Widespread air pollutants of the North China Plain during the Asian summer monsoon season: A case study

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Abstract: During the Asian summer monsoon season, prevailing southeasterly - southwesterly winds are subject to delivering air pollutants from the North China Plain (NCP) to the Northeast and Northwest China. In the present study, the WRF-CHEM model is used to evaluate contributions of trans-boundary transport of the NCP emissions to the air quality in the Northeast and Northwest China during a persistent air pollution episode from 22 to 28 May 2015. The WRF-CHEM model generally performs well in capturing the observed temporal variation and spatial distribution of fine particulate matters (PM$_{2.5}$), ozone (O$_3$), and NO$_2$. The simulated temporal variation of aerosol species is also in good agreement with measurements in Beijing during the episode. Model simulations show that the NCP emissions contribute substantially to the PM$_{2.5}$ level in Liaoning and Shanxi provinces, the adjacent downwind areas of the NCP, with an average of 24.2 and 13.9 µg m$^{-3}$ during the episode, respectively. The PM$_{2.5}$ contributions in Jilin and Shaanxi provinces are also appreciable, with an average of 9.6 and 6.5 µg m$^{-3}$, respectively. The average percentage contributions of the NCP emissions to the PM$_{2.5}$ level in Liaoning, Jilin, Shanxi, Shaanxi provinces are 40.6%, 27.5%, 32.2%, and 20.9%, respectively. The NCP emissions contribute remarkably to the O$_3$ level in Liaoning province, with an average of 46.5 µg m$^{-3}$, varying from 23.9 to 69.5 µg m$^{-3}$. The O$_3$ level in Shanxi province is also influenced considerably by the NCP emissions, with an average contribution of 35.1 µg m$^{-3}$. The O$_3$ level in Shanxi province is also influenced considerably by the NCP emissions, with an average contribution of 35.1 µg m$^{-3}$. The average O$_3$ contributions of the NCP emissions to Jilin and Shaanxi provinces are 28.7 and 20.7 µg m$^{-3}$, respectively. The average percentage contributions of the NCP emissions to the afternoon O$_3$ level in Liaoning, Jilin, Shanxi, and Shaanxi provinces are 27.4%, 19.5%, 21.2%, and 15.8%, respectively. However, the effect of the NCP emissions on the air quality in Inner Mongolia is generally insignificant. Therefore, effective mitigations of the NCP emissions not only improve the local air quality, but also are beneficial to the air quality in the Northeast and Northwest China during the Asian summer monsoon season.
1 Introduction

With the rapid growth of industrialization, urbanization and transportation, China has experienced severe air pollution with high levels of fine particulate matters (PM\(_{2.5}\)) and ozone (O\(_3\)) recently (e.g., Chan and Yao, 2008; Zhang et al., 2013; Kurokawa et al., 2013; Li et al., 2017b). Although the Chinese State Council has issued the ‘Atmospheric Pollution Prevention and Control Action Plan’ in September 2013 with the aim of improving China’s air quality, heavy haze or photochemical smog still frequently plagues China, especially in the North China Plain (NCP), Yangtze River Delta (YRD), and Pearl River Delta (PRD). Elevated O\(_3\) and PM\(_{2.5}\) concentrations in the atmosphere not only perturb regional and global climates, also exert adverse effects on air quality, ecosystems, and human health (Weinhold, 2008; Parrish and Zhu, 2009).

The NCP has become one of the most polluted areas in the world due to a large amount of emissions of pollutants and frequent occurrence of unfavorable meteorological situations, as well as the topography (e.g., Tang et al., 2012; Zhang et al., 2013; Zhuang et al., 2014; Pu et al., 2015; Long et al., 2016). Heavy haze with extremely high PM\(_{2.5}\) concentrations often covers the NCP during wintertime, partially attributable to the coal combustion for domestic heating (e.g., He et al., 2001; Cao et al., 2007; H. Li et al., 2017a). However, even in summer, with improvement of the evacuation condition and increase of precipitation, photochemical smog with high levels of PM\(_{2.5}\) and O\(_3\) still engulfs the NCP (e.g., Gao et al., 2011; Hu et al., 2014; Cao et al., 2015; Wu et al., 2017). The PM\(_{2.5}\) concentrations during summertime in the NCP are generally lower than those in winter, but still much higher than 35 \(\mu\)g m\(^{-3}\), the first grade of National Ambient Air Quality Standards (NAAQS) in China (Feng et al., 2016; Z. S. Wang et al., 2016; Sun et al., 2016). The average summertime PM\(_{2.5}\) concentrations in the NCP are 77.0 ± 41.9 \(\mu\)g m\(^{-3}\) in 2013, much more than those in other regions of China and also exceeding the second grade of NAAQS (Hu et al., 2014). In addition, increasing O\(_3\)
precursors emissions has caused serious O₃ pollution during summertime in the NCP (e.g., Zhang et al., 2009; Xu et al., 2011; Kurokawa et al., 2013). Li et al. (2017b) have reported that the maximum 1h O₃ concentration exceeds 200 µg m⁻³ in almost all the cities in Eastern China from April to September 2015, mainly concentrated in the NCP and YRD, showing a widespread and persistent O₃ pollution. Ma et al. (2016) have found a growth trend of the surface O₃ at a rural site in the NCP from 2003 to 2015, with an average rate of 1.13 ppb per year. Wu et al. (2017) have shown that the average afternoon O₃ concentration in the summer of 2015 in Beijing is about 163 µg m⁻³.

