Effects of the meteorological variability on regional air quality in East Asia
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HIGHLIGHTS
• Examine the effects of the meteorological variability on regional air quality.
• Cloudiness and temperature account for 30% of total O3 increases in East Asia.
• Increases in mixing depths suppress increases of (NH4)2SO4 in summer.
• Decreases in mixing depths in winter result in enhancement of NH4NO3.

ABSTRACT
We examine the effects of the meteorological variability on O3 and SO4/NO3/NH4 aerosol concentrations in East Asia using 3-D chemical transport model (GEOS-Chem) simulations for the period of 1985–2006. The model was driven by the GEOS assimilated meteorology with the emission estimates from the Streets et al. inventory with annual scale factors of Regional Emission inventory in Asia (REAS). Over the past two decades precursor emissions have been dramatically increased. Our model simulations however show strong non-linear responses of oxidation products to the increases of those precursors. The analysis of simulated results shows significant effects of meteorological variability on O3 and SO4/NO3/NH4 concentrations. Springtime O3 concentration has been generally increased over the past two decades mainly due to increases in anthropogenic precursor emissions but concurrent changes in meteorology including decreases of cloud covers and increases of temperature further enhance O3 increases in East Asia. Our analysis reveals that changes in meteorology account for 30% of total O3 increases in East Asia over the past two decades. On the contrary, increases in mixing depth suppress increases of (NH4)2SO4 concentrations in summer but decreases in mixing depth in winter result in enhancement of NH4NO3 aerosols concentrations up to 4 µg m–3 in eastern China. Effects of meteorological variability on SO4/NO3/NH4 aerosol concentrations are thus seasonally dependent such as a decrease in summer by 4% but an increase by 7% in winter over the past two decades. This result indicates that the meteorological conditions have changed more favorable for the PM air quality degradation in winter.

1. Introduction
East Asia is one of the largest source regions of gaseous pollutants and aerosols due to the rapid industrialization, urbanization and the population growth (Zhang et al., 2007; Singh et al., 2009). Therefore, air quality in East Asia has been deteriorated over the past and has caused serious concerns for human health and economic losses. Previous studies have shown that the observed springtime background O3 concentrations at Hatto in Japan have increased by 0.6 ppbv yr–1 during the last decade (Tanimoto et al., 2009) and other surface O3 values in western and central Japan (WCJ) have also shown similar increasing trends of 0.37 ppbv yr–1 for the past depending on regions and time of years (Kurokawa et al., 2009). These increases are primarily attributed to increases in anthropogenic precursor emissions in East Asia based on the analysis of chemical transport model simulations (Kurokawa et al., 2009; Wang et al., 2009; Chatani and Sudo, 2011).

Together with O3, the other important air pollutant, aerosol concentrations have also increased during the past decade. Previous studies on aerosol concentrations and their long-term changes are relatively few in East Asia and recently, Kim et al. (2011) showed that the observed SO2−4, NO3, and NH4 mass concentrations at a background site of East Asia (Gosan, Cheju island, Korea) increased by 21%, 107%, and 42% from 1992 to 2008, respectively. They suggested that the observed increasing SO2−4
NO$_3$ and NH$_4^+$ concentrations were likely related to the emission change of China and Korea.

The changes of air pollutant concentrations over the past decades might have largely reflected their precursor emissions changes (Streets et al., 2009). However, because the simultaneous increases of long-lived greenhouse gas concentrations have affected climate on global and regional scales (IPCC, 2007) corresponding changes in meteorological variables may indirectly affect pollutant concentrations (Jacob and Winner, 2009). For example, O$_3$ is highly sensitive to air temperature (Cox and Chu, 1996; Lin et al., 2001). Increases in temperature may also lead to the enhanced SO$_4^{2-}$ concentration with the faster SO$_2$ oxidation, but NO$_3$ is expected to decrease as a result of NO$_3$ and NH$_4^+$ partitioning to the gas-phase (Dawson et al., 2007). SO$_4^{2-}$ concentration is also expected to increase with increasing cloudiness due to increases in aqueous production but an accompanied increase in precipitation may have an opposite effect due to increases in wet scavenging (Pye et al., 2009; Tai et al., 2010). Air pollutant concentrations are thus subject to changes in meteorology (Jacob and Winner, 2009).

