



# Weekly variability of precipitation induced by anthropogenic aerosols: A case study in Korea in summer 2004



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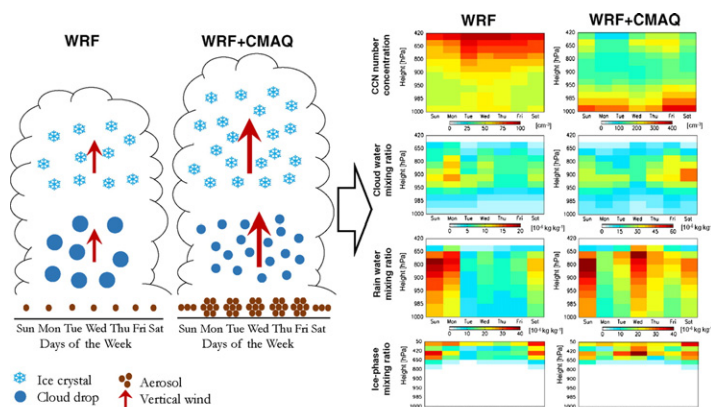
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## HIGHLIGHTS

- Effect of anthropogenic aerosols on weekly variability of precipitation in Korea
- Aerosols in Aitken and accumulation modes were considered in this work.
- Aerosols suppressed auto-conversion process from cloud water to rain water.
- Aerosols induced significantly stronger vertical updrafts in the mid-atmosphere.

## GRAPHICAL ABSTRACT



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## ABSTRACT

We examine the effect of anthropogenic aerosols on the weekly variability of precipitation in Korea in summer 2004 by using Weather Research and Forecasting (WRF) and Community Multiscale Air Quality (CMAQ) models. We conduct two WRF simulations including a baseline simulation with empirically based cloud condensation nuclei (CCN) number concentrations and a sensitivity simulation with our implementation to account for the effect of aerosols on CCN number concentrations. The first simulation underestimates observed precipitation amounts, particularly in northeastern coastal areas of Korea, whereas the latter shows higher precipitation amounts that are in better agreement with the observations. In addition, the sensitivity model with the aerosol effects reproduces the observed weekly variability, particularly for precipitation frequency with a high  $R$  at 0.85, showing 20% increase of precipitation events during the weekend than those during weekdays. We find that the aerosol effect results in higher CCN number concentrations during the weekdays and a three-fold increase of the cloud water mixing ratio through enhanced condensation. As a result, the amount of warm rain is generally suppressed because of the low auto-conversion process from cloud water to rain water under high aerosol conditions. The inefficient conversion, however, leads to higher vertical development of clouds in the mid-atmosphere with stronger updrafts in the sensitivity model, which increases by 21% cold-phase hydrometeors including ice, snow, and graupel relative to the baseline model and ultimately results in higher precipitation amounts in summer.

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## 1. Introduction

Aerosols in the atmosphere play an important role in cloud formation as cloud condensation nuclei (CCN). The chemical composition and number–size distribution of aerosols significantly modify cloud properties and precipitation processes by altering the droplet number concentration, droplet effective radius, cloud albedo, cloud liquid water content, and cloud lifetime (Abdul-Razzak and Ghan, 2002; Lohmann and Feichter, 2005; Ramanathan et al., 2001; Rosenfeld et al., 2014). The resulting formations of clouds and precipitation effectively remove aerosols from the atmosphere and affect their atmospheric lifetime and burden (Andronache, 2003; Bae et al., 2012). This interaction among aerosols, clouds, and precipitation is a large uncertainty in atmospheric models and requires an improved understanding to predict future Earth system changes induced by anthropogenic activities (Andreae and Rosenfeld, 2008; Lee, 2011).

Rapid economic development in East Asia has caused dramatic increases of aerosol precursor emissions such as SO<sub>2</sub> and NO<sub>x</sub> in recent decades (Kurokawa et al., 2013; Ohara et al., 2007; Zhang et al., 2009). Resulting aerosol concentrations have also increased in East Asia over the past two decades (Jeong and Park, 2013; Wang and Shi, 2010), whereas stringent air quality regulations in developed countries throughout Europe and North America have resulted in significant decreases of aerosol concentrations in the past (Richter et al., 2005; Zhang et al., 2007b) that are expected to continue in the future (IPCC, 2007). Although the effect of aerosols on clouds and precipitation is manifested in East Asia among the industrialized regions of the world, such conditions have been poorly represented in hydrological simulations of atmospheric models. In this work, we use a regional meteorological model to examine the effect of aerosols on clouds and precipitation development by improving the existing model simulations of cloud microphysics with the inclusion of explicit CCN activation. We focus our analysis on Korea, where long-term precipitation data are available.

Previous numerical studies have developed numerous methods on various scales to consider aerosols–cloud interaction (Isaksen et al., 2009; Khain, 2009; Kulmala et al., 2011; Quaas et al., 2009; Valipour, 2012) and have shown that the effect of aerosols on precipitation is highly sensitive to environmental conditions and cloud type (Fan et al., 2007; Khain, 2009; Makkonen et al., 2009; Rosenfeld et al., 2008; Tao et al., 2007). That is, the balance between the generation and the loss of the condensate mass determines the sign of the effect of aerosol on precipitation (Khain, 2009), which should also be critically sensitive to the generation and loss of aerosols. Although some studies showed that the distribution of CCN can have a significant impact on the cloud microphysics by affecting the droplet distribution (Cruz and Pandis, 1997; Feingold et al., 1999; Liu and Li, 2014; McFiggans et al., 2006), most cloud-resolving and regional meteorological models have included aerosols in a very simple manner by prescribing the fixed distributions of aerosols or CCN spectra (Bangert et al., 2011). The chemical composition and dynamics of aerosols have not been explicitly considered in those models. Instead, the CCN activation equation suggested by Twomey (1959) has been widely used in such models including the latest Weather Research and Forecasting (WRF) model (Khairoutdinov and Kogan, 2000).

