Contributions of Trans-boundary Transport to the Summertime Air Quality in Beijing, China

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Abstract: In the present study, the WRF-CHEM model is used to evaluate the contributions of trans-boundary transport to the air quality in Beijing during a persistent air pollution episode from 5 to 14 July 2015 in Beijing-Tianjin-Hebei (BTH), China. Generally, the predicted temporal variations and spatial distributions of PM₂.₅ (fine particulate matter), O₃ (ozone), and NO₂ are in good agreement with observations in BTH. The WRF-CHEM model also reproduces reasonably well the temporal variations of aerosol species compared to measurements in Beijing. The factor separation approach is employed to evaluate the contributions of trans-boundary transport of emissions outside of Beijing to the PM₂.₅ and O₃ levels in Beijing. On average, in the afternoon during the simulation episode, the pure local emissions contribute 22.4% to the O₃ level in Beijing, less than 36.6% from pure emissions outside of Beijing. The O₃ concentrations in Beijing are decreased by 5.1% in the afternoon due to interactions of local emissions with those outside of Beijing. The pure emissions outside of Beijing play a dominant role in the PM₂.₅ level in Beijing, with a contribution of 61.5%, much more than 13.7% from pure Beijing local emissions. The emissions interactions enhance the PM₂.₅ concentrations in Beijing, with a contribution of 5.9%. Therefore, the air quality in Beijing is primarily determined by the trans-boundary transport of emissions outside of Beijing during summertime, showing that the cooperation with neighboring provinces to mitigate pollutant emissions is a key for Beijing to improve air quality. Considering the uncertainties in the emission inventory and the meteorological field simulations, further studies need to be performed to improve the WRF-CHEM model simulations to reasonably evaluate trans-boundary transport contributions to the air quality in Beijing for supporting the design and implementation of emission control strategies.
Beijing, the capital of China, has become an environmentally stressed city due to growing population, increasing transportation activity, and city expansion (Parrish and Zhu, 2009). Beijing is situated in northeastern China, surrounded from the southwest to the northeast by the Taihang Mountains and the Yanshan Mountains and open to the North China Plain (NCP) in the south and east. Unfortunately, NCP has become one of the most polluted areas in China due to rapid industrialization and urbanization (Zhang et al., 2013). When south or east winds are prevalent in NCP, air pollutants originated from NCP are transported to Beijing and surrounding areas and subject to be accumulated due to the mountain blocking, causing heavy air pollution in Beijing (Long et al., 2016).

PM$_{2.5}$ (fine particulate matter) and O$_3$ (ozone) are considered to be the most serious air pollutants of concern in Beijing during summertime (e.g., Xie et al., 2015; Zheng et al., 2015; Chen et al., 2015; Wang et al., 2016). The mean summertime PM$_{2.5}$ mass concentration is about 80 µg m$^{-3}$ in 2013 (Li et al., 2015a), exceeding the second grade of National Ambient Air Quality Standards (NAAQS) in China and also higher than the average PM$_{2.5}$ concentration of 78.1µg m$^{-3}$ during the period from 2004 to 2012 (Liu et al., 2015). During haze pollution events in summer 2014, the PM$_{2.5}$ concentration generally reaches 100 µg m$^{-3}$, and even exceeds 150 µg m$^{-3}$ in Beijing (Wang et al., 2016). An increasing O$_3$ trend has been observed in Beijing from 2002 to 2010 (Wang et al., 2012; Wang et al., 2013). The average maximum 1-h O$_3$ concentration has been reported to achieve 140 µg m$^{-3}$ during summertime of 2013 in Beijing (Wang et al., 2014a). Wang et al. (2016) have demonstrated that the summertime O$_3$ mass concentration holds a high level in 2014 in Beijing, with a daily
average of up to 110 µg m$^{-3}$. Chen et al. (2015) have further shown that the average
maximum daily O$_3$ concentrations are higher than 150 µg m$^{-3}$ during the summer in 2015 at
most of monitoring sites in Beijing.