China is located in a large monsoon domain, and the Asia summer monsoon (ASM) tends to substantially influence the distribution and trans-boundary transport of air pollutants in China. Zhu et al. (2004) have proposed that the summertime high O₃ concentration over Western China is due to the monsoonal transport from Eastern China and long-range transport from South/central Asia and even Europe. Zhao et al. (2010) have also indicated that O₃ transported from South/Central Asia to Western China increases from May to August because of the northward movement of the India summer monsoon. Huang et al. (2015) have suggested that an earlier onset of the ASM would lead to more O₃ enhancement in the lower troposphere over the NCP in later spring and early summer. Numerous studies have also reported that the strength and tempo-spatial extension of the ASM influences the spatial and temporal distribution of aerosol mass concentrations over Eastern China (Cao et al., 2015; Li et al., 2016; Cheng et al., 2016). For example, Zhang et al. (2010) have emphasized that the East ASM plays a major role in determining the seasonal and interannual variations of the PM₂.₅ concentration over Eastern China. Using the GEOS-CHEM model, Zhu et al. (2012) have shown that the weakening of the ASM increases the aerosol concentration in Eastern China. Wu et al. (2016) have pointed out that the regional transport and tempo-spatial distribution of air pollutants are directly influenced by the East Asian monsoon at seasonal,
inter-annual, and decadal scales. During the ASM season, meteorological conditions are characterized by prevailing southwesterly-southeasterly winds over Eastern China. Air pollutants originated from the NCP are likely to be transported northwards and affect the air quality in its downwind areas, so it is imperative to quantitatively evaluate the effect of the NCP emissions on the air quality in its neighboring regions. Previous studies have concentrated on the composition, characteristics, and sources of the air pollutants over the NCP (e.g., Han et al., 2006; Liu et al., 2012; Zhao et al., 2013; Li et al., 2015). However, few studies have been performed to investigate the effect of trans-boundary transport of air pollutants originated from the NCP on the air quality in the Northeast and Northwest China under the prevailing southerly wind associated with the ASM.

In this study, we first analyze the role of synoptic situations during the ASM (from May to September) in the trans-boundary transport over Northern China and further evaluate the contribution of trans-boundary transport of pollutants originated from the NCP to the air quality in the Northeast and Northwest China using the WRF-CHEM model. The model configuration and methodology are described in Section 2. Analysis results and discussions are presented in Section 3, and conclusions are given in Section 4.

2 Model and Methodology

2.1 WRF-CHEM Model and Configuration

A persistent air pollution episode with high levels of PM$_{2.5}$ and O$_3$ from 22 to 28 May 2015 in Northern China is simulated using the WRF-CHEM model which is developed by Li et al. (2010, 2011a, b, 2012) at the Molina Center for Energy and the Environment. Table 1 provides detailed model configurations and Figure 1 shows the WRF-CHEM model simulation domain. It is worth noting that the horizontal resolution of 10 km adopted in this
study is the lower bound for the WRF model to turn on the cumulus scheme, so the new Kain-Fritch scheme is used in the present study (Table 1). Further description of the model is presented in Supplementary Information (SI).

The key characteristics of the aerosol pollution in China are frequently associated with rather efficient secondary formation, including aerosol nucleation and rapid growth under favorable conditions (Zhang et al., 2012; Qiu et al., 2013; Guo et al., 2014; Zhang et al., 2015). The new particle production rate in the WRF-CHEM model is calculated due to the binary nucleation of H$_2$SO$_4$ and H$_2$O vapor. The nucleation rate is a parameterized function of temperature, relative humidity, and the vapor-phase H$_2$SO$_4$ concentration, following the work of Kulmala et al. (1998), and the new particles are assumed to be 2.0 nm diameter. Recent studies have shown that organic vapors are involved in the nucleation process (Zhang et al., 2012) and further studies need to be conducted to consider the contributions of organic vapors to the nucleation process. The secondary organic aerosol (SOA) formation is simulated using a non-traditional SOA model including the volatility basis-set modeling method in which primary organic components are assumed to be semi-volatile and photochemically reactive and are distributed in logarithmically spaced volatility bins (Li et al., 2011a). The contributions of glyoxal and methylglyoxal to the SOA formation are also included in the SOA module. The SOA formation from glyoxal and methylglyoxal is parameterized as a first-order irreversible uptake by aerosol particles, with a reactive uptake coefficient of $3.7 \times 10^{-3}$ for glyoxal and methylglyoxal (Zhao et al., 2006). The simulation of inorganic aerosols in the WRF-CHEM model adopts the ISORROPIA Version 1.7 (Nenes et al., 1998).

For the discussion convenience, Northern China is divided into 3 regions (Figure S1): 1) the North China Plain (including Beijing, Tianjin, Hebei, Shandong, Henan, the south of Jiangsu and Anhui, hereafter referred to as the NCP), 2) the Northeast China (including
Heilongjiang, Jilin, Liaoning and the east part of Inner Mongolia, hereafter referred to as the 140 NEC), and 3) the Northwest China (including Shanxi, Shaanxi and the west part of Inner 141 Mongolia, hereafter referred to as the NWC). During the episode, the observed average daily 142 PM$_{2.5}$ concentration was 75.5 µg m$^{-3}$ and the mean O$_3$ concentration in the afternoon was 143 151.2 µg m$^{-3}$ in the NCP. Figure S2 presents the distributions of the anthropogenic emission 144 rates of volatile organic compounds (VOCs), nitrogen oxide (NO$_x$), organic carbon (OC), and 145 SO$_2$ in Mainland China, showing that the high emission rates of VOCs, NO$_x$, OC, and SO$_2$ 146 are generally concentrated in the NCP. It is worth noting that uncertainties in the emission 147 inventory used in this study are rather large considering the rapid changes in anthropogenic 148 emissions that are not fully reflected in the current emission inventory and the complexity of 149 pollutants precursors.