In this work, we used a regional meteorological model coupled with chemical transport model simulations. Our main focus in this study is to improve the model simulations of cloud microphysics by updating the CCN activation in the WRF model and to show the resulting aerosol effects on precipitation. We compare the observed and simulated seasonal precipitation to validate the effect of aerosols on our cloud microphysics simulation in Korea. Moreover, we examine the effect of aerosols on the weekly variability of simulated precipitation as an additional validation of our improvement through comparison with the observation.

## 2. Model description

We used the WRF version 3.1.1 model to conduct seasonal simulations over East Asia for June–August 2004. The WRF is a mesoscale numerical weather prediction model designed to serve both operational forecasting and atmospheric research needs. As in other regional models, the WRF employs several numerical schemes for simulating cloud microphysics. In this study, we selected the WRF Double-Moment 6-class (WDM6) microphysics scheme, which was developed by Lim and Hong (2010) based on the WRF single-moment 6-class (WSM6) microphysics scheme (Hong et al., 2004; Hong and Lim, 2006). The improved scheme includes the prognostic calculation of number concentrations for cloud and rainwater together with CCN number concentrations. The explicit calculation of CCN number concentrations allows for examination of model sensitivity to changes in CCN number concentrations driven by changes in aerosol number concentrations. We discuss the CCN activation calculation in the model in more detail in Section 2.1; other detailed information on the microphysics scheme has been reported previously (Lim and Hong, 2010). The physics packages include the Grell–Devenyi cumulus parameterization scheme (Grell and Devenyi, 2002), the Noah land-surface model (Chen and Dudhia, 2001), the Yonsei University planetary boundary layer (YSU PBL; Hong et al., 2006; Hong, 2010), the longwave radiation schemes from the Rapid Radiative Transfer Model (RRTM; Mlawer et al., 1997), and a simple cloud-interactive radiation scheme (Dudhia, 1989).

### 2.1. CCN activation

The WDM6 scheme explicitly computes CCN number concentrations by using an empirical power law ( $N_{CCN} = CS^k$ ) as a function of the supersaturation ( $S$ ) and the index in the power law ( $k$ ; Twomey, 1959; Khairoutdinov and Kogan, 2000). However, the use of this equation is known to overestimate CCN concentrations, particularly at high supersaturation levels, because of the functional form of the power law and the use of a single value for  $k$  at 0.6 (Khvorostyanov and Curry, 2006; Lim and Hong, 2010). Many field and laboratory measurements have shown that CCN number concentrations are not linear in log–log coordinates, as would occur when  $k$  is constant, but vary with a concave curvature; that is, the  $k$  index decreases with an increase in  $S$  (Khvorostyanov and Curry, 2006). Many previous studies have developed various methods for correcting this issue (Khvorostyanov and Curry, 2006, and references therein). In the present study, we implemented in the WRF a modified power law developed by Khvorostyanov and Curry (2006), hereafter referred to as KC06, who corrected the overestimation of CCN concentrations by using a functional form of power law parameters  $C$  and  $k$  as functions of the size distribution and the solubility of aerosol.

To explore the effects of our implementation of the modified power law of KC06 on CCN concentrations in the WRF, we conducted simple calculations of CCN concentrations based on the assumed aerosol distributions by using three equations: 1) the power law (default in WDM6), 2) the Köhler equation with 100 sectional bins, and 3) the modified power law from KC06. We assumed that aerosols are activated when critical supersaturation of each bin is lower than supersaturation.

Fig. 1 compares the activated fractions of aerosols for two modes, Aitken and accumulation, as a function of supersaturation by using the Twomey relationship, the Köhler equation, and the modified power law of KC06. Aerosol size distributions are generally divided into three modes: Aitken (diameters nominally between 0.01 and 0.1 μm), accumulation (diameters nominally between 0.1 and 2.5 μm), and coarse (diameters greater than 2.5 μm) modes. The smaller Aitken mode represents fresh particles either from nucleation or from direct emission, while the larger accumulation and coarse modes represent aged particles (Binkowski and Roselle, 2003). In this study, however, only Aitken and accumulation modes were considered because anthropogenic aerosols exist mainly in the first two modes.

The activated fractions using the Twomey relationship were identically distributed with supersaturation for the two different aerosol modes; however, they were significantly higher than those calculated with other two methods for the Aitken mode and were lower for the accumulation mode at high supersaturation. This discrepancy occurred because aerosol size distributions were disregarded for CCN activation. On the contrary, the results using the modified power law of KC06 showed remarkably consistent distributions with the sectional results using the Köhler equation. Therefore, CCN activation with aerosol size changes is computed more effectively with the KC06 method than that by the Köhler equation.

## 2.2. Aerosol simulation

We conducted air chemistry model simulation by using the Community Multiscale Air Quality (CMAQ) model to obtain the spatial and temporal distributions of aerosol size and chemical compositions in East Asia. The CMAQ model is driven by meteorological fields from the fifth-generation mesoscale model (MM5) in the  $x$ ,  $y$ , and  $\sigma$  coordinates (Grell et al., 1994). The horizontal resolutions of the CMAQ model are  $45 \text{ km} \times 45 \text{ km}$  with 14 vertical layers. We used daily varying anthropogenic emissions for the CMAQ modeling system from the Sparse Matrix Operator Kernel Emissions—Asia (SMOKE—Asia) version 1.1. This system was developed by Woo et al. (2009) based on the Intercontinental

Chemical Transport Experiment—Phase B (INTEX-B) emission inventory for the base year of 2006 (Zhang et al., 2009) and includes the most updated fuel statistics in Asia (Woo et al., 2009). The aerosol simulation includes secondary inorganic aerosols (e.g., sulfate, nitrate, and ammonium), primary organic carbon (OC) and elemental carbon (EC) aerosols, and secondary organic aerosol (SOA). CMAQ also includes natural aerosols such as sea-salt and soil-derived aerosols. Atmospheric aerosols have mixed chemical composition, with a variety of inorganic and organic species often present in a single particle (Cruz and Pandis, 1997, 1998; Frosch et al., 2011). In this study, however, we only used anthropogenic aerosols in the Aitken and accumulation modes to examine the effect of anthropogenic aerosols on the weekly variability of precipitation. Additional details on the CMAQ model simulations can be found in Bae et al. (2012).