In recent years, Beijing has implemented aggressive emission control strategies to ameliorate the air quality (Parrish and Zhu, 2009). Both NO$_x$ (NO+NO$_2$) and total VOCs (volatile organic compounds) in Beijing have decreased linearly since 2002, while the daytime average O$_3$ concentration still increases rapidly (Tang et al., 2009; Wang et al., 2012; Zhang et al., 2014). Zhang et al. (2014) have highlighted the importance of the trans-boundary transport and the cooperation with neighboring provinces to control the O$_3$ level in Beijing. Pollutants transported from outside of Beijing and formed locally together determine the air quality in Beijing (Meng et al., 2006; Zhang et al., 2012).

Several studies have been performed to investigate the role of trans-boundary transport in the air quality of Beijing based on observational analysis and model simulations. Using the US EPA’s Model-3/CMAQ model simulation in the Beijing area, Streets et al. (2007) have pointed out that non-Beijing sources can contribute 50-70% of Beijing’s PM$_{2.5}$ concentration and 20-30% of O$_3$ concentration. Wang et al. (2009) have indicated that O$_3$ formation in Beijing is not only affected by local emissions, but also influenced by Tianjin and the south of Hebei Province. The intense regional transport of pollutants from south to north in NCP has been proposed to be the main reason for the heavy haze pollution in January 2013 in Beijing (Sun et al., 2014; Tao et al., 2014; Wang et al., 2014b). Jiang et al. (2015) have demonstrated that the transport from the environs of Beijing contributes about 55% of the peak PM$_{2.5}$ concentration in the city during a heavy haze event in December 2013.
Since September 2013, the ‘Atmospheric Pollution Prevention and Control Action Plan’ (hereafter referred to as APPCAP) has been implemented, which is released by the Chinese State Council to reduce PM$_{2.5}$ by up to 25% by 2017 relative to 2012 levels. After implementation of the APPCAP, high PM$_{2.5}$ mass concentrations still can be observed and the O$_3$ pollution has deteriorated during summertime since 2013 in Beijing (Chen et al., 2015; Wang et al., 2016). Hence, studies are imperative to explore the O$_3$ and PM$_{2.5}$ formation from various sources and evaluate the pollutants contributions from local production and trans-boundary transport in Beijing, to support the design of mitigation strategies.

The purpose of the present study is to evaluate the contributions of trans-boundary transport of emissions outside of Beijing to the air quality in Beijing and interaction of emissions in and outside of Beijing using the WRF-CHEM model. The model configuration and methodology are described in Section 2. Model results and sensitivity studies are presented in Section 3, and conclusions and discussions are given in Section 4.

2 Model and Methodology

2.1 WRF-CHEM Model

The WRF-CHEM model used in the study is developed by Li et al. (2010, 2011a, b, 2012) at the Molina Center for Energy and the Environment, with a new flexible gas phase chemical module and the CMAQ aerosol module developed by US EPA. The wet deposition follows the method used in the CMAQ and the surface deposition of chemical species is parameterized following Wesely (1989). The photolysis rates are calculated using the FTUV (Li et al., 2005; Li et al., 2011a), in which the effects of aerosols and clouds on photolysis are
The inorganic aerosols are predicted in the WRF-CHEM model using ISORROPIA Version 1.7 (Nenes et al., 1998). The secondary organic aerosol (SOA) formation is calculated using a non-traditional SOA module. The volatility basis-set (VBS) modeling method is used in the module, assuming that primary organic components are semi-volatile and photochemically reactive and are distributed in logarithmically spaced volatility bins. Detailed information about the volatility basis-set approach can be found in Li et al (2011b). The SOA formation from glyoxal and methylglyoxal is also parameterized as a first-order irreversible uptake by aerosol particles and cloud droplets.