In this study we attempt to better understand the effects of meteorological variability on regional air quality degradation. By doing this we would obtain better quantitative information on how anthropogenic emission can directly affect pollutant concentrations as well as how meteorological variability can indirectly affect pollutant concentrations. This quantitative segregation offers a valuable metrics for air quality protection measures and can further be used to assess the effect of the future climate changes on air pollutants. The understanding on the past changes up to the present would be a valuable basis to evaluate the models used for predicting future changes.

In particular, the interaction between the climate and air pollutants has been under intense scrutiny for the future (Jacob and Winner, 2009, and references therein). Several studies using chemical transport models have investigated the effects of future climate change on O$_3$ and aerosols and its radiative climate forcing (Levy et al., 2008; Menon et al., 2008; Liao et al., 2009). Jacob and Winner (2009) summarized that climate change alone will vary emissions and meteorological climate change on O$_3$ and aerosols and its radiative climate forcing (Winner, 2009, and references therein). Several studies using chemical transport models have investigated the effects of future climate change on O$_3$ and aerosols and its radiative climate forcing (IPCC, 2007) corresponding changes in meteorological variables may indirectly affect pollutant concentrations (Jacob and Winner, 2009). For example, O$_3$ is highly sensitive to air temperature (Cox and Chu, 1996; Lin et al., 2001). Increases in temperature may also lead to the enhanced SO$_4^{2-}$ concentration with the faster SO$_2$ oxidation, but NO$_3$ is expected to decrease as a result of NO$_3$ and NH$_4^+$ partitioning to the gas-phase (Dawson et al., 2007). SO$_4^{2-}$ concentration is also expected to increase with increasing cloudiness due to increases in aqueous production but an accompanied increase in precipitation may have an opposite effect due to increases in wet scavenging (Pye et al., 2009; Tai et al., 2010). Air pollutant concentrations are thus subject to changes in meteorology (Jacob and Winner, 2009).

Fig. 1 shows the trends of Asian anthropogenic emissions in the model for 1985–2006. Values have increased 40% for CO, 12% for NO$_x$, and 60% for SO$_2$ over the past two decades due to increases in combustion facilities of fossil fuel (Ohara et al., 2007; Zhang et al., 2009; Lu et al., 2011). Meanwhile, NH$_3$ emission shows a gradual increase (44% for 1985–2006) due to the steady growth in agricultural production and fertilizer uses. The scale factor of CO was applied to anthropogenic VOCs in the model. Biogenic emissions of isoprene and monoterpenes are from the Model of Emissions of Gases and Aerosols from Nature (MEGAN) inventory (Guenther et al., 2006), and are dependent on solar radiation and temperature. NH$_3$ emissions are superimposed relative seasonal variation based on the length of the growing season for fertilizer use and on temperature (Fisher et al., 2011). Fig. 2 shows the GEOS assimilated meteorological data: monthly anomalies (differences between the monthly values and the average for 1986–2006) of mean surface temperature, cloud fractions, and

**2. Model description**

We use the global chemical transport model (GEOS-Chem) to conduct a fully coupled oxidant–aerosol simulation (Bey et al., 2001; Park et al., 2006). The GEOS-Chem model v8-03-01 (http://www.as.harvard.edu/chemistry/trop/geos) uses the assimilated meteorological data from the Goddard Earth Observing System (GEOS-4) of the NASA Global Modeling and Assimilation Office. The data include winds, convective mass fluxes, temperature, clouds, and precipitation at 6-h frequencies with a horizontal resolution of $1^\circ \times 1^\circ$ and 55 hybrid pressure-sigma levels up to 0.01 hPa. We degrade these meteorological fields to a horizontal resolution of $2^\circ \times 2.5^\circ$ and 30 vertical levels for computational expediency. The GEOS-Chem includes more than 80 species and 300 reactions for the detailed O$_3$−NO$_x$−hydrocarbon chemistry coupled with aerosol chemistry. The aerosol simulation includes H$_2$SO$_4$−HNO$_3$−NH$_3$ aerosol thermodynamics (Park et al., 2004), and the formation of these aerosols is computed locally with the ISORROPIA II aerosol thermodynamic equilibrium model (Fountoukis and Nenes, 2007).