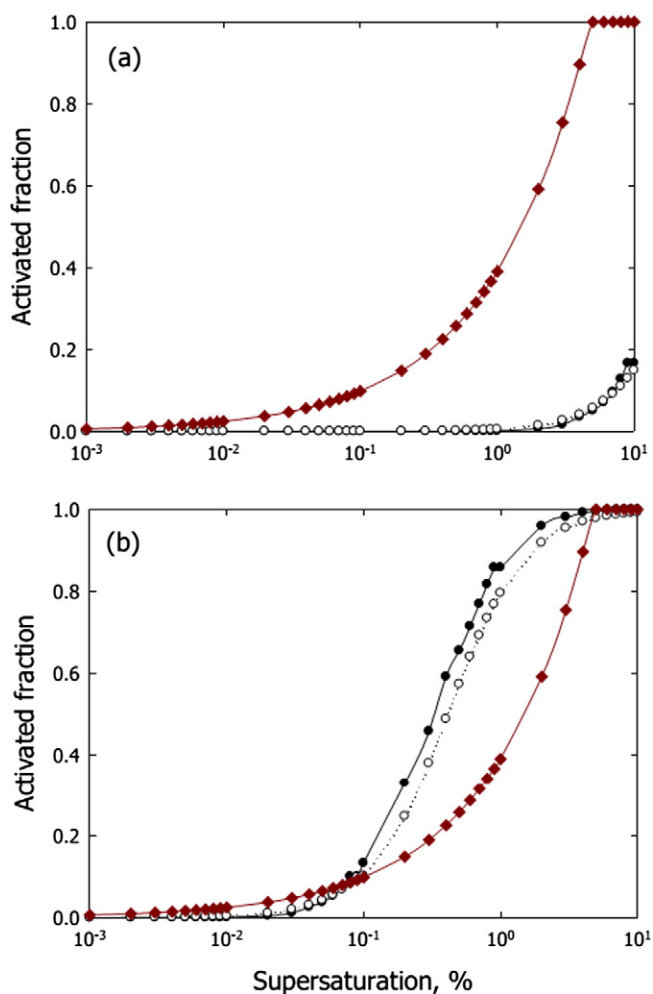
Anthropogenic aerosol concentrations can vary weekly owing to human activities that differ between weekdays and weekends (Gong et al., 2007). Fig. 2 shows weekly anthropogenic emissions of  $\text{SO}_2$  and  $\text{NO}_x$  over Korea ( $34.5\text{--}38^\circ\text{N}$ ,  $126.5\text{--}129.5^\circ\text{E}$ ). Although sulfur trioxide ( $\text{SO}_3$ ) is formed during the combustion of sulfur-containing fuels, an observed  $\text{SO}_3/\text{SO}_x$  ratio is very low in the range of 0.001–0.01 in flue gases emitted (Kikuchi, 2001; Fleig et al., 2011). Industrial activities and traffic throughout Korea are reduced on Saturdays and Sundays, leading to lower levels of emitted pollutants known as the “weekend effect”. Compared with that on Mondays through Fridays, the weekend emissions of  $\text{SO}_2$  and  $\text{NO}_x$  are reduced by approximately 30% (Fig. 2). Although the degree of reduction of industrial activities and traffic may differ among regions, weekend minima of anthropogenic emissions are generally expected for all regions.

Similar weekly variations are shown in aerosol number concentrations from the CMAQ model. Fig. 3 shows the weekly variability of simulated seasonal mean anthropogenic aerosol number concentrations for the accumulation mode from the CMAQ model in summer 2004 over Korea. The simulated aerosol number concentrations in surface air were  $1500\text{--}2500 \text{ cm}^{-3}$ , which is a typical range observed in East Asia (Gao et al., 2009; Shen et al., 2011; Wang et al., 2014). In general, the simulated concentrations during the weekends were lower than those of weekdays as a direct result of the decreased anthropogenic emissions with a maximum on Friday. The vertical profiles of the aerosol number concentrations show similar weekly variability with a sharp decrease with altitude. Values above 800 hPa were lower than  $500 \text{ cm}^{-3}$ , which is consistent with observations around the Korean Peninsula (Kim et al., 2014).

## 2.3. Experimental setup

To investigate the effect of aerosols on precipitation, two numerical simulations were conducted: a baseline and a sensitivity run. The baseline simulation used an empirical power law, which is a default value in WDM6, with a fixed CCN number concentration of  $100 \text{ cm}^{-3}$ . The sensitivity simulation used explicitly calculated aerosol number concentrations at each time step from the CMAQ model as input with the modified power law of KC06 for CCN activation. The results from these simulations were used for model evaluation and were analyzed to investigate the effect of aerosols on the weekly variability of precipitation over Korea.

We used one-way nested domains starting at a resolution of 45 km over East Asia including the Korean Peninsula (Fig. 4). The two nested domains have 15-km and 5-km resolutions, respectively. Our analysis primarily focused on the results from the innermost nested domain because the simulations at resolutions of 45 km and 15 km are too coarse to resolve the fine-scale variation in precipitation in Korea. Sub-grid cumulus parameterization was used only for simulations with horizontal resolutions of 45 km and 15 km because no cumulus scheme is generally needed for simulations with horizontal resolutions of 5 km or less (Hong and Dudhia, 2012). The model has 14 vertical layers with a top at 50 hPa. For initial and boundary conditions for the WRF simulations,



**Fig. 1.** Activated fractions of aerosols for (a) Aitken (top) and (b) accumulation (bottom) modes as a function of supersaturation using the Twomey equation in the original Weather Research and Forecasting (WRF) model (red diamonds), the Köhler equation (closed circles), and the modified power law from Khvorostyanov and Curry (2006; open circles), respectively.

