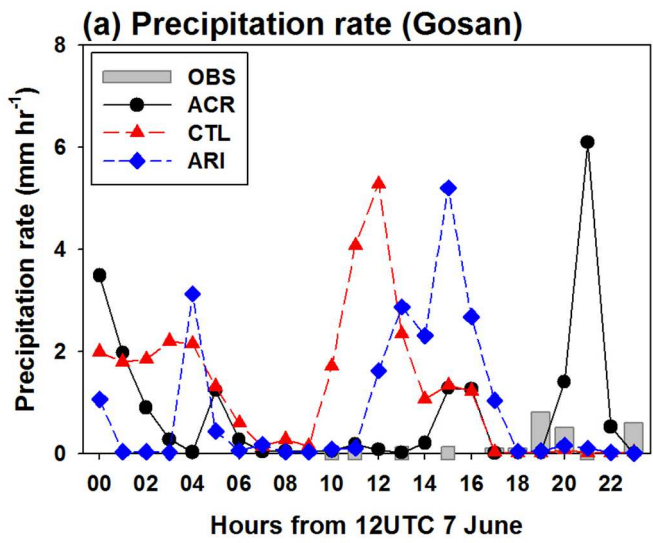


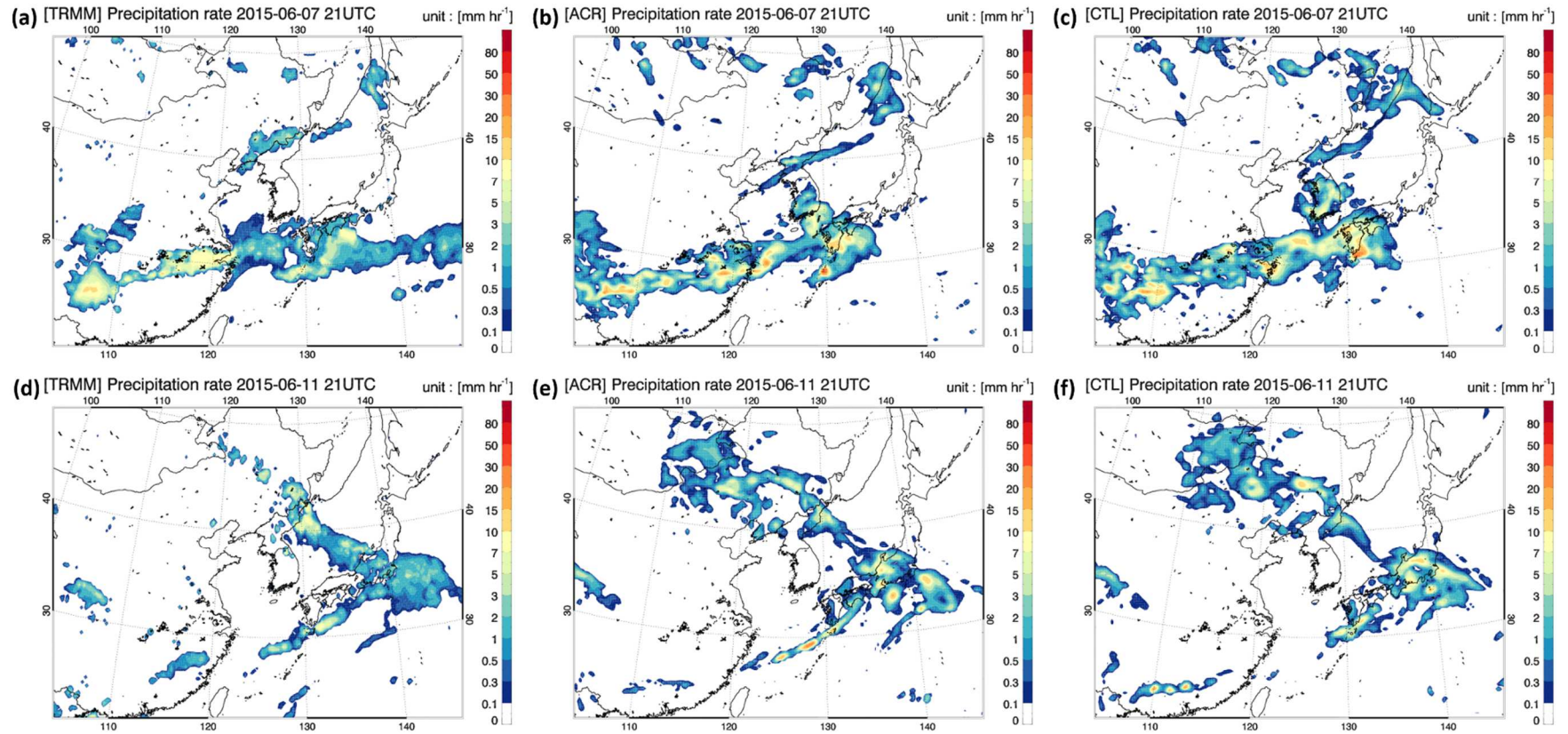
(c) Total precip. and Wind Vectors [2015.06.11 00UTC - 2015.06.11 23UTC] unit : [mm] -10m s