We use the latest gridded anthropogenic monthly emissions for 2006 over Asian domain (60°E–158°E and 13°S–54°N) (Streets et al., 2006; Zhang et al., 2009). The inventory was based upon the studies by Streets et al. (2006), which improved estimations for Chinese industrial sources that had been underestimated in the inventory developed earlier (Streets et al., 2003). We then apply annual scale factors of Regional Emission inventory in Asia (REAS; Ohara et al., 2007) for 1985–2006 to the Streets et al. emissions to impose interannual variations in the model. The REAS inventory includes anthropogenic emissions for 1985–2003 estimated based on the fuel combustion and industrial sources and projected emissions after 2004.

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convective and large-scale precipitations in East Asia. Fig. 2 also displays the 12-months running mean of monthly anomalies of those variables. Surface temperature has increased by about 0.9 K, whereas cloud fraction has decreased by about 4% in East Asia over the past two decades. Those long-term observed changes are well consistent with the climate model simulations driven by the greenhouse gas changes. But in 1998/1999, cloud fraction was dramatically increased in East Asia, which was associated with one of the strongest ENSO events during the recent decades. A previous study by Xia (2012) found that significant decrease of cloud cover is evident over much of China over recent decades due to larger increase in clear sky frequency and larger decreases in overcast frequency, and largely attributable to high and middle-layer clouds (Warren et al., 2007).

The GEOS convective and large-scale precipitations also show dramatic increases due to that ENSO event but the long-term changes are opposite such that the convective precipitation has increased but the large-scale precipitation has decreased in East Asia over the past two decades. Convective precipitation is in general highest during the Asian summer monsoon in summer-time. Large-scale precipitation is more important than convective precipitation in other seasons and is well correlated with changes in cloud fraction. In Sections 4 and 5 we will examine the effects of these changes of meteorological variables on air pollutant concentrations in East Asia.

3. Model evaluation

The data from the Acid Deposition Monitoring Network (EANET, available at http://www.eanet.cc) are used to evaluate the model, focusing on O3, SO4^2-, NO3 and NH4^+ concentrations. EANET sites are mainly located in islands, rural regions and mountains, to avoid any direct influence from local pollution. The data include the monthly averaged surface concentrations of gaseous pollutants, soluble inorganic aerosols, and particulate matter mass concentrations.

First, we focus our evaluation on simulated O3 and aerosol concentrations. Fig. 3 compares the observed and simulated 5-yr (2002-2006) averaged O3 and SO4^2-, NO3 and NH4^+ concentrations in surface air at the EANET sites, which are mostly located in South Korea and Japan. The EANET observations are not available for the years earlier than 2002. In general, O3 values are high in the continent and the highest concentration is shown over Tibet owing to high elevation; Background O3 concentration increases with altitude (Logan, 1999). It is quite interesting that O3 is not high in eastern China where a large amount of NOx is emitted and this is due to the NOx titration. On the contrary, the highest SO4^2- and NH4^+ concentrations occur in eastern China, reflecting the combined effects of intense industrial activities and agricultural NH3 emissions. The spatial distribution of NO3 is similar to that of NH4 and reflects the general limitation of NH4NO3 formation by the availability of ammonia (Seinfeld and Pandis, 2006). Despite the lack of observations in China the spatial distribution of simulated O3 and aerosols is relatively in good agreement with observations in the downwind regions (Fig. 3).