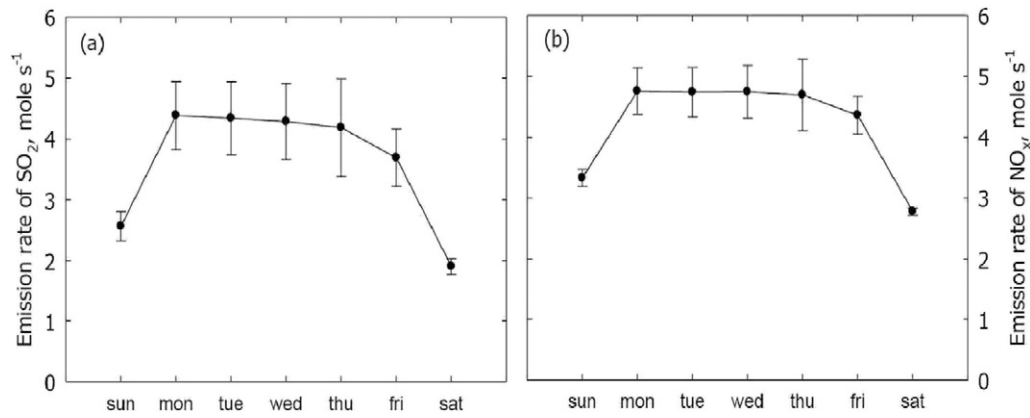


Fig. 2. Weekly emission rates of (a)  $\text{SO}_2$  and (b)  $\text{NO}_x$  at the surface layer in Korea.

we used the 6-h operational global analysis data from the National Centers for Environmental Prediction Final (NCEP FNL) with a  $1^\circ \times 1^\circ$  resolution every 6 h. The daily mean multi-scale ultra-high-resolution sea surface temperature (MUR SST) on a  $0.1^\circ$  grid was used as the ocean surface boundary condition. We conducted WRF simulations for 107 days with the first 15 days used as spin-up time. The precipitation fields within the third domain were analyzed from June 1 to August 31, 2004. All simulations were conducted by using the same initial and boundary conditions except for the CCN activation, as described above.

### 3. Effects of aerosols on simulated precipitation

Fig. 5 shows simulated precipitation for June–August 2004 in Korea from the baseline and the sensitivity models. For evaluation of the models, we used the observed daily precipitation amounts at 62 weather stations from the Korea Meteorological Administration (KMA) for June–August 2004. The observed precipitation in summer was typically high in the northeastern coastal areas of Korea owing to the topographic effect (Jung et al., 2012; Park and Lee, 2007). The models poorly captured the observed spatial patterns in the Korean Peninsula with low correlation coefficients at  $R < 0.2$  and tended to underestimate the precipitation amounts with mean biases of  $< -3.1 \text{ mm day}^{-1}$ . Many factors may have contributed to the simulated discrepancies, reflecting our poor understanding of the hydrological processes; however, such a topic is beyond the scope of this paper. We determined that the sensitivity model with explicit aerosols resulted in higher precipitation than that of the baseline model and is in better agreement with the observations with a slight increase in  $R$  and a reduced mean bias. Thus, we focused on the resulting precipitation changes driven by aerosols.

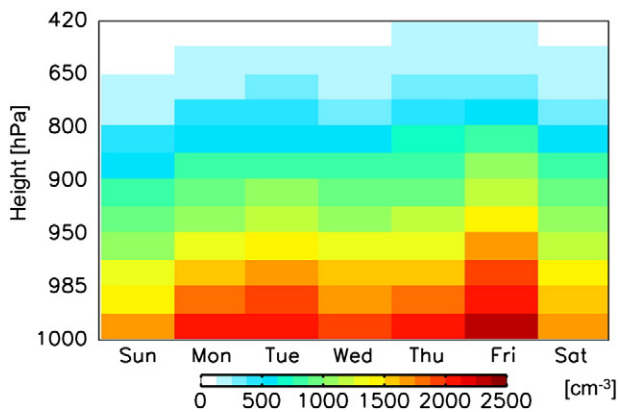


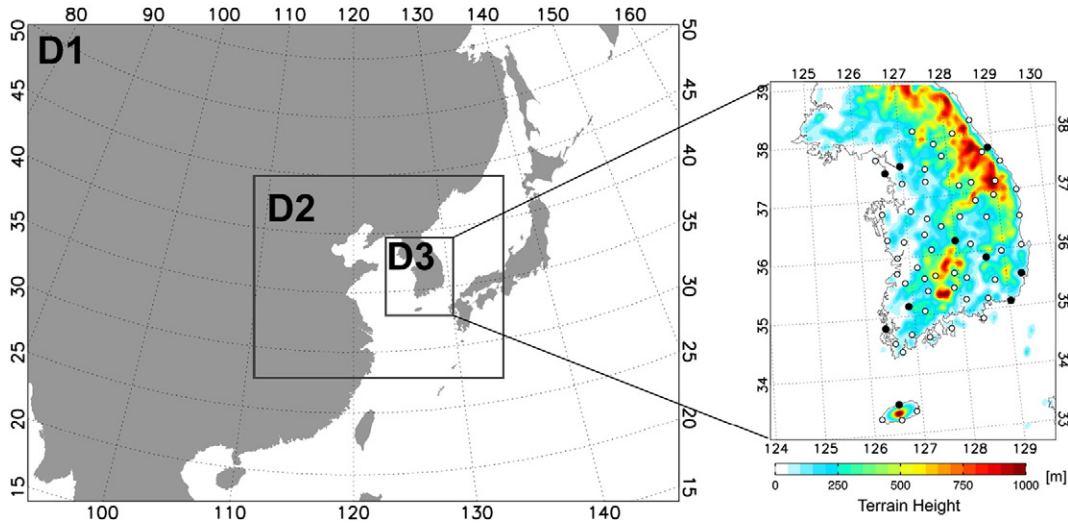
Fig. 3. Weekly variability of simulated seasonal mean aerosol number concentrations for the accumulation mode from the Community Multiscale Air Quality (CMAQ) model in summer 2004 in Korea.