(d) 550nm AOD and LWP at 500hPa [2015.06.11 00UTC - 2015.06.11 23UTC]

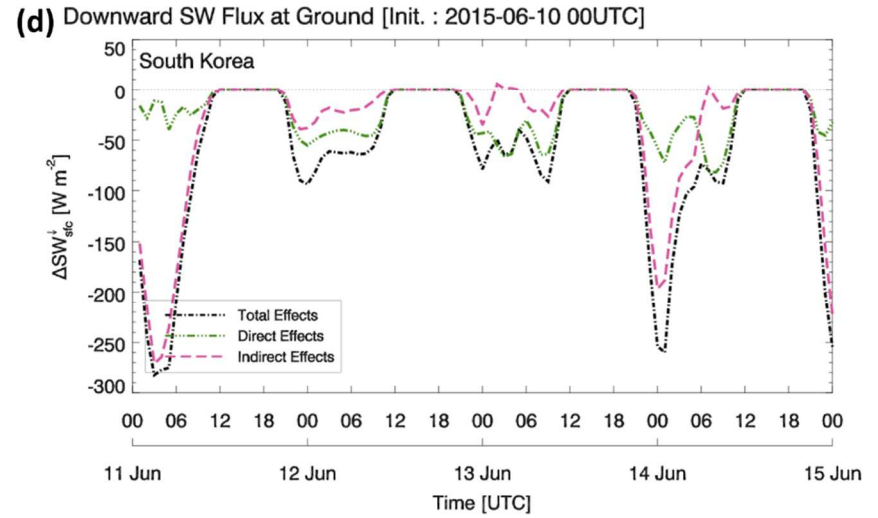