Fig. 4 shows scatterplots of the simulated vs. observed annual and seasonal mean O3 and SO4^2-, NO3 and NH4^+ concentrations in surface air. Regression lines are computed here and elsewhere with the reduced major axis method (Hirsch and Gilroy, 1984). The observed O3 values show a maximum value in spring and a minimum in summer. The highest concentration in spring is due to an increase in photochemical O3 production, whereas the lowest concentration in summer is attributed to the Asian summer monsoon, which accompanies precipitating cloud and clean marine air mass (Kajii et al., 1998; Kondo et al., 2004; He et al., 2008).
simulated values show an overestimate of 10 ppbv especially in summer, likely due to the model’s incapability of simulating marine boundary layer in summer. The model tends to overestimate 1–7 ppbv in other seasons. Site by site comparisons show poor correlation coefficients between the model and the observations. These discrepancies reflect too coarse horizontal resolutions to reproduce observed O3 that is significantly affected by local emissions of NOx and VOCs (Fiore et al., 2002; Park et al., 2003). Despite of this model weakness, the model generally captures the variation of seasonal mean concentrations averaged over East Asia, which can be used to analyze the effect of meteorology on O3 on a regional scale.

Fig. 4 also shows a comparison of the observed and simulated annual and seasonal mean concentrations of SO4−/NO3−/NH4+ aerosols at the EANET sites. From spring to summer, observed SO4− concentrations are higher than those in the winter. This is because of the enhanced photochemical production through both gas-phase and aqueous-phase chemical oxidation. Whereas, the observed NO3− concentrations display an opposite seasonal variation with a winter maximum and a summer minimum because of aerosol NO3− formation favored with lower temperature in winter. NH4+ aerosol largely follows those of SO4− and NO3− and therefore no strong seasonal variation is shown in the observed NH4+ concentrations at the EANET sites. Overall, the model reproduces quite successfully observed SO4− and NH4+ aerosol concentrations with R² of 0.5–0.8 and regression slopes of 0.6–1.0. In addition, the regression slopes of the mean equivalent ratios of observed and simulated [NH4+]/(2[SO4−] + [NO3−]) are 0.69 and 0.96, respectively. The model is generally less acidic than the observations because the anions in particles can be neutralized by other alkaline cations (e.g., Ca2+) that are not included in the model. This also indicates that NO3− in the model is mostly NH4NO3. ISORROPIA II module in GEOSChem computes gas-aerosol equilibrium partitioning of HNO3, NH3 on other minerals. However, mineral components such as Ca2+, Mg2+ and K+ aerosols are not considered in the present study.

4. Long-term trend of springtime O3 concentrations

This section discusses the long-term trend of the simulated O3 in spring when O3 in ambient air is highest over East Asia due to the favorable sunny and warm conditions for photochemical O3 production. Fig. 5 shows the simulated trends of seasonal surface O3 concentrations in East Asia for April–June of 1985–2006. Values are anomalies, computed as deviations from the mean value averaged for 1985–2006. As discussed above, two model simulations driven by the assimilated meteorology are conducted. The first denoted with EyyMyy uses annually varying anthropogenic emissions and the latter (E06Myy) uses fixed emissions for 2006. The surface O3 concentrations from the EyyMyy simulations show an increasing trend of 0.30 ppbv yr⁻¹ in East Asia. East Asian countries including eastern China, Korea, and Japan show different increasing
trends of 0.45, 0.28, and 0.14 ppbv yr\(^{-1}\), respectively. Previous studies showed that the observed springtime O\(_3\) concentration at Happo and WCJ increased by 0.60 and 0.42 ppbv yr\(^{-1}\) (Kurokawa et al., 2009; Tanimoto et al., 2009), respectively, and our model is also consistent with respective rates of increase of about 0.65 and 0.23 ppbv yr\(^{-1}\), respectively.

With the gradual long-term trends of O\(_3\) concentrations, year-to-year variations are more pronounced such as sharp decreases of O\(_3\) in 1998 and 2002–2003 (Fig. 5). A previous study by Kurokawa et al. (2009) showed that observed springtime low O\(_3\) over Japan in spring 1998 and 2003 was strongly related to El Niño with surface pressure positive anomaly in the western Pacific. Japan is generally influenced significantly by Asian continental outflow, but during the El Niño periods a massive anticyclone centered in the eastern Pacific weakens Asian continental outflow and consequently an influence of maritime air masses from the western Pacific becomes larger than during ordinary years resulting in the decrease in O\(_3\).