2.2 Data and Methodology

In the present study, the model performance is validated using the hourly measurements 152 of O$_3$, NO$_2$, and PM$_{2.5}$ concentrations released by the China’s Ministry of Environment 153 Protection (China MEP), which can be accessed at http://www.aqistudy.cn/. In addition, the 154 simulated submicron sulfate, nitrate, ammonium, and organic aerosols are compared to the 155 measurements by the Aerodyne Aerosol Chemical Speciation Monitor (ACSM), which was 156 deployed at the National Center for Nanoscience and Technology (NCNST), Chinese 157 Academy of Sciences in Beijing (Figure 1). The observed mass spectra of organic aerosols 158 are analyzed using the Positive Matrix Factorization (PMF) technique and four components 159 are identified: hydrocarbon-like organic aerosol (HOA), cooking organic aerosol (COA), coal 160 combustion organic aerosol (CCOA), and oxygenated organic aerosol (OOA). HOA, COA, 161 and CCOA are interpreted as a surrogate of primary organic aerosols (POA), and OOA is a 162 surrogate of secondary organic aerosols (SOA). Furthermore, the reanalysis data from the
European Centre for Medium-Range Weather Forecasts (ECMWF) are used to analyze the synoptic patterns during the ASM season from May to September 2015.

The mean bias (MB), root mean square error (RMSE) and the index of agreement (IOA) are utilized to evaluate the performance of the WRF-CHEM model simulations against measurements. To assess the contributions of the NCP emissions to the near-surface concentrations of O$_3$ and PM$_{2.5}$ in the NEC and NWC, the factor separation approach (FSA) is used in this study (Stein and Alpert, 1993; Gabusi et al., 2008; Li et al., 2014). The detailed description of methodology can be found in SI-2.

3 Results and Discussion

3.1 Synoptic Patterns during the ASM Season

The ASM commences to prevail from May to September each year in China, with strong winds blowing from oceans to Eastern China and bringing warm and moist airflow to the continent. Furthermore, the Western Pacific subtropical high gradually intensifies, and moves from south to north to influence the weather and climate over China, also transporting water vapor from the sea to Eastern China. During the ASM season, due to the influence of the Western Pacific subtropical high, rain belts and associated deep convections move from Southeastern China to Northern China (Ding et al., 1992, 2005; Lau et al., 1988, 1992; Kang et al., 2002). Figure 2 shows the geopotential heights at 500 hPa and mean sea level pressure with wind vectors during the ASM season in 2015. At 500 hPa, the main part of subtropical high, which is represented by the scope of the contour of 5880 geopotential meter, is located in Northwest Pacific Ocean. The mean ridgeline of the Western Pacific subtropical high is located at 25°N, moving from south to north from May to September, which substantially affects the synoptic conditions in China. Flat westerly wind at 500 hPa prevails over the NCP and its surrounding regions, indicating a stable weather condition. The mean sea level
pressure shows that most of areas in the NCP are continually influenced by the ASM and the high-pressure system centering in the Western Pacific, causing the prevailing southeasterly -southwesterly wind over the NCP and its surrounding areas. The detailed description of the synoptic conditions during the study episode can be found in SI.

In the region controlled by the Western Pacific subtropical high, a subsidence airflow is dominant with calm or weak winds, and the temperature is extremely high due to the strong sunlight, which is favorable for the accumulation and formation of air pollutants. The air pollutants are likely to be transported from south to north under the persistent effect of southerly winds.

Figures 3 and 4 present the relationship of PM$_{2.5}$ and O$_3$ concentrations in the NCP with those in the NEC and NWC during the ASM season from 2013 to 2016, respectively. The observed PM$_{2.5}$ and O$_3$ concentrations in the NCP exhibit a positive correlation with those in the NEC and NWC, with the correlation coefficients generally exceeding 0.55. There are two possible reasons for the positive correlation of PM$_{2.5}$ and O$_3$ concentrations between the NCP and its surrounding regions. One is that when the NCP and its neighboring areas are controlled by the same large-scale synoptic pattern, the concentrations of air pollutants generally vary synchronously. The other is the trans-boundary transport of air pollutants originated from the NCP to its surrounding areas due to the southerly wind associated with the ASM. The correlation coefficients of PM$_{2.5}$ and O$_3$ concentrations in the provinces of the NEC with those in the NCP generally decrease from south to north, with the coefficients of 0.69, 0.56 and 0.52 for PM$_{2.5}$, and of 0.86, 0.76, and 0.76 for O$_3$ in Liaoning, Jilin and Heilongjiang, respectively. The decreasing trend of the correlation coefficients also exists from east to west in the NWC, with coefficients of 0.69 and 0.62 for PM$_{2.5}$, and 0.87 and 0.84 for O$_3$ in Shanxi and Shaanxi, respectively. Hence, when severe air pollution occurs in the NCP in summer, the air quality in its adjacent provinces is likely to be deteriorated, possibly
caused by the trans-boundary transport of air pollutants originated from the NCP.

It is worth noting that the intensity of ASM substantially influences the temporal variation and spatial distribution of air pollutants (Wu et al., 2016). The East Asia summer monsoon index proposed by Zhang et al. (2003) is defined as a difference of anomalous zonal wind between the (10°-20°N, 100°-150°E) and (25°-35°N, 100°-150°E) at 850hPa during summer (June-August). The year of monsoon index greater than or equal to 2 is defined as the strong summer monsoon year, and the year of monsoon index less than or equal to -2 is defined as the weak summer monsoon year. The monsoon index calculated by China Meteorological Administration shows that the intensity of the summer monsoon in 2015 is close to the normals (SI-Figure S5).