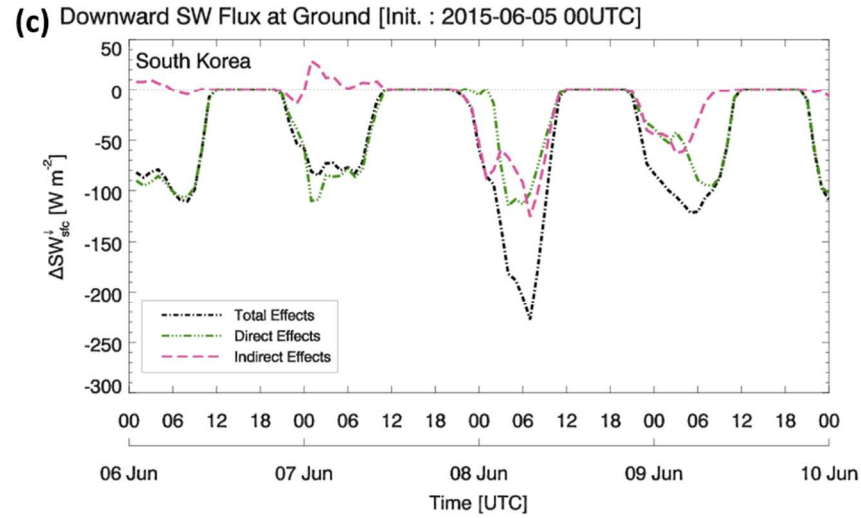
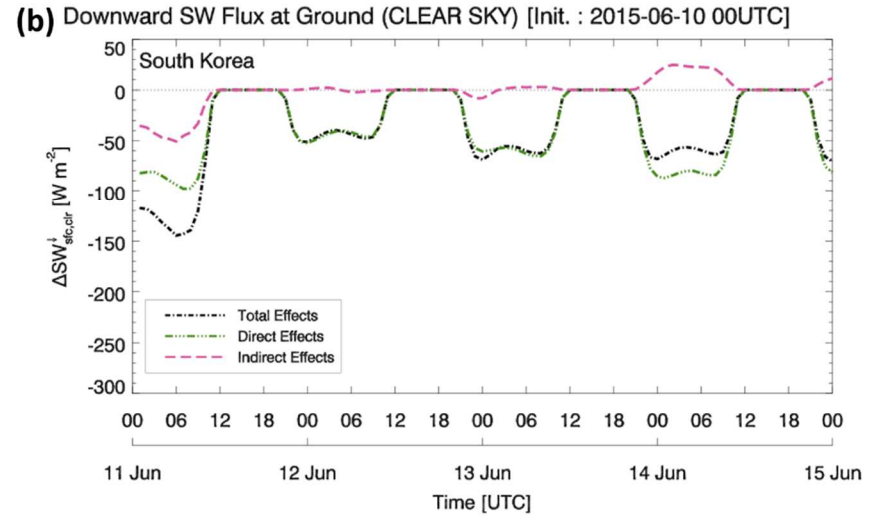
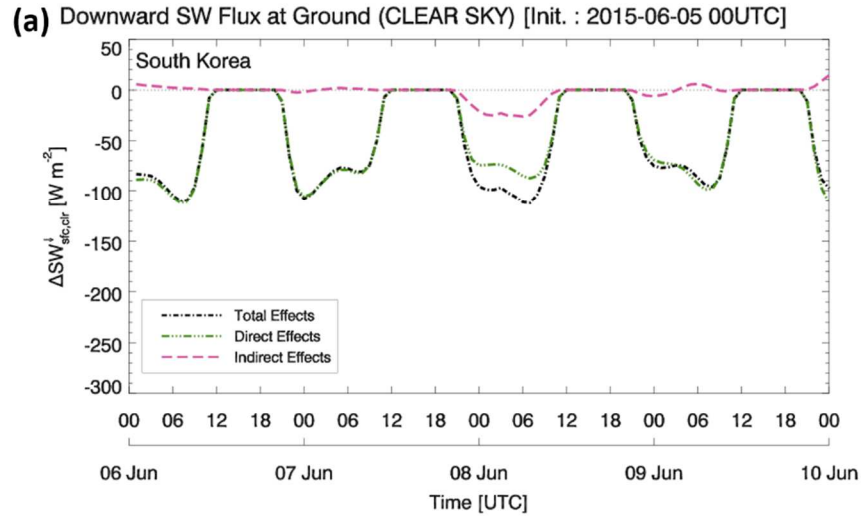