\[ y = 0.14x + 17.0, \quad R^2 = 0.38 \]

\[ y = 0.80x + 0.69, \quad R^2 = 0.58 \]

\[ y = 0.99x + 0.45, \quad R^2 = 0.67 \]

\[ y = 0.29x + 0.23, \quad R^2 = 0.75 \]

\[ y = 0.75x + 0.57, \quad R^2 = 0.75 \]

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Fig. 4. Scatterplots of simulated versus observed annual mean O\(_3\) (units: ppbv) and SO\(_4^{2-}\)–NO\(_3^-\)–NH\(_4^+\) (units: \(\mu g \text{ m}^{-3}\)) concentrations at EANET sites. Values are annual mean (top panel) and seasonal means for 2002–2006. Sites in East Asia (Fig. 3) are shown. Reduced major axis regressions for the ensemble of the data (thick lines) are shown; regression equations and \(R^2\) are shown inset. Dashed lines denote a factor of 2 departures. The simulated (red color) and observed (green color) mean values are shown in the upper left and right corner of each panel, respectively. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)
emissions. Therefore, the results reflect the changes in springtime O$_3$ caused solely by the meteorological variability. We find that values also show increasing trends, but to a less degree, which are up to about 0.08, 0.13, 0.10, and 0.01 ppbv yr$^{-1}$ in East Asia, China, Korea, and Japan, respectively. Compared with the trends from the $E_{yy}M_{yy}$ simulation as discussed above, these results indicate that the interannual meteorological variability accounts for 30% of total O$_3$ increases in East Asia.

To understand the impact of interannual meteorological variability on spatial distribution of surface O$_3$ and correlations with meteorological variables, we compare the averages of two 5-yr periods, 1985–1989 and 2002–2006. We consider that the 5-yr averaged concentrations are representative values devoid of the noise due to short-term meteorological variability. Fig. 6 shows the spatial distribution of the simulated springtime O$_3$ concentrations for 1985–1989 and 2002–2006, respectively, from the $E_{06}M_{yy}$ simulation. High O$_3$ concentrations in eastern China reflect the primary source region of anthropogenic emissions and extend over northeast Asia indicating pollution transport from the continent. Differences of O$_3$ concentrations between two periods are also shown in Fig. 6. They are significantly positive, with the continental average of 1.3 ppbv. In particular, the largest increases are found over eastern China up to 5 ppbv. But O$_3$ is slightly decreased in northeastern Japan mainly due to ENSO events in 2002–2003 over the western Pacific Ocean. As mentioned above, the lower springtime O$_3$ over northeastern Japan is related to meteorological variations caused by El Niño.

In order to find out significant meteorological factors contributing to O$_3$ changes during past two decades, the spatial distributions of differences in meteorological fields and statistics between surface O$_3$ and meteorological fields over East Asia are shown in Fig. 6. A few previous studies suggest that O$_3$ is positively correlated with high temperature and solar radiation, as these enhance the photochemical O$_3$ production ([Wise and Comrie, 2005; Steiner et al., 2006; Dawson et al., 2007]). As previously found, O$_3$ concentrations in our statistical results are positively correlated with temperature in East Asia ($R = 0.48$), but correlation coefficient is relatively low. Previous studies showed that the probability of observed high O$_3$ exceeding the 80 ppb can double for a few degrees increases in temperature, which in case of high temperature as over 300 K ([Lin et al., 2001]). However, the average temperature in springtime in East Asia is 290 K and so resulting correlation between temperature and O$_3$ is relatively weaker than previous studies. The spatial pattern of O$_3$ changes corresponds to that of cloud fractions (Fig. 6) as shown in eastern China. Our results indicate that the changes of O$_3$ concentrations are negatively correlated with changes in cloudiness ($R = -0.61$). The decreases in large scale precipitation ($R = -0.49$) and relative humidity ($R = -0.71$) are also associated with cloudiness, and thus they are negatively correlated with surface O$_3$. These results suggest that the changes of O$_3$ in East Asia are affected by increased solar radiation due to decreased cloud cover and increased temperature over the past two decades.