2.2 Model Configuration

A persistent air pollution episode from 5 to 14 July 2015 in Beijing-Tianjin-Hebei (BTH) is simulated using the WRF-CHEM model. During the episode, the observed mean daily PM$_{2.5}$ concentration is 73.8 µg m$^{-3}$ and the average O$_3$ concentration in the afternoon reaches 237.0 µg m$^{-3}$ in Beijing. The WRF-CHEM model adopts one grid with horizontal resolution of 6 km and 35 sigma levels in the vertical direction, and the grid cells used for the domain are 200 × 200 (Figure 1). The physical parameterizations include the microphysics scheme of Hong et al (Hong and Lim, 2006), the Mellor, Yamada, and Janjic (MYJ) turbulent kinetic energy (TKE) planetary boundary layer scheme (Janjić, 2002), the Unified Noah land-surface model (Chen and Dudhia, 2001), the rapid radiative transfer model (RRTM) long wave radiation scheme (Mlawer et al., 1997) and the Goddard shortwave parameterization (Suarez and Chou, 1994). The NCEP $1^\circ \times 1^\circ$ reanalysis data are used to obtain the meteorological initial and boundary conditions. The chemical initial and boundary
conditions are interpolated from the 6h output of MOZART (Horowitz et al., 2003). The
spin-up time of the WRF-CHEM model is 28 hours. The SAPRC-99 (Statewide Air Pollution
Research Center, version 1999) chemical mechanism is used in the present study. The
anthropogenic emissions are developed by Zhang et al. (2009), including contributions from
agriculture, industry, power generation, residential, and transportation sources. The biogenic
emissions are calculated online using the MEGAN (Model of Emissions of Gases and

2.3 Factor Separation Approach

The formation of the secondary atmospheric pollutant, such as O₃, secondary organic
aerosol, and nitrate, is a complicated nonlinear process in which its precursors from various
emission sources and transport react chemically or reach equilibrium thermodynamically.
Nevertheless, it is not straightforward to evaluate the contributions from different factors in a
nonlinear process. The factor separation approach (FSA) proposed by Stein et al. (1993) can
be used to isolate the effect of one single factor from a nonlinear process and has been widely
used to evaluate source effects (Gabusi et al., 2008; Weinroth et al., 2008; Carnevale et al.,
2010; Li et al., 2014). The total effect of one factor in the presence of others can be
decomposed into contributions from the factor and that from the interactions of all those
factors.

Suppose that field \( f \) depends on a factor \( \varphi \):

\[ f = f(\varphi) \]

The FSA decomposes function \( f(\varphi) \) into a constant part that does not depend on \( \varphi \) \( (f(0)) \)
and a \( \varphi \)-depending component \( (f'(\varphi)) \), as follows:
Considering that there are two factors $X$ and $Y$ that influence the formation of secondary pollutants in the atmosphere and also interact with each other. Denoting $f_{XY}$, $f_X$, $f_Y$, and $f_0$ as the simulations including both of two factors, factor $X$ only, factor $Y$ only, and none of the two factors, respectively. The pure contributions of factor $X$ and $Y$ can be isolated as follows:

\[
\begin{align*}
    f'_X &= f_X - f_0 \\
    f'_Y &= f_Y - f_0
\end{align*}
\]

Note that term $f'_{X(Y)}$ represents the pure impacts of factor $X(Y)$, while $f_0$ is the term independent of factors $X$ and $Y$.

The simulation including both factors $X$ and $Y$ is given by:

\[
f_{XY} = f_0 + f'_X + f'_Y + f_{XY}
\]

The mutual interaction between $X$ and $Y$ can be expressed as:

\[
f'_{XY} = f_{XY} - f_0 - f'_X - f'_Y = f_{XY} - (f_X - f_0) - (f_Y - f_0) - f_0 = f_{XY} - f_X - f_Y + f_0
\]

The above equation shows that the study needs four simulations, $f_{XY}, f_X, f_Y$ and $f_0$, to evaluate the contributions of two factors and their synergistic interactions.