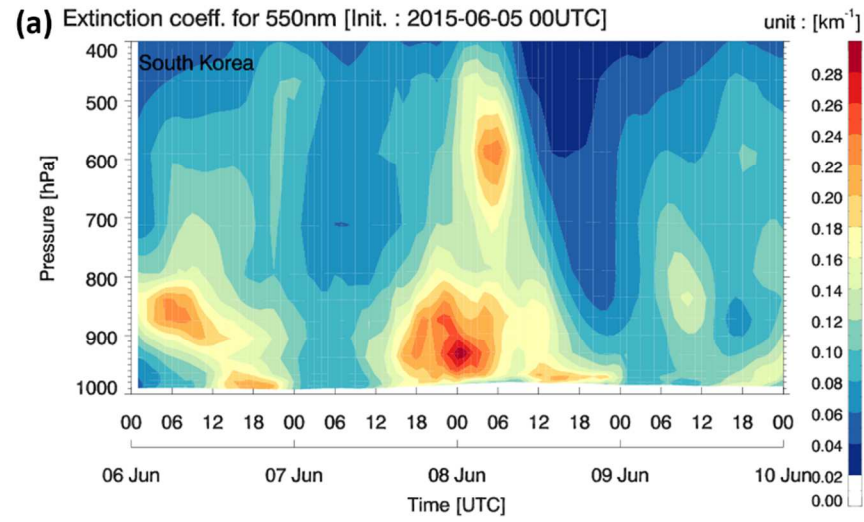


858

859

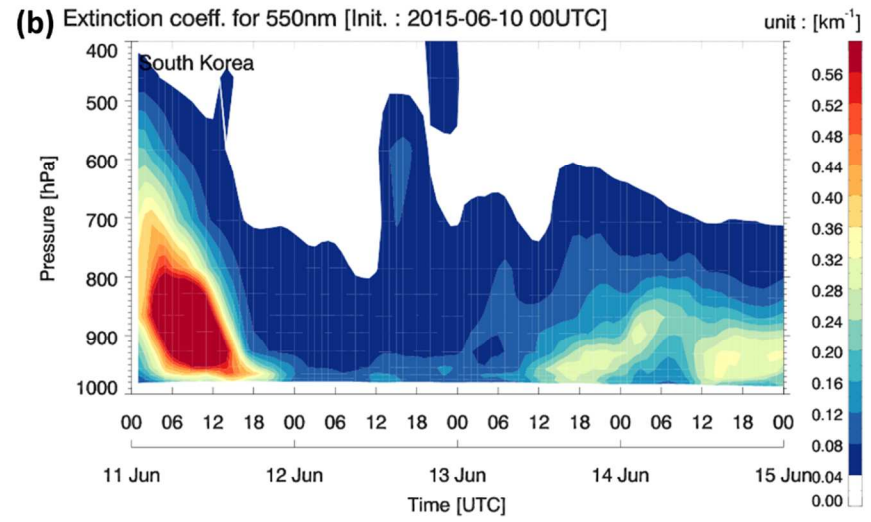


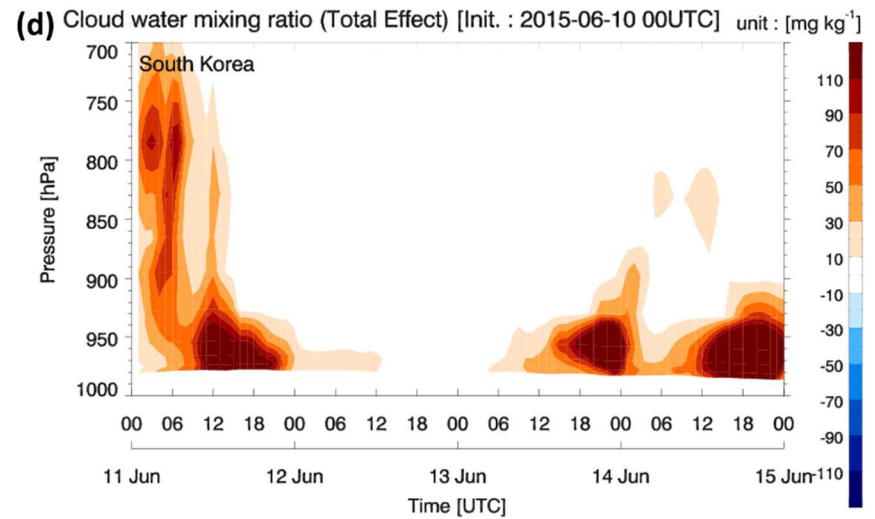