5. Change of SO$_4^{2-}$–NO$_3^-$–NH$_4^+$ concentrations in East Asia

In this section, we investigate SO$_4^{2-}$–NO$_3^-$–NH$_4^+$ aerosols that are primary air pollutants in East Asia. Unlike O$_3$, those inorganic aerosol concentrations show both summer and winter maxima depending on chemical species. As described above in Section 3, SO$_4^{2-}$ concentrations were the largest in summer and the smallest in winter. Whereas, NO$_3^-$ concentrations display an opposite seasonal variation with a winter maximum and a summer minimum. Therefore, we focus our results on both summer for ammonium sulfate ([NH$_4$]$_2$SO$_4$) and winter for ammonium nitrate (NH$_4$NO$_3$) in East Asia.

Fig. 7 shows simulated summertime surface (NH$_4$)$_2$SO$_4$ concentrations from the $E_{06}M_{yy}$ for 1985–1989 and 2002–2006, respectively. High (NH$_4$)$_2$SO$_4$ concentrations in eastern China are mainly due to the anthropogenic emissions ([Streets et al., 2003]) and extend northeastward including Korea and Japan. Differences of summertime (NH$_4$)$_2$SO$_4$ concentrations for the past two periods...
are also shown in the figure reflecting the effect of regional meteorological variability. Those effects are significantly negative, up to $-7 \, \mu g \, m^{-3}$ in central eastern China and extend to the northeastern China, but $(NH_4)_2SO_4$ concentrations are increased over southern and northern China and Korean peninsula.

We find that the spatial patterns of $(NH_4)_2SO_4$ changes are generally consistent with that of mixing depth and convective precipitation changes between the two periods. Changes in mixing depth have stronger effects on PM concentrations (Jacob and Winner, 2009). Dawson et al. (2007) find that mixing depth changes led to mixing and dilution effect, with PM$_{2.5}$ concentrations generally decreasing as mixing depth was increased. Our results clearly show that increases in mixing depth result in decreases of $(NH_4)_2SO_4$ aerosols in central eastern China. Whereas, decreases in convective precipitation lead to less washout losses of aerosols and thus enhance $(NH_4)_2SO_4$ aerosol concentrations in southern China and Korean peninsula. Increase in temperature is also known to enhance $SO_4^{2-}$ concentrations due to faster SO$_2$ oxidation (Chin et al., 2000) but shows small variability ($+0.2\%$) as well as opposite effects. Changes in humidity affect PM water content and hence the uptake of semi-volatile components, mainly NO$_3$ and organics but little sensitivity of $(NH_4)_2SO_4$ aerosols (Dawson et al., 2007). Decreases in cloudiness are generally known to enhance aerosol concentrations but the result in this study shows opposite effects with low statistical significance.

Fig. 8 shows simulated wintertime NH$_4$NO$_3$ aerosols in surface air from the E06Myy over East Asia. The spatial distribution of NH$_4$NO$_3$ in winter is quite similar to that of $(NH_4)_2SO_4$ in summer. Differences of wintertime NH$_4$NO$_3$ concentrations between two periods are shown in Fig. 8 with a mean increase of $0.3 \, \mu g \, m^{-3}$ over East Asia and the highest value is up to $4 \, \mu g \, m^{-3}$ in eastern China. This increase in aerosol concentrations can be explained by decreases in mixing depth that lead to less dilution of aerosols in the later period relative to the earlier. The strong negative correlation between wintertime aerosol concentrations and mixing
depth supports our finding but other meteorological variables have relatively weak correlations.

The changes of SO$_4^{2-}$, NO$_3^-$, and NH$_4^+$ concentrations over the past decades have been largely influenced by their precursor emission changes. Table 1 summarizes our simulated results. Over the past two decades, SO$_2$, NO$_x$, and NH$_3$ emissions are increased by 51%, 107% and 31%, respectively, in East Asia. But the changes of SO$_4^{2-}$, NO$_3^-$, and NH$_4^+$ aerosols show much higher increases than those of NO$_x$ and NH$_3$ emissions, but the SO$_4^{2-}$ aerosol increase is lower than that of SO$_2$ emission. This result indicates that the inorganic aerosol formation in East Asia is ammonia-poor in 1980s and becomes ammonia-rich in 2000s (Song et al., 2008). Interestingly, the increasing rates of wintertime SO$_4^{2-}$, NO$_3^-$, and NH$_4^+$ concentrations are higher than those of summertime, implying that meteorological conditions in winter become more favorable for the PM air quality degradation. This is also clearly shown in the change of SO$_4^{2-}$, NO$_3^-$, and NH$_4^+$ concentrations due to changes in meteorological variability (Table 1). The effects of meteorological variability between two periods on the seasonal mean concentrations are opposite in summer and winter such as a reduction of 4% in summer but an increase of 7% in winter and this is mainly due to the meteorological variability for each season over the past two decades.