3.2 Model performance

3.2.1 PM$_{2.5}$, O$_3$ and NO$_2$ Simulations in Northern China

Figure 5 shows the temporal variations of observed and simulated near-surface PM$_{2.5}$, O$_3$ and NO$_2$ concentrations averaged over monitoring sites in Northern China. The WRF-CHEM model generally simulates well the diurnal variation of PM$_{2.5}$ concentrations in Northern China, with $IOA$ of 0.91. The model successfully reproduces the temporal variations of surface O$_3$ concentrations compared with observations in Northern China, e.g., peak O$_3$ concentrations in the afternoon due to active photochemistry and low O$_3$ concentrations during nighttime caused by the NO$_x$ titration, with $IOA$ of 0.98. However, the model underestimation still exists in simulating the O$_3$ concentration, with a $MB$ of -4.0 µg m$^{-3}$. The model also reasonably yields the NO$_2$ diurnal profiles, but frequently overestimates the NO$_2$ concentrations in the late evening due to the simulated low PBL height, and underestimates the concentration in the early morning because of the uncertainties in the NO$_x$ emissions. The further analysis of the model performance of PM$_{2.5}$, O$_3$ and NO$_2$ concentrations in Northern China can be found in SI.
3.2.2 Aerosol Species Simulations in Beijing

Figure 6 presents the temporal variations of simulated and observed aerosol species at NCNST site in Beijing from 22 to 28 May 2015. Generally, the WRF-CHEM model predicts reasonably the temporal variations of the aerosol species against the measurements, especially for POA and nitrate aerosol, with IOAs of 0.81 and 0.90, respectively. The model has difficulties in well simulating the SOA concentrations, with the IOA and MB of 0.69 and -3.6 µg m\(^{-3}\), respectively. It is worth noting that many factors influence the SOA simulation, including measurements, meteorology, precursors emissions, SOA formation mechanisms and treatments (Bei et al., 2012, 2013). The model reasonably tracks the temporal variation of the observed sulfate concentration, but the bias is still large, and the MB and IOA are -1.2 µg m\(^{-3}\) and 0.68, respectively. The sulfate source in the atmosphere is various, including SO\(_2\) gas-phase oxidations by hydroxyl radicals (OH) and stabilized criegee intermediates (sCI), aqueous reactions in cloud or fog droplets, and heterogeneous reactions on aerosol surfaces, as well as direct emissions from power plants and industries (G. H. Li et al., 2017a). G. Wang et al. (2016) have also reported that the aqueous oxidation of SO\(_2\) by NO\(_2\) is important to the efficient sulfate formation. Considering that the model fails to well resolve convective clouds due to the 10km horizontal resolution, the sulfate formation from the cloud process is generally underestimated. Additionally, large amount of SO\(_2\) is emitted from point sources, such as power plants or agglomerated industrial zones, which is much more sensitive to wind fields simulations (Bei et al., 2010). The model performs reasonably well in simulating the ammonium aerosol, with the IOA and MB of 0.77 and -0.4 µg m\(^{-3}\), respectively.

3.2.3 Simulations of the Spatial Distribution of PM\(_{2.5}\) and O\(_3\) Concentrations

The peak PM\(_{2.5}\) concentration generally occurs from 08:00 to 10:00 Beijing Time (BJT) due to the rush hour. Figure 7 provides the distributions of calculated and observed near-surface PM\(_{2.5}\) concentrations along with the simulated wind fields at 08:00 BJT from 23
to 28 May 2015. The calculated PM$_{2.5}$ spatial patterns generally agree well with the observations at the monitoring sites. The NCP experiences severer PM$_{2.5}$ pollution than its surrounding areas, with PM$_{2.5}$ concentrations frequently exceeding 150 µg m$^{-3}$ in the Beijing-Tianjin-Hebei region. During the study episodes, the pollutants are likely to be transported to the NEC and NWC under the prevailing southwesterly or southeasterly winds in Northern China, causing the PM$_{2.5}$ concentrations in most of areas of the NEC and NWC frequently to be higher than 75 µg m$^{-3}$.

The O$_3$ concentration during summertime generally reaches its peak from 14:00 to 16:00 BJT in Northern China (Figure 5). Figure 8 shows the spatial distribution of calculated and measured near-surface O$_3$ concentrations at 14:00 BJT from 23 to 28 May 2015, along with the simulated wind fields. Generally, the simulated O$_3$ spatial patterns are consistent with the observations, but the model overestimation or underestimation still exists. The simulated high O$_3$ concentrations at 14:00 BJT, exceeding 200 µg m$^{-3}$, are frequently concentrated in the NCP, which is also consistent with the measurements. The O$_3$ transport to the NEC and NWC from the NCP is obvious when the winds are southeasterly or southwesterly, inducing the severe O$_3$ pollution in the NEC and NWC.

In general, the simulated variations of PM$_{2.5}$, O$_3$, NO$_2$ and aerosol species are in good agreement with observations, indicating that the simulations of meteorological conditions, chemical processes and the emission inventory used in the WRF-CHEM model are reasonable, providing a reliable base for the further investigation.

### 3.3 Effects of the NCP Emissions on the Air Quality in the NEC and NWC

To evaluate the contribution of the NCP emissions to the air quality in its neighboring areas, four model simulations are performed, including $f_{NS}$ with all anthropogenic emissions from the NCP and non-NCP areas, $f_{N}$ with anthropogenic emissions from the NCP only, $f_{S}$ with anthropogenic emissions from the non-NCP areas only, and $f_{0}$ without
all anthropogenic emissions. Consequently, the air pollutants concentrations in the NEC and NWC can be separated into four components, including contributions from the local emissions \((f'_S, f_S - f_0)\), the trans-boundary transport of the NCP emissions \((f'_N, f_N - f_0)\), the interactions of these two emissions \((f'_{NS}, f_{NS} - f_N - f_S + f_0)\) and the background \((f_0)\).