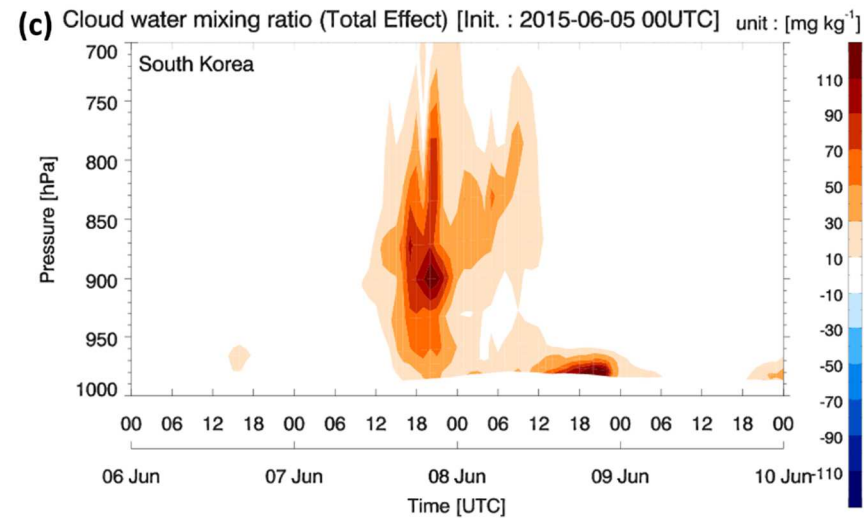
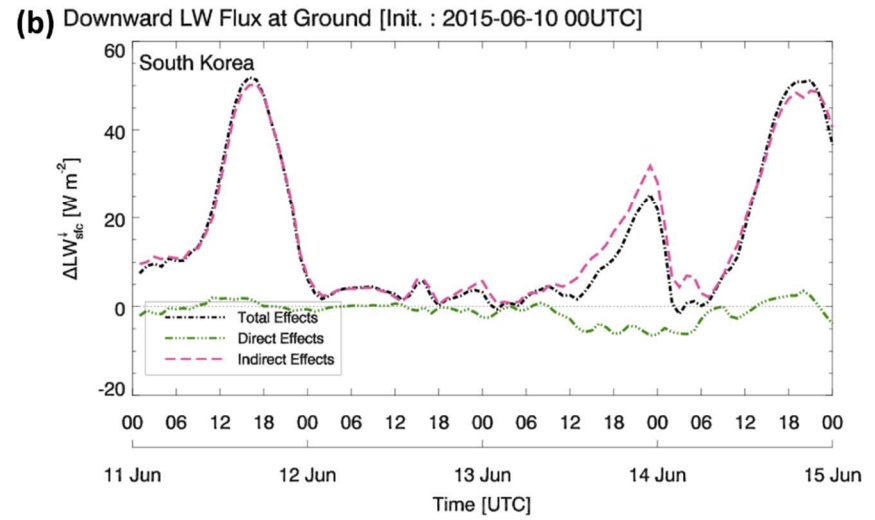
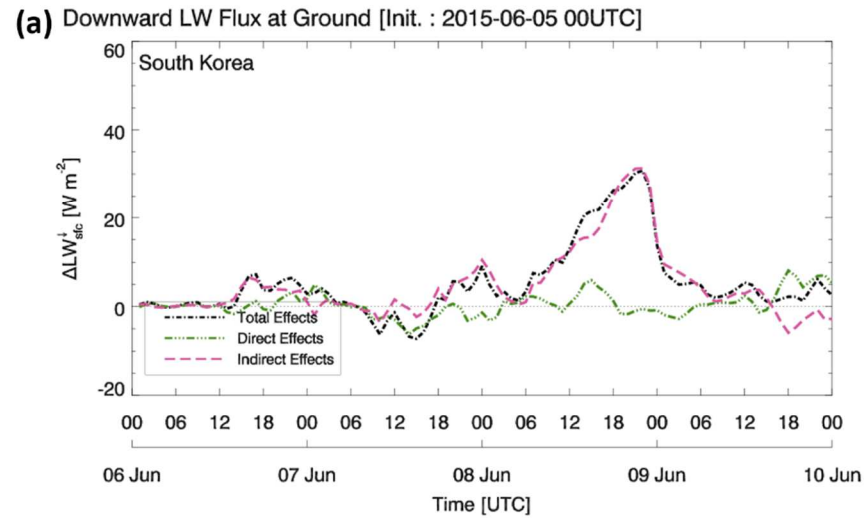
863 Fig. 5