Numerous studies have reported that an increase in aerosols generally tends to decrease precipitation in shallow clouds (Andreae et al., 2004; Li et al., 2011; Rosenfeld et al., 2008). In other cases, however, aerosols may enhance precipitation by invigorating deep convection and accelerating the conversion of cloud water to precipitation (Bell et al., 2008; Koren, 2005). Fig. 6 shows the time series anomalies of 11-year centered running means summer precipitation frequencies averaged at the 11 long-term monitoring KMA stations (see Fig. 4) in Korea for 1956–2005. The observed precipitation does not typically include trace precipitation of  $< 0.1 \text{ mm day}^{-1}$ . In our analysis, the trace precipitation days were considered as those of light rain at  $< 10 \text{ mm day}^{-1}$ ; however, the results are insensitive to the inclusion of trace precipitation events. Fig. 6 also shows the long-term trends for summer precipitation frequencies estimated using the least squares technique. The observed light rain frequency shows a pronounced decreasing trend of 1.4% per decade in Korea, particularly after the early 1980s. In addition, the seasonal precipitation amount in summer contributed by light rain showed a decrease (not shown). On the contrary, a significant change was shown in the frequency of heavy rain at  $\geq 20 \text{ mm day}^{-1}$  such that heavy precipitation occurred more frequently with an increasing trend of 1.2% per decade. This observed trend was also detected over East China, where Qian et al. (2009) showed a decreasing trend in light rain and an increasing trend in heavy rain from 1956 to 2005 and suggested that an increase in aerosol concentrations is partly responsible for the observed precipitation trends.

Fig. 7 shows the probability distribution functions (PDFs) of observed and simulated precipitation intensities for the baseline and the sensitivity simulations in summer 2004 in Korea. Compared with the results from the baseline, the sensitivity simulation appeared to simulate decreased light rain frequency, whereas an increase in heavy rain frequency was evident. This result is attributed to the suppressed auto-conversion process in the sensitivity simulation with high CCN concentrations (Lim and Hong, 2010). Our sensitivity simulations are consistent with the long-term observation trends, which show a decrease (increase) in light (heavy) rain frequency, and imply that the increase in aerosols is a likely cause for the observed precipitation trend over the past decades.

### 4. Effects of aerosols on weekly variability of precipitation

We further examined the effect of aerosols on the weekly variability of precipitation in Korea. Kim et al. (2009) previously analyzed long-term observed precipitation frequencies in the same region during 1975–2005 and suggested that the weekly cycles of precipitation might be attributed to changes in cloud fraction driven by aerosol variations between weekdays and weekends. Furthermore, Choi et al. (2008) analyzed observed daily precipitation frequencies in Korea during 1961–2007 and determined that after the mid-1990s, the

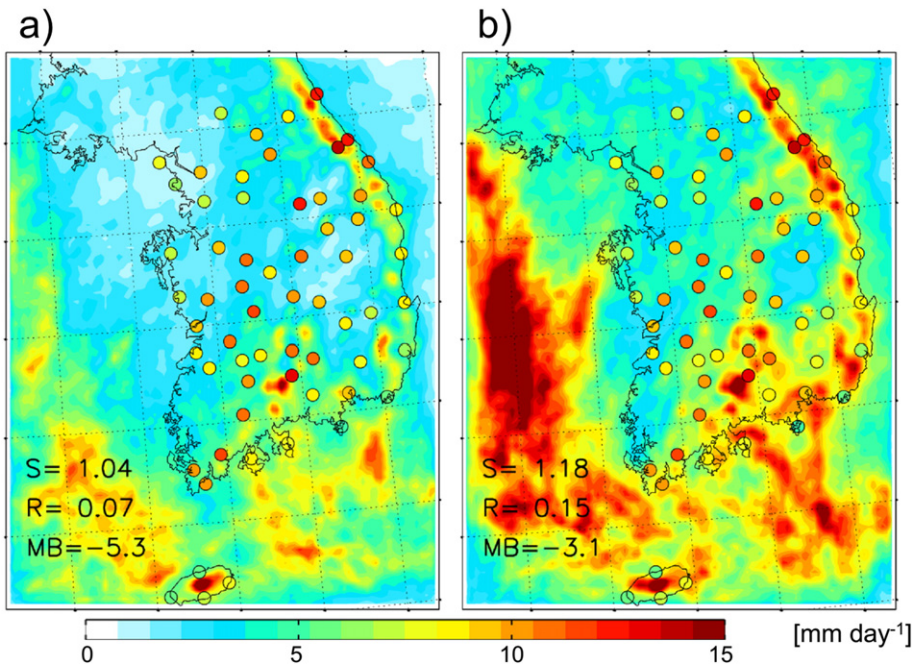


**Fig. 4.** Model domains with the 45-km resolution  $132 \times 97$  (D1), 15-km resolution  $132 \times 120$  (D2), and 5-km resolution  $105 \times 132$  (D3) grids. Surface sites of the Korea Meteorological Administration (KMA) are denoted with open circles in the innermost nested domain (D3); long-term observation sites are denoted with black dots (D3).

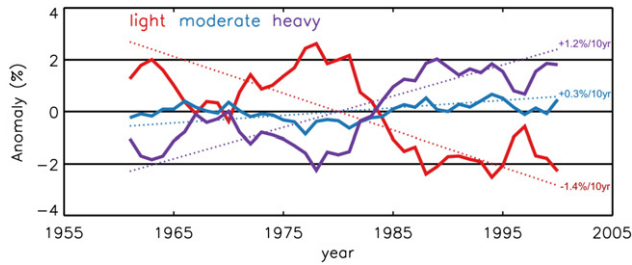
precipitation frequency in summer was significantly higher during the weekends than that on weekdays.