In the present study, the effect of the NCP emissions on the PM\(_{2.5}\) and O\(_3\) concentrations in the NEC and NWC is evaluated, considering that they have the long lifetime of several days in the troposphere and often constitute the primary air pollutant during summertime (Seinfeld and Pandis, 2006). However, the trans-boundary transport of PM\(_{10}\) is not considered due to its short lifetime of several hours caused by the dry deposition and gravity and the fact that PM\(_{10}\) is generally confined to its source region when the wind is not strong enough (Sun et al., 2006).

### 3.3.1 Contributions of the NCP Emissions to PM\(_{2.5}\) Levels in the NEC and NWC

Figure 9 shows the simulated spatial distribution of daily mean PM\(_{2.5}\) concentrations contributed by the NCP emissions in the NEC and NWC from 23 to 28 May 2015. The contribution of trans-boundary transport of the NCP emissions to the PM\(_{2.5}\) concentration is remarkable in Liaoning, frequently exceeding 30 µg m\(^{-3}\) in most areas of the province during the episode. The NCP emissions also considerably influence the PM\(_{2.5}\) concentration in Jilin, contributing 5~30 µg m\(^{-3}\) in most areas and occasionally exceeding 40 µg m\(^{-3}\). The effect of the NCP emissions on the PM\(_{2.5}\) level in Shanxi and Shaanxi is increasingly evident from 23 to 28 May 2015, with the contribution of up to 50~60 µg m\(^{-3}\) in southeast of Shanxi and to a lesser extent of 30~40 µg m\(^{-3}\) in the middle part of Shaanxi on 27-28 May. The contribution of trans-boundary transport of the NCP emissions to the PM\(_{2.5}\) level in Inner Mongolia is not significant, which may be attributed to the location of the low pressure and terrain characteristics. Obviously, the effect of trans-boundary transport shows a stepwise
characteristic: the closer to the NCP emission sources, the more remarkable the impact on the downwind areas. As a result, Liaoning and Shanxi provinces are substantially influenced by the NCP emissions, while Jilin and Shaanxi provinces are affected to a lesser extent.

The impact of the NCP emissions on the daily average PM$_{2.5}$ concentration in the NEC and NWC from 22 to 28 May 2015 is summarized in Table 2. On average, the NCP emissions increase the PM$_{2.5}$ concentrations by 24.2, 9.6, 13.9, 6.5, and 2.6 $\mu$g m$^{-3}$ in Liaoning, Jilin, Shanxi, Shaanxi, and Inner Mongolia, with the average percentage contribution of 40.6%, 27.5%, 32.2%, 20.9%, and 16.7%, respectively. Figure 10 shows the episode-averaged PM$_{2.5}$ percentage contribution from the NCP emissions to the surrounding areas. The NCP emissions markedly affect the air quality in Liaoning, accounting for around 20%-50% of the PM$_{2.5}$ concentration during the episode and with the most substantial impact on the west part of the province. The NCP emissions contribute about 15%-30% of the PM$_{2.5}$ concentration in Jilin. Shanxi province is also remarkably affected by the NCP emissions, with more than 25% of PM$_{2.5}$ concentration contributed by the NCP emissions in most areas. Although Shaanxi province is a little far from the NCP, the NCP emissions still contribute about 10%-35% of the PM$_{2.5}$ concentration. The NCP emissions also enhance the PM$_{2.5}$ concentration by 5-50% in the southern edge of Inner Mongolia, which is adjacent to the NCP.

3.3.2 Contributions of the NCP Emissions to O$_3$ Concentrations in the NEC and NWC

Figure 11 shows the simulated spatial distribution of the average afternoon O$_3$ concentrations contributed by the NCP emissions from 23 to 28 May 2015. Similar to the PM$_{2.5}$ case, the contribution of the NCP emissions to the O$_3$ formation in Liaoning and Jilin province is increasingly enhanced during the episode (except on 26 May), and on 25 and 27 May, the NCP emissions account for more than 70 $\mu$g m$^{-3}$ of the O$_3$ concentration in most areas of Liaoning. On 25 and 28 May, the NCP emissions contribute more than 70 $\mu$g m$^{-3}$ of the O$_3$ concentration in some regions of Jilin. A less impact of the NCP emissions on Jilin
province on 26 May is due to the weakening of the low pressure. The NCP emissions play a progressively important role in $O_3$ concentrations in Shanxi and Shaanxi provinces during the episode, especially on 27 and 28 May when the contribution can be up to 60 µg m$^{-3}$. The impact of the NCP emissions on $O_3$ concentrations in Inner Mongolia is insignificant overall.