### 2.4 Statistical Methods for Comparisons

In the present study, the mean bias ($MB$), root mean square error ($RMSE$) and the index of agreement ($IOA$) are used as indicators to evaluate the performance of WRF-CEHM model in simulation against measurements. $IOA$ describes the relative difference between the model and observation, ranging from 0 to 1, with 1 indicating perfect agreement.
\[ MB = \frac{1}{N} \sum_{i=1}^{N} (P_i - O_i) \]

\[ RMSE = \left( \frac{1}{N} \sum_{i=1}^{N} (P_i - O_i)^2 \right)^{1/2} \]

\[ IOA = 1 - \frac{\sum_{i=1}^{N} (P_i - O_i)^2}{\sum_{i=1}^{N} (P_i - \bar{P})^2 + \sum_{i=1}^{N} (O_i - \bar{O})^2} \]

where \( P_i \) and \( O_i \) are the predicted and observed pollutant concentrations, respectively. \( N \) is the total number of the predictions used for comparisons, and \( \bar{P} \) and \( \bar{O} \) represents the average of the prediction and observation, respectively.

2.5 Pollutants Measurements

The hourly measurements of \( \text{O}_3 \), \( \text{NO}_2 \), and \( \text{PM}_{2.5} \) used in the study are downloaded from the website [http://www.aqistudy.cn/](http://www.aqistudy.cn/). The submicron sulfate, nitrate, ammonium, and organic aerosols are observed by the Aerodyne Aerosol Chemical Speciation Monitor (ACSM), which is deployed at the National Center for Nanoscience and Technology (NCNST), Chinese Academy of Sciences, Beijing (Figure 1). The mass spectra of organic aerosols are analyzed using the Positive Matrix Factorization (PMF) technique to separate into four components: hydrocarbon-like organic aerosol (HOA), cooking organic aerosol (COA), coal combustion organic aerosol (CCOA), and oxygenated organic aerosol (OOA). HOA, COA, and CCOA are interpreted as surrogates of primary organic aerosol (POA), and OOA is a surrogate of SOA.

3 Results and Discussions

3.1 Summertime Air Quality in Beijing

The APPCAP has been implemented since 2013 September, so comparisons of summertime pollutants between 2013 and 2015 can show the mitigation effects on the air
Considering that high O$_3$ concentrations generally take place in the afternoon during summertime, Table 1 presents the summertime hourly concentrations of pollutants in the afternoon (12:00 – 18:00 Beijing Time (BJT)) averaged at 12 monitoring sites in Beijing in 2013 and 2015. Apparently, implementation of the APPCAP has considerably decrease the concentrations of primary species of CO and SO$_2$, particularly with regard to SO$_2$, reduced by more than 40% from 2013 to 2015. Most of NO$_x$ exist in the form of NO$_2$ in the afternoon during summertime due to active photochemical processes. Therefore, 25.1% decrease of NO$_2$ in the afternoon from 2013 to 2015 shows that the NO$_x$ emission mitigation is also effective in Beijing. The PM$_{2.5}$ concentrations are decreased by about 24.0% from 2013 to 2015, approaching the expected 25% reduction by 2017 relative to 2012 levels. However, the O$_3$ trend is not anticipated in Beijing, and O$_3$ concentrations are increased from 133.0 µg m$^{-3}$ in 2013 to 163.2 µg m$^{-3}$ in 2015, enhanced by 22.8%. For the discussion convenience, we have defined the O$_3$ exceedance with hourly O$_3$ concentrations exceeding 200 µg m$^{-3}$ and PM$_{2.5}$ exceedance with hourly PM$_{2.5}$ concentrations exceeding 75 µg m$^{-3}$. Although the PM$_{2.5}$ exceedance frequency in the afternoon has been decreased by 25.0% from 2013 to 2015, but still remains 32.7% in 2015. The O$_3$ exceedance frequency in 2015 is 31.8%, enhanced by 57.6% compared to 20.2% in 2013. Hence, during the summertime of 2015, two years after implementation of the APPCAP, Beijing still has experienced high O$_3$ and/or PM$_{2.5}$ pollutions frequently.