In the present study, we analyzed the observed changes in the weekly variability of precipitation frequencies in Korea by separating two periods: 1956–1975 and 1986–2005, referred to respectively as the 1960s and 1990s. We believe that the first period was less affected by anthropogenic activities relative to the latter, during which time rapid economic development has occurred in Korea. Fig. 8 shows the 1960s–1990s changes in weekly variability of the observed precipitation frequencies averaged at the 11 long-term monitoring KMA stations in Korea. We determined that the observed changes are positive for weekends and negative for weekdays, indicating that precipitation occurred more frequently during weekends during the 1990s relative to that during the 1960s.

The observed changes of the weekly variability between the two periods may be due to the variability of aerosol concentrations such that the weekday high aerosol concentrations during the 1990s reduced the precipitation occurrences. This conclusion is consistent with the results of previous findings such that aerosol loading caused by human activities suppresses the warm rain processes (Gong et al., 2007; Koren et al., 2004; Rosenfeld et al., 2007). Fig. 8 also shows the weekly variability of simulated precipitation frequencies from the baseline and the sensitivity simulations in summer 2004 at the 11 stations. Both models captured the observed variability to some degree. The sensitivity model, however, reproduced the weekly variability of precipitation frequency with high correlation at  $R = 0.85$ , exhibiting more consistent weekday and weekend variability of precipitation driven by the weekly variation of aerosol concentrations.



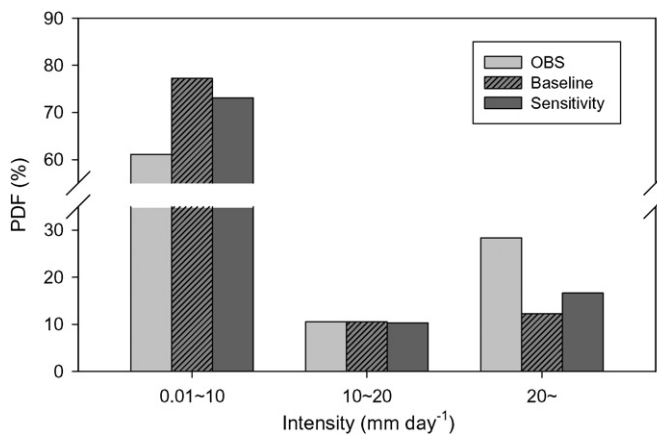
**Fig. 5.** Spatial distributions of the simulated precipitation for (a) baseline and (b) sensitivity runs in summer 2004 in Korea. The closed circles represent the observed data obtained from the 62 stations of the Korea Meteorological Administration (KMA). The inset shows reduced major-axis regression slopes ( $S$ ), correlation coefficients ( $R$ ) and mean bias ( $MB = M - O$ , where  $M$  and  $O$  are the modeled and observed values, respectively).



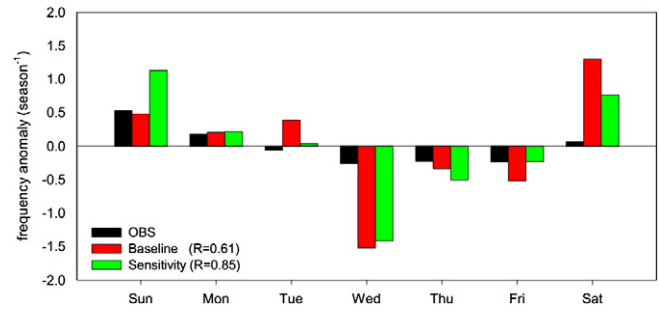
**Fig. 6.** Time series anomaly of 11-year centered running means of summer precipitation frequency for light rain ( $<10 \text{ mm day}^{-1}$ ), moderate rain ( $10\text{--}20 \text{ mm day}^{-1}$ ), and heavy rain ( $\geq 20 \text{ mm day}^{-1}$ ) intensities from 1956 to 2005 recorded at the 11 long-term monitoring Korea Meteorological Administration (KMA) stations represented in Fig. 4 with black closed circles. The linear-least squares fits of the data are shown.

To understand the mechanism for the simulated weekly variability of precipitation we examined the differences between the two models by comparing weekly CCN number concentrations, cloud water mixing ratios, and rainwater mixing ratios averaged over the model domain ( $34.5\text{--}38.0^\circ\text{N}$ ,  $126.5\text{--}129.5^\circ\text{E}$ ) from the baseline and the sensitivity simulations shown in Fig. 9. As previously discussed, the baseline model uses a fixed initial CCN number concentration of  $100 \text{ cm}^{-3}$  at each model grid and shows increasing CCN number concentrations with altitude because of the conversonal loss to raindrops in the low troposphere. Conversely, the sensitivity model generally shows decreasing CCN number concentrations with altitude and the highest CCN number in surface air driven by aerosol concentrations, as shown in Fig. 3. For Wednesday–Friday, the sensitivity model showed relatively high CCN concentrations above 800 hPa that are transported vertically from the surface. They subsequently formed clouds, with the maximum occurrence on Saturday. Compared with the baseline model, the sensitivity model showed a three-fold increase in cloud water content due to the high CCN number concentration. However, no such increase was detected in simulated rainwater content in the sensitivity model because the increase in CCN number and the resulting decrease in cloud droplet size suppressed the collection/coalescence processes and the raindrop formation. The reduced conversion thus allowed for greater cloud development, which after a few days resulted in the precipitation that occurred frequently during the weekends in the sensitivity model.

Interestingly, the rainwater mixing ratio from the sensitivity model also showed a high value around 700 hPa on Wednesday caused by a strong vertical updraft, as shown in Fig. 10, which produced cold-phase hydrometeors. In a cloud modeling study, Khain et al. (2005)



**Fig. 7.** Probability distribution functions (PDFs) of the observed and simulated precipitation intensities for light rain ( $<10 \text{ mm day}^{-1}$ ), moderate rain ( $10\text{--}20 \text{ mm day}^{-1}$ ), and heavy rain ( $\geq 20 \text{ mm day}^{-1}$ ) in summer 2004 at the 62 stations in Korea.