Table 3 summarizes the effects of the NCP emissions on the average afternoon $O_3$ concentration in the NEC and NWC from 22 to 28 May 2015. During the episode, the NCP emissions substantially influence the $O_3$ level in Liaoning province, and the afternoon $O_3$ contribution is about 46.5 µg m$^{-3}$ on average, ranging from 23.9 to 69.5 µg m$^{-3}$. The NCP emissions also contribute an average of 28.7 µg m$^{-3}$ to the $O_3$ concentration in Jilin province, varying from 12.4 to 45.7 µg m$^{-3}$. The contribution of NCP emissions to Shanxi and Shaanxi provinces becomes increasingly significant during the episode, with an average of 35.1 µg m$^{-3}$ for Shanxi province and 20.7 µg m$^{-3}$ for Shaanxi province, respectively. The $O_3$ concentration in Inner Mongolia is less influenced by the NCP emissions, with an average of 8.4 µg m$^{-3}$. Figure 12 illustrates the episode-averaged afternoon $O_3$ percentage contribution of the NCP emissions to the surrounding areas. In the NEC, the NCP emissions account for 15-35% of the afternoon $O_3$ concentration in most areas of Liaoning province, and 10-30% in Jilin province. In the NWC, the NCP emissions contribute 10-35% of the $O_3$ concentration in Shanxi province, and 10-25% in Shaanxi. In Inner Mongolia, the impact of the NCP emissions on $O_3$ formation is small, generally less than 15% except in the southern area adjacent to the NCP and Liaoning province where a contribution of more than 10% is found. On average, the NCP emissions distinctly increase the afternoon $O_3$ concentrations in Liaoning, Jilin, Shanxi, Shaanxi, and Inner Mongolia, with the average percentage of 27.4%, 19.5%, 21.2%, 15.8%, and 8.0%, respectively (Table 3).

Additional sensitivity studies have also been performed to examine the potential influences of the cumulus parameterization on evaluation of the contribution of the NCP emissions.
emissions to the PM$_{2.5}$ and O$_3$ concentrations in the NEC and NWC, in which the cumulus parameterization is turned off. The difference of the contribution of NCP emissions to the PM$_{2.5}$ and O$_3$ concentrations in the NEC and NWC is less than 0.8% between the simulations with and without the cumulus parameterization. Furthermore, it is worth noting that uncertainties from meteorological field simulations, emission inventories, and the chemical mechanism used in simulations, have large potentials to influence evaluation of the effect of the NCP emissions on the PM$_{2.5}$ and O$_3$ concentrations in the NEC and NWC (Carter and Atkinson, 1996; Lei et al., 2004; Song et al., 2009; Bei et al., 2017).

4 Summary and Conclusions

Analyses of the synoptic pattern during the ASM season show that the southeasterly-southwesterly winds prevail in Northern China, facilitating the trans-boundary transport of air pollutants from the NCP to the NEC and NWC. The good relationships of PM$_{2.5}$ and O$_3$ concentrations in the NCP with those in the NEC and NWC during the ASM season also indicate the possibility that the air quality in the NEC and NWC is influenced by the trans-boundary transport of air pollutants originated from the NCP.

A widespread and severe pollution episode from 22 to 28 May 2015 in Northern China is further simulated using the WRF-CHEM model to investigate the impact of trans-boundary transport of the NCP emissions on PM$_{2.5}$ and O$_3$ concentrations in the NEC and NWC, when the region is affected by prevailing southeasterly-southwesterly winds associated with the ASM.

In general, the WRF-CHEM model well reproduces the temporal variations and spatial distributions of PM$_{2.5}$, O$_3$, and NO$_2$ concentrations compared to observations in Northern China, although the model biases still exist due to the uncertainties in simulated meteorological fields and the emission inventory. The model also performs reasonably well in
simulating the variations of aerosol constituents against the ACSM measurement at the NCNST site in Beijing.

The FSA method is used to investigate the contribution of trans-boundary transport of the NCP emissions to O3 and PM$_{2.5}$ levels in the NEC and NWC. Model results show that the NCP emissions contribute approximately an average of 24.2 and 13.9 µg m$^{-3}$ to the PM$_{2.5}$ concentration in Liaoning and Shanxi during the episode, with the average percentage contribution of 40.6% and 32.2%, respectively. The NCP emissions enhance the PM$_{2.5}$ level by 9.6 and 6.5 µg m$^{-3}$ in Jilin and Shaanxi on average, with the percentage contribution of 27.5% and 20.9%, respectively. The NCP emissions also substantially influence the O$_3$ concentration in the NEC and NWC. The NCP emissions increase the afternoon (12:00 - 18:00 BJT) O$_3$ concentration in Liaoning by 46.5 µg m$^{-3}$ on average during the episode, followed by 35.1 µg m$^{-3}$ in Shanxi, 28.7 µg m$^{-3}$ in Jilin, and 20.7 µg m$^{-3}$ in Shaanxi, with the average percentage contribution of 27.4%, 21.2%, 19.5%, and 15.8%, respectively. In contrast, the contribution of trans-boundary transport of the NCP emissions to the PM$_{2.5}$ and O$_3$ concentration in Inner Mongolia are less, with an average of 2.6 and 8.4 µg m$^{-3}$, respectively. Our results demonstrate that when southerly winds are prevailing in Northern China, air pollutants originated from the NCP are likely to be transported northwards and profoundly affect the air quality in the NEC and NWC. Stringent control of the NCP emissions not only mitigates the local air pollution, also is beneficial to the air quality in the NEC and NWC during the ASM season.

It is worth noting that interactions between the air pollution in China and ASM are two-way and their relationships are complicated and interrelated, especially with regard to the aerosol-meteorology interaction. Aerosol impacts on meteorology is significant due to its direct and indirect effects, which further influence the air pollution condition in the lower troposphere. Aerosol semi-direct effect induced by the light absorbing aerosols in the
atmosphere stabilizes planetary boundary layer (PBL) and thus reduces the PBL height to exacerbate accumulation of air pollutants within the PBL, particularly for the aging process of black carbon which considerably enhances light absorption (Wang et al., 2013; Khalizov et al., 2009; Peng et al., 2016). In addition, aerosol plays an important role in the process of cloud formation and precipitation via acting as cloud condensation nuclei (CCN) and ice nuclei (IC). Therefore, aerosol-cloud interactions modify temperature and moisture profiles and influence precipitation, leading to potential feedback on the atmospheric chemistry (Wang et al., 2011). In addition, the ASM substantially influence spatial characteristics of the air pollutants transport and distribution in Eastern China on seasonal, inter-annual, and decadal scales (Wu et al., 2016). Further studies need to be performed to investigate the impacts of the ASM variation on the air pollutants transport, which is modulated by climate changes.