864

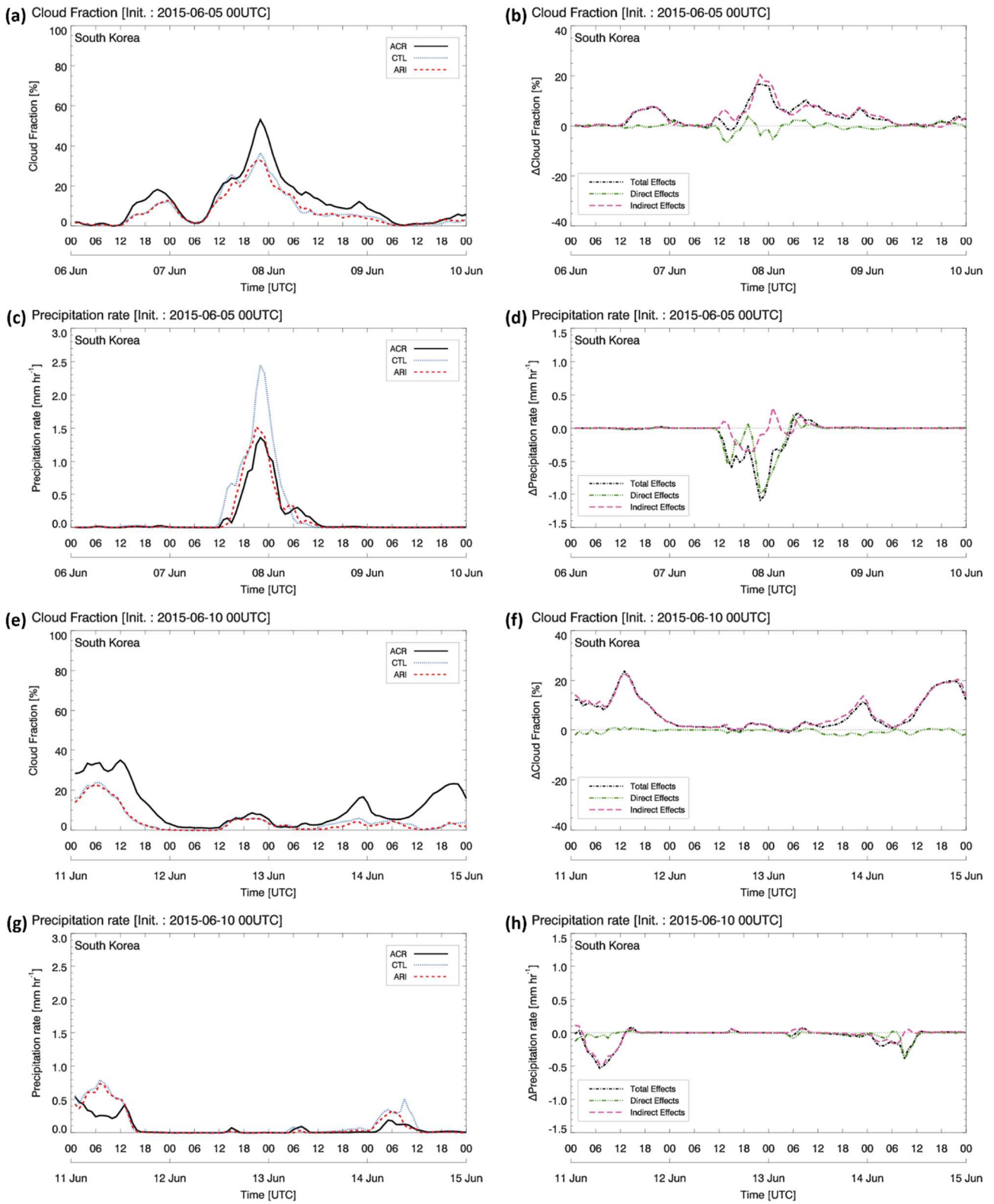
865





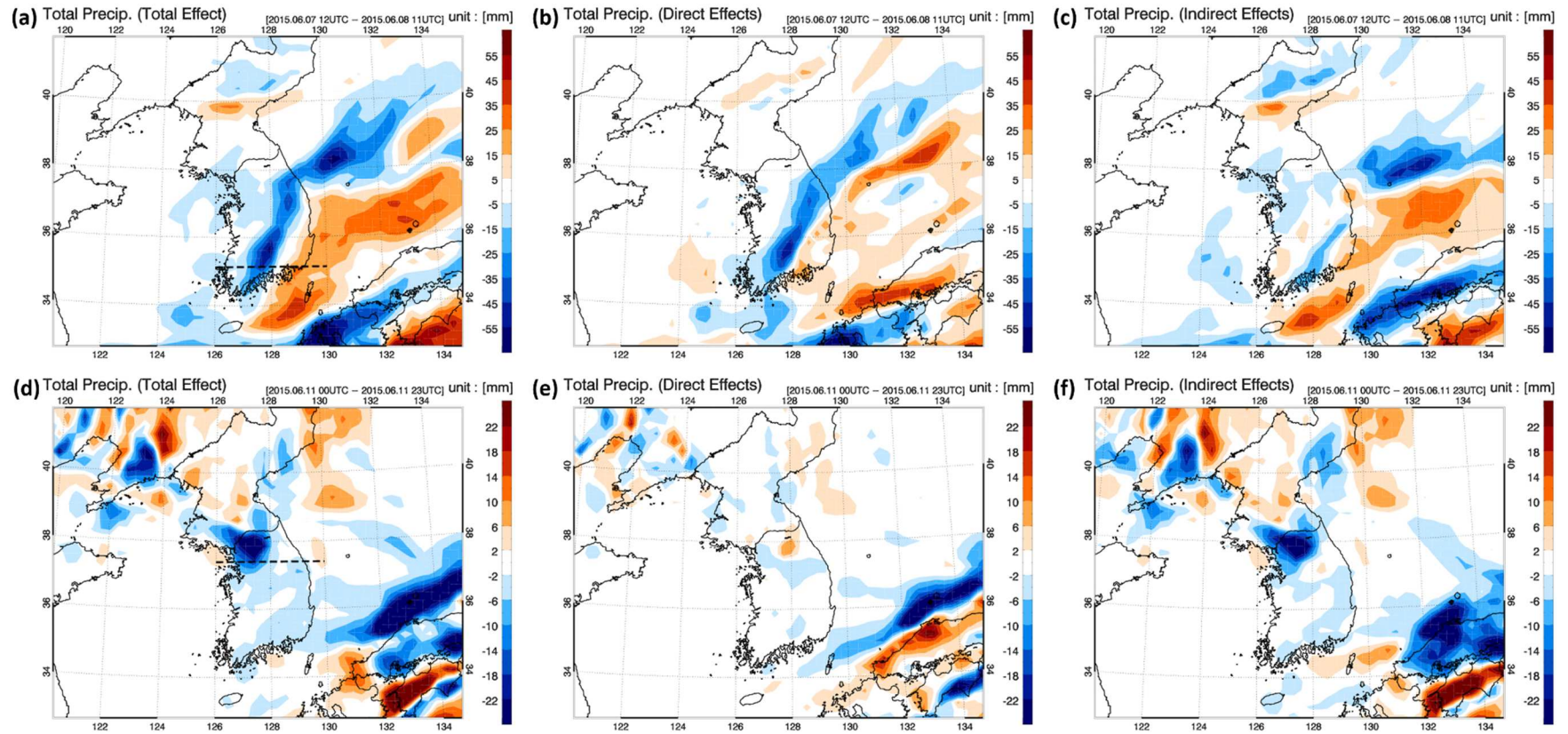