**Fig. 8.** Observed (black bars) change in the weekly variability of precipitation frequency (difference between 1986–2005 and 1956–1975) averaged at the 11 long-term monitoring sites in Korea. Simulated weekly anomalies of precipitation frequencies were computed by using the results from the baseline (red bars) and the sensitivity (green bars) simulations for summer 2004. Correlation coefficients between the observation and the model results are shown to reflect the effect of aerosols on the weekly variability of precipitation.

showed that an increased updraft was generally associated with an increased aerosol amount by allowing additional moisture to reach the freezing level. Compared with the results of the baseline simulation, the sensitivity model showed relatively high vertical velocity in the mid-troposphere, particularly during the weekends, causing frequent precipitation. The stronger updraft caused the cloud water to extend vertically to higher levels, leading to enhanced cold rain processes that generate ice phase contents such as ice, snow, and graupel (Lim and Hong, 2012).

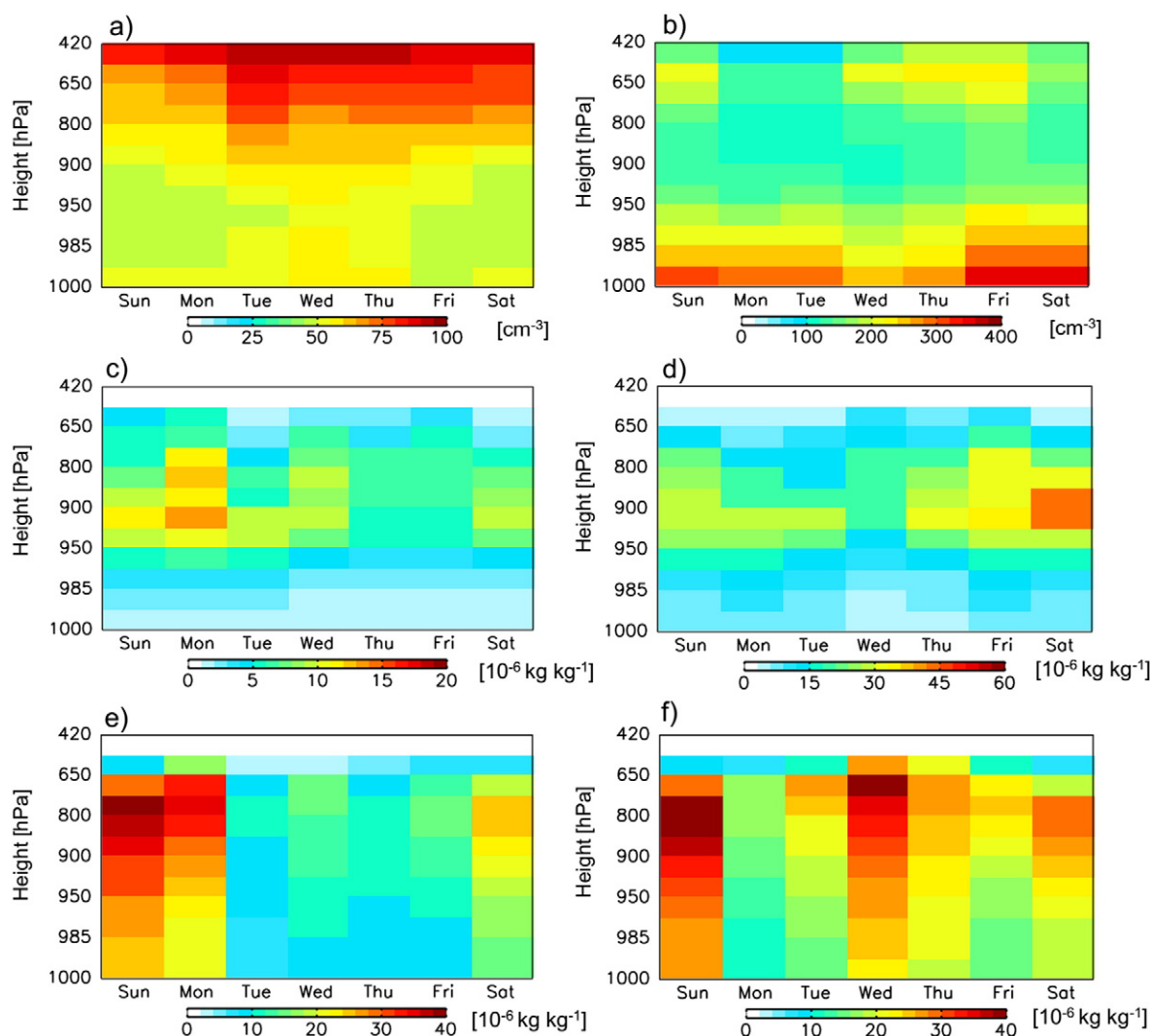
Fig. 10 also shows the weekly variability of simulated ice-phase hydrometeors such as ice, snow, and graupel determined in the baseline and the sensitivity simulations. The sensitivity model produced higher amounts of ice phases than the baseline model, which was also caused by the aerosol effect. The weekly variability of ice phases generally correlated with that of the updraft velocities, although it did not appear to contribute significantly to the weekly variability of precipitation frequencies. Owing to high temperature in summer, the ice phases quickly melt and evaporate while they precipitate. Occasionally, if a high cloud develops, the large amount of ice-phase content causes a heavy rain event in summer (Lim and Hong, 2012).

## 5. Summary and discussion

Atmospheric aerosols are known to have considerable direct and indirect impacts on the atmospheric processes. Changes in CCN number concentrations as a function of ambient aerosol number concentrations affect cloud formation and precipitation. In this work, we used a regional meteorological model coupled with chemical transport model simulations to examine the effect of anthropogenic aerosols on precipitation in Korea. We paid particular attention on the weekly variability of precipitation by conducting model simulations using fixed CCN number concentrations in the baseline model and simulated aerosol concentrations from the CMAQ in the sensitivity model.