Although the model performs well in simulating PM$_{2.5}$, O$_3$ and NO$_2$ during the episode in northern China, the uncertainties from meteorological fields and the emission inventory still exist. Future studies need to be conducted to improve the WRF-CHEM model simulations, and to further assess the contributions of trans-boundary transport of the NCP emissions under specific synoptic patterns, considering the rapid changes in anthropogenic emissions, which is not reflected in the present study. Therefore, more episode simulations during the ASM season should be performed to comprehensively evaluate the contribution of trans-boundary transport of the NCP emissions to the air quality in its downwind regions and support the design and implementation of effective emission control strategies.
Acknowledgements. This work is financially supported by the National Key R&D Plan (Quantitative Relationship and Regulation Principle between Regional Oxidation Capacity of Atmospheric and Air Quality (2017YFC0210000)). Naifang Bei is supported by the National Natural Science Foundation of China (no. 41275101 and no. 41430424) and the Fundamental Research Funds for the Central Universities of China. Guohui Li is supported by the Hundred Talents Program of the Chinese Academy of Sciences and the National Natural Science Foundation of China (no. 41661144020).
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Contributions of trans-boundary transport to summertime air quality in Beijing, China, Atmospheric Chemistry and Physics, 17, 1-46, 2017.


<table>
<thead>
<tr>
<th><strong>Regions</strong></th>
<th>Northern China</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Simulation period</strong></td>
<td>May 22 to 28, 2015</td>
</tr>
<tr>
<td><strong>Domain size</strong></td>
<td>$350 \times 350$</td>
</tr>
<tr>
<td><strong>Domain center</strong></td>
<td>$35^\circ$N, $114^\circ$E</td>
</tr>
<tr>
<td><strong>Horizontal resolution</strong></td>
<td>$10\text{km} \times 10\text{km}$</td>
</tr>
<tr>
<td><strong>Vertical resolution</strong></td>
<td>35 vertical levels with a stretched vertical grid with spacing ranging from 30 m near the surface, to 500 m at 2.5 km and 1 km above 14 km</td>
</tr>
<tr>
<td><strong>Microphysics scheme</strong></td>
<td>WSM 6-class graupel scheme (Hong and Lim, 2006)</td>
</tr>
<tr>
<td><strong>Boundary layer scheme</strong></td>
<td>MYJ TKE scheme (Janjić, 2002)</td>
</tr>
<tr>
<td><strong>Surface layer scheme</strong></td>
<td>MYJ surface scheme (Janjić, 2002)</td>
</tr>
<tr>
<td><strong>Cumulus scheme</strong></td>
<td>Kain-Fritsch (new Eta) scheme (Kain, 2004)</td>
</tr>
<tr>
<td><strong>Land-surface scheme</strong></td>
<td>Unified Noah land-surface model (Chen and Dudhia, 2001)</td>
</tr>
<tr>
<td><strong>Longwave radiation scheme</strong></td>
<td>Goddard longwave scheme (Chou and Suarez, 2001)</td>
</tr>
<tr>
<td><strong>Shortwave radiation scheme</strong></td>
<td>Goddard shortwave scheme (Chou and Suarez, 1999)</td>
</tr>
<tr>
<td><strong>Meteorological boundary and initial conditions</strong></td>
<td>NCEP 1°×1° reanalysis data</td>
</tr>
<tr>
<td><strong>Chemical initial and boundary conditions</strong></td>
<td>MOZART 6-hour output (Horowitz et al., 2003)</td>
</tr>
<tr>
<td><strong>Anthropogenic emission inventory</strong></td>
<td>SAPRC-99 chemical mechanism emissions (Zhang et al., 2009)</td>
</tr>
<tr>
<td><strong>Biogenic emission inventory</strong></td>
<td>MEGAN model developed by Guenther et al. (2006)</td>
</tr>
<tr>
<td><strong>Model spin-up time</strong></td>
<td>28 hours</td>
</tr>
</tbody>
</table>
Table 2 Daily average PM$_{2.5}$ contributions (µg m$^{-3}$) of the NCP emissions in the NEC and NWC from 22 to 28 May 2015.

<table>
<thead>
<tr>
<th>Date</th>
<th>Jilin (µg m$^{-3}$)</th>
<th>Liaoning (µg m$^{-3}$)</th>
<th>Shanxi (µg m$^{-3}$)</th>
<th>Shaanxi (µg m$^{-3}$)</th>
<th>Inner Mongolia (µg m$^{-3}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>22</td>
<td>0.7±0.4</td>
<td>6.1±4.5</td>
<td>0.7±1.1</td>
<td>0.1±0.0</td>
<td>0.2±0.1</td>
</tr>
<tr>
<td>23</td>
<td>6.1±2.1</td>
<td>15.4±4.6</td>
<td>4.7±5.5</td>
<td>0.5±0.3</td>
<td>1.0±0.5</td>
</tr>
<tr>
<td>24</td>
<td>10.0±2.1</td>
<td>19.6±5.8</td>
<td>12.7±5.0</td>
<td>3.5±1.6</td>
<td>2.2±1.3</td>
</tr>
<tr>
<td>25</td>
<td>14.4±4.4</td>
<td>33.6±8.9</td>
<td>14.6±5.1</td>
<td>6.0±1.7</td>
<td>2.6±1.8</td>
</tr>
<tr>
<td>26</td>
<td>6.4±2.8</td>
<td>24.1±9.5</td>
<td>16.3±5.7</td>
<td>9.1±1.8</td>
<td>1.9±0.8</td>
</tr>
<tr>
<td>27</td>
<td>11.4±3.6</td>
<td>46.7±12.3</td>
<td>20.7±7.3</td>
<td>11.6±2.2</td>
<td>3.2±1.9</td>
</tr>
<tr>
<td>28</td>
<td>18.0±7.4</td>
<td>23.7±8.5</td>
<td>27.5±9.0</td>
<td>14.9±4.4</td>
<td>6.9±3.4</td>
</tr>
<tr>
<td>Average (µg m$^{-3}$)</td>
<td>9.6±3.3</td>
<td>24.2±7.7</td>
<td>13.9±5.5</td>
<td>6.5±1.7</td>
<td>2.6±1.4</td>
</tr>
<tr>
<td>Average (%)</td>
<td>27.5±7.8</td>
<td>40.6±9.7</td>
<td>32.2±9.4</td>
<td>20.9±4.1</td>
<td>16.7±6.5</td>
</tr>
</tbody>
</table>
Table 3 Daily afternoon (12:00-18:00 BJT) average $O_3$ contributions ($\mu g \text{ m}^{-3}$) of the NCP emissions in the NEC and NWC from 22 to 28 May 2015.