6. Summary and discussions

Over the past two decades, meteorological changes have occurred in East Asia, but their impacts on regional air quality are difficult to understand from measurements alone. In this study, we have quantified the effects of the meteorological variability on O$_3$ and SO$_4^{2-}$, NO$_3^-$, and NH$_4^+$ concentrations in East Asia using the 3-D global chemical transport model (GEOS-Chem).

First, we conducted a model evaluation by comparing the observed and simulated O$_3$, and SO$_4^{2-}$, NO$_3^-$, and NH$_4^+$ concentrations at EANET sites. The simulated O$_3$ showed an overestimate of 10 ppbv in summer and 1–7 ppbv in other seasons, reflecting too coarse horizontal resolutions to reproduce observed O$_3$ that is significantly affected by local emissions of O$_3$ precursors. On the other hand, SO$_4^{2-}$ and NH$_4^+$ aerosols generally reproduced the observed spatial variability with $R^2$ of 0.5–0.8 and regression slopes of 0.6–1.0. However, NO$_3^-$ aerosol is relatively poorly simulated. Despite of these discrepancies, we found that the model generally captured the variation of seasonal mean concentrations averaged over East Asia.
Asia, which can be used to analyze the effects of meteorology on regional air quality over East Asia.

We then used the sensitivity simulations with the fixed anthropogenic emissions to examine the effects of the meteorological variability on the changes of O$_3$ and SO$_4^{2-}$/CO$_e$/NO$_3$/CO$_e$/NH$_4^+$ concentrations in East Asia. We found that springtime O$_3$ concentration has been generally increased over the past two decades mainly due to increases in anthropogenic precursor emissions, and also concurrent meteorological variability including decreases of cloud covers, precipitation, relative humidity and increases of temperature further enhanced O$_3$, accounting for 30% of total O$_3$ increases in East Asia. On the other hand, the effects of meteorological variability on SO$_4^{2-}$/NO$_3$/NH$_4^+$ aerosols have seasonal dependency such as negative in summer and positive in winter. Increases in mixing depth suppress (NH$_4$)$_2$SO$_4$ aerosol concentrations in summer up to 6 $\mu$g m$^{-3}$ in central eastern China, whereas, decreases in convective precipitation enhance (NH$_4$)$_2$SO$_4$ aerosol concentrations in southern China and Korean peninsula. Decreases in mixing depth in winter result in enhancement of NH$_4$NO$_3$ aerosol concentrations up to 4 $\mu$g m$^{-3}$ in eastern China but other meteorological variables have relatively weak correlations.

The changes of SO$_4^{2-}$/NO$_3$/NH$_4^+$ concentrations over the past two decades have been largely influenced by increases in anthropogenic precursor emissions but not linearly proportional to those of precursor emissions changes. NO$_x$ and NH$_x$ aerosols were increased much higher than those of NO$_x$ and NH$_x$ emissions, but SO$_4^{2-}$ aerosol increase was lower than that of SO$_2$ emission change. These results corresponded well with decreases of (NH$_4$)$_2$SO$_4$ in summer and increases of NH$_4$NO$_3$ in winter from sensitivity simulation with fixed anthropogenic emissions and implied that meteorological conditions in winter have become more favorable for the PM air quality degradation over the past two decades.

<table>
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<td>SO$_2$ (+51%)</td>
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<td>SUM (+63%)</td>
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Fig. 8. Same as Fig. 6 except for NH$_4$NO$_3$ concentrations (μg m$^{-3}$) and meteorological variables in winter.
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