First, we conducted model evaluation by comparing the simulated and observed precipitation amounts for summer 2004 in Korea. The baseline model generally underestimated the observed precipitation, particularly in northeastern coastal areas of Korea, whereas the sensitivity model showed increased precipitation amounts, resulting in closer agreement with the observations. Moreover, the sensitivity model successfully reproduced the 1960s and 1990s changes in the observed weekly variability of precipitation frequency with a high  $R$  at 0.85. These changes were presumably caused by increases in aerosol concentrations, indicating the considerable effect of anthropogenic aerosols on the weekly variability of precipitation in Korea.

For understanding the mechanism of the simulated response to the aerosol effect in the model, we examined the simulated hydrometeor



**Fig. 9.** Domain-averaged weekly variability of the simulated (a and b) cloud condensation nuclei (CCN) number concentration, (c and d) cloud water mixing ratio, and (e and f) rain water mixing ratio for the baseline (left panel) and the sensitivity (right panel) simulations in summer 2004 over Korea (34.5–38°N, 126.5–129.5°E). It should be noted that the plots use different scales in the CCN number concentration and cloud water mixing ratio.

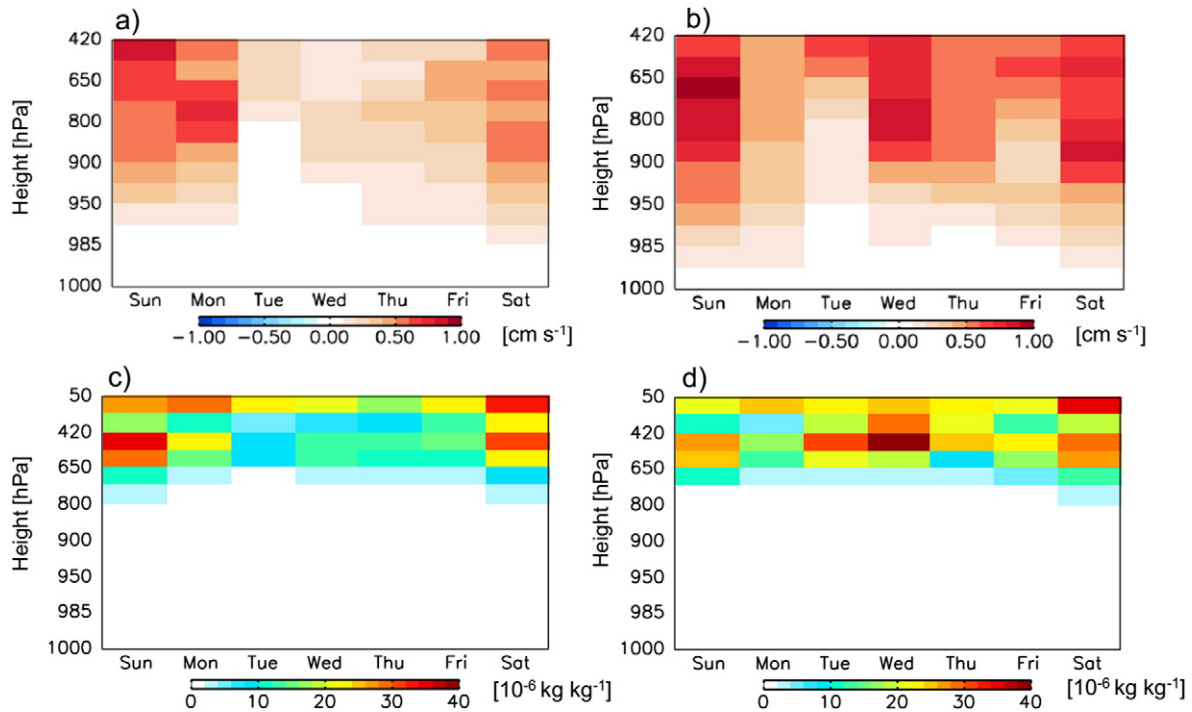
concentrations. The cloud water contents from the baseline and the sensitivity simulations showed similar weekly variability; however, the sensitivity model showed a three-fold increase over the results of the baseline model owing to the high CCN number concentration. Unlike the cloud water contents, the rainwater contents of the baseline and the sensitivity simulations were similar due to the low auto-conversion rate caused by the relatively small cloud droplet size in the high aerosol environment in the sensitivity simulation. The reduced conversion allows for higher cloud development and thus resulted in frequent precipitation during the weekends in the sensitivity model. Moreover, the aerosol effect induced significantly stronger vertical updrafts, which produced increased amounts of ice-phase hydrometeors such as ice, snow, and graupel. However, these ice phases did not appear to contribute significantly to the weekly variability of precipitation frequency.

Although we examined the effect of the aerosols on the weekly variability of precipitation in Korea by using our best simulations, some limitations of our work should be noted. First, the sensitivity simulation was conducted in off-line fashion by using the simulated aerosol concentrations from CMAQ as input; thus, it did not fully account for two-way interactions between meteorology and aerosols. Second, laboratory experiments have shown that the hydrophobic organic coatings on inorganic cores could modify the CCN activation behavior (Abbatt et al.,

2005; Cruz and Pandis, 1998; Raymond and Pandis, 2003; Yu et al., 2013), so that a consideration of the CCN activity of mixed materials (Andreae and Rosenfeld, 2008; Sun and Ariya, 2006) is required in future studies. This could be particularly important for East Asia where organic aerosol has become important these days (Heald et al., 2005; Zhang et al., 2007a). Third, we conducted the model evaluation for summer 2004. Although the meteorological conditions in this period were similar to those of the previous years, such short-term simulation might be insufficient for fully representing the effect of anthropogenic aerosols on the weekly variability of precipitation. Therefore, further research using an improved model for a long-term simulation is necessary for examining the effect of aerosols on precipitation in East Asia with a particular focus on the rapid economic development periods in the recent decade.

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**Fig. 10.** Domain-averaged weekly variability of simulated (a and b) vertical velocity and (c and d) ice-phase hydrometeor (including ice, snow, graupel) mixing ratios for the baseline (left panel) and the sensitivity (right panel) simulations in summer 2004 over Korea (34.5–38°N, 126.5–129.5°E).

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