<table>
<thead>
<tr>
<th>Date</th>
<th>Jilin</th>
<th>Liaoning</th>
<th>Shanxi</th>
<th>Shaanxi</th>
<th>Inner Mongolia</th>
</tr>
</thead>
<tbody>
<tr>
<td>22</td>
<td>12.4±0.1</td>
<td>23.9±2.7</td>
<td>12.7±0.0</td>
<td>7.7±0.0</td>
<td>2.8±0.6</td>
</tr>
<tr>
<td>23</td>
<td>25.8±2.5</td>
<td>38.9±6.2</td>
<td>21.5±1.1</td>
<td>13.1±0.3</td>
<td>5.1±0.2</td>
</tr>
<tr>
<td>24</td>
<td>35.0±3.6</td>
<td>47.5±8.1</td>
<td>31.3±3.9</td>
<td>21.2±1.9</td>
<td>8.5±0.5</td>
</tr>
<tr>
<td>25</td>
<td>45.7±8.4</td>
<td>69.5±15.5</td>
<td>39.7±6.4</td>
<td>21.5±2.5</td>
<td>9.9±0.6</td>
</tr>
<tr>
<td>26</td>
<td>16.6±1.6</td>
<td>41.0±5.9</td>
<td>36.4±4.6</td>
<td>21.7±2.4</td>
<td>10.8±0.7</td>
</tr>
<tr>
<td>27</td>
<td>23.9±5.0</td>
<td>69.3±16.4</td>
<td>51.7±7.8</td>
<td>33.5±4.5</td>
<td>9.6±1.0</td>
</tr>
<tr>
<td>28</td>
<td>41.7±5.5</td>
<td>35.1±6.5</td>
<td>52.3±9.0</td>
<td>26.5±4.7</td>
<td>12.2±1.8</td>
</tr>
</tbody>
</table>

Average ($\mu g \text{ m}^{-3}$) | 28.7±3.8 | 46.5±8.8 | 35.1±4.7 | 20.7±2.3 | 8.4±0.7 |
Average (%)                     | 19.5±2.9 | 27.4±5.9 | 21.2±3.2 | 15.8±2.0 | 8.0±0.7 |
Figure 1 WRF-CHEM simulation domain with topography. The blue circles represent centers of cities with ambient monitoring sites and the red circle denotes the NCNST site. The size of the blue circle represents the number of ambient monitoring sites of cities.

Figure 2 (a) Geopotential heights and (b) the mean sea level pressures with wind vectors during the summer monsoon season in 2015.

Figure 3 Relationships of observed PM$_{2.5}$ and O$_3$ concentrations in NCP with those in the NEC during May to September from 2013 to 2016.

Figure 4 Same as Figure 3, but for the NWC.

Figure 5 Comparison of measured (black dots) and predicted (blue line) diurnal profiles of near-surface hourly (a) PM$_{2.5}$, (b) O$_3$, and (c) NO$_2$ averaged over all ambient monitoring stations in Northern China from 22 to 28 May 2015.

Figure 6 Comparison of measured (black dots) and simulated (black line) diurnal profiles of submicron aerosol species of (a) POA, (b) SOA, (c) sulfate, (d) nitrate, and (e) ammonium at NCNST site in Beijing from 22 to 28 May 2015.

Figure 7 Pattern comparison of simulated vs. observed near-surface PM$_{2.5}$ at 08:00 BJT during from 23 to 28 May 2015. Colored circles: PM$_{2.5}$ observations; color contour: PM$_{2.5}$ simulations; black arrows: simulated surface winds.

Figure 8 Same as Figure 7, but for the near-surface O$_3$ at 14:00 BJT.

Figure 9 Contributions of NCP emissions to the daily mean near-surface PM$_{2.5}$ concentration in the NEC and NWC from 23 to 28 May 2015.

Figure 10 Average percentage contribution of NCP emissions to PM$_{2.5}$ concentrations in the NEC and NWC from 22 to 28 May 2015.

Figure 11 Same as Figure 9, but for the afternoon (12-18:00 BJT) O$_3$ concentration.

Figure 12 Same as Figure 10, but for the afternoon (12-18:00 BJT) O$_3$ concentration.
Figure 1 WRF-CHEM simulation domain with topography. The blue circles represent centers of cities with ambient monitoring sites and the red circle denotes the NCNST site. The size of the blue circle represents the number of ambient monitoring sites of cities.
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Figure 12 Same as Figure 10, but for the afternoon (12-18:00 BJT) O₃ concentration.