### 3.2 Model Performance

The hourly measurements of O$_3$, NO$_2$, and PM$_{2.5}$ in Beijing-Tianjin-Hebei (BTH) and ACSM measured aerosol species in Beijing are used to validate the WRF-CHEM model.
3.2.1 \textbf{O}_3, \textit{NO}_2, \textit{and PM}_{2.5} \textbf{Simulations in Beijing}

Figure 2 shows the temporal variations of observed and simulated near-surface \textit{O}_3, \textit{NO}_2,
and \textit{PM}_{2.5} concentrations averaged over monitoring sites in Beijing from 5 to 14 July 2015.
The WRF-CHEM model performs reasonably well in simulating the \textit{PM}_{2.5} variations
compared with observations in Beijing. The \textit{MB} and \textit{RMSE} are -3.6 \text{ µg m}^{-3} and 22.5 \text{ µg m}^{-3},
respectively, and the \textit{IOA} is 0.86. The underestimation of \textit{PM}_{2.5} concentrations on July 8 and
overestimation on July 11 and 14 are still rather large, perhaps caused by the simulated wind
field uncertainties that influence the pollutants transports from outside of Beijing or lack of
resolving convective clouds due to the 6 km horizontal resolution. The model well
reproduces the temporal variations of \textit{O}_3 concentrations, with an \textit{IOA} of 0.92. The model
considerably underestimates the \textit{O}_3 concentration during daytime on July 5, 6 and 13. Most
of monitoring sites in Beijing are concentrated in the urban area. Therefore, if the simulated
winds cause the \textit{O}_3 plume formed in the urban area to leave early or deviate the \textit{O}_3 plume
transported from outside of Beijing from the urban area, the model is subject to underestimate
the \textit{O}_3 concentration in Beijing (Bei et al., 2010). The WRF-CHEM model also reasonably
yields the \textit{NO}_2 diurnal profiles, but frequently overestimates the \textit{NO}_2 concentrations during
nighttime, which is likely caused by the failure of boundary layer simulations.

3.2.2 \textbf{Aerosol Species Simulations in Beijing}

Figure 3 shows the temporal variations of simulated and observed aerosol species at
\textit{NCNST} site in Beijing from 5 to 14 July 2015. The WRF-CHEM model generally performs
reasonably in simulating the aerosol species variations compared with ACSM measurements.
As a primary aerosol species, the POA in Beijing is determined by direct emissions from various sources and transport from outside of Beijing, so uncertainties from emissions and meteorological fields remarkably affect the model simulations (Bei et al., 2012; Bei et al., 2013). Although the MB and RMSE for POA are 0.0 µg m\(^{-3}\) and 3.1 µg m\(^{-3}\), respectively, the IOA is less than 0.60, indicating the considerable biases in POA simulations. The WRF-CHEM model has difficulties in well simulating the sulfate aerosol, with an IOA lower than 0.60. The model cannot produce the observed high peaks of sulfate aerosols around noontime on 8, 11, and 12 July 2015. The sulfate aerosol in the atmosphere is produced from multiple sources, 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. There are two possible reasons for the biases in sulfate simulations. Firstly, the model is not able to resolve well convective clouds due to the 6 km horizontal resolution used in simulations, reducing the sulfate production from cloud processes. Secondly, a large amount of SO\(_2\) is released from point sources, such as power plants or agglomerated industrial zones, and the transport of SO\(_2\) from point sources is much sensitive to wind field simulations. The model reasonably well reproduces the observed temporal variations of SOA, nitrate, and ammonium, with IOA\(_s\) exceeding 0.75.

**3.2.2 \(O_3\), NO\(_2\), and PM\(_{2.5}\) Simulations in BTH**

Figure 4 shows the diurnal profiles of observed and simulated near-surface \(O_3\), NO\(_2\), and PM\(_{2.5}\) concentrations averaged over monitoring sites in BTH from 5 to 14 July 2015. The WRF-CHEM model exhibits good performance in predicting the temporal variations of \(O_3\),
NO$_2$, and PM$_{2.5}$ concentrations compared with measurements in BTH, with IOAs higher than 0.80. In addition, O$_3$ and NO$_2$ simulations are also improved in BTH compared to those in Beijing, indicating better model performance for regional simulations in a large scale.