867

868



870

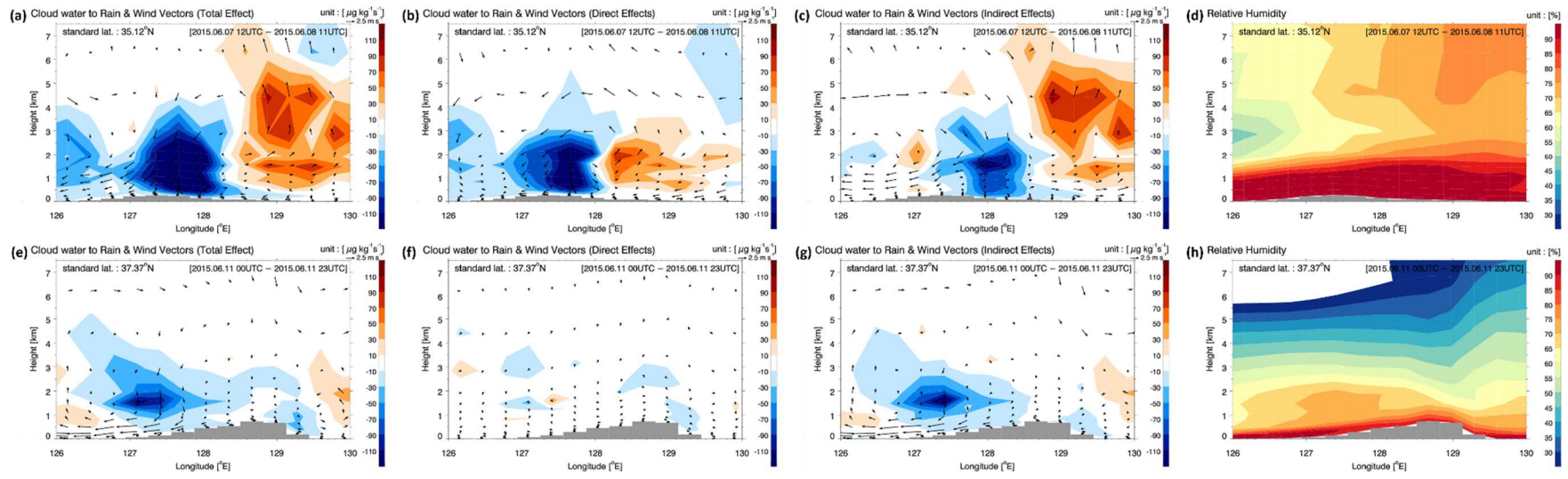
871



873

874

875 Fig. 9



876

877