Figure 5 presents the distributions of calculated and observed near-surface PM$_{2.5}$ concentrations along with the simulated wind fields at 10:00 Beijing Time (BJT) on the six selected representative days with high O$_3$ and PM$_{2.5}$ concentrations. The calculated PM$_{2.5}$ spatial patterns generally agree well with the observations at the monitoring sites. The observed PM$_{2.5}$ concentrations in BTH are still high even after implementation of the APPCAP, frequently exceeding 75 µg m$^{-3}$ on the selected six days. The PM$_{2.5}$ concentrations in Beijing are higher than 115 µg m$^{-3}$ at 10:00 BJT on 8, 11, and 12 July 2015, causing moderate air pollution.

Further analyses are provided to interpret the high PM$_{2.5}$ formation in Beijing on July 8 and 11, 2015. Figure 6 presents the haze formation process from 20:00 BJT on July 7 to 18:00 BJT on July 8 in Beijing. From 20:00 to 22:00 on July 7, the prevailing southeast winds bring the pollutants formed in Tianjin to Beijing. Meanwhile, a PM$_{2.5}$ plume in the east of Beijing has been enhanced and commenced to be transported to Beijing. From 00:00 BJT to 06:00 BJT on July 8, the formed PM$_{2.5}$ plume in the east of Beijing has been transported to the city, but only partially influenced the urban area due to the prevailing northeast winds.

From 08:00 to 10:00 on July 8, the PM$_{2.5}$ pollution in the urban area of Beijing is enhanced due to rush hour emissions. After 12:00 BJT on July 8, the PM$_{2.5}$ plume formed in the urban area of Beijing has been transported to the northwest of Beijing and prevailing southeast winds continuously carry the PM$_{2.5}$ formed in Tianjin to Beijing. Figure 7 shows the
dominant role of trans-boundary transport in the haze formation in Beijing from 20:00 BJT on July 10 to 18:00 BJT on July 11. From 20:00 on July 10 to 08:00 BJT on July 11, a PM$_{2.5}$ plume formed in the east of Beijing has been transported to Beijing forced by east winds, causing continuous increase of the PM$_{2.5}$ concentration in Beijing. From 08:00 to 12:00 BJT on July 11, the transported PM$_{2.5}$ plume from outside of Beijing has been further enhanced due to rush hour emissions in Beijing, but the WRF-CHEM model overestimates the PM$_{2.5}$ concentration in the urban area due to the simulated weak or calm winds. After 12:00 BJT on July 11, the enhanced south winds commence to transport the PM$_{2.5}$ plume in the urban area to the north of Beijing.

The O$_3$ concentration during summertime reaches its peak during the period from 14:00 to 16:00 BJT in Beijing (Tang et al., 2012). Figure 8 presents the spatial distribution of calculated and measured near-surface O$_3$ concentration at 15:00 Beijing Time (BJT) on the selected six days, along with the simulated wind fields. In general, the simulated O$_3$ spatial patterns are consistent with the measurements, but model biases still exist, possibly caused by the active convections in the afternoon, which cannot be well resolved in WRF-CHEM model. High O$_3$ concentrations at 15:00 BJT in Beijing are observed and also simulated by the model, frequently exceeding 250 µg m$^{-3}$. The O$_3$ transport to Beijing from its surrounding areas is also obvious when the winds are easterly or southerly. Figure 9 provides the spatial distribution of simulated and observed near-surface NO$_2$ concentration on the selected six days at 08:00 BJT when the NO$_2$ concentration reaches it peak due to rush hour NO$_x$ emissions and low planetary boundary layer (PBL). The simulated near-surface NO$_2$ concentrations highlights the dominant impact of the anthropogenic emissions, primarily
concentrated in cities or their downwind areas, which generally agree well with the measurements. Beijing is surrounded from south to east by cities with high NO\textsubscript{2} concentrations, which can influence the O\textsubscript{3} formation in Beijing when south or east winds are prevalent.

The good agreements between predicted PM\textsubscript{2.5}, O\textsubscript{3}, NO\textsubscript{x} and aerosol species and the corresponding measurements show that the modeled meteorological fields and emissions used in simulations are generally reasonable.

### 3.3 Contributions of Trans-boundary Transport to the O\textsubscript{3} and PM\textsubscript{2.5} Levels in Beijing

The FSA is used in the present study to evaluate the contributions and interactions of emissions from Beijing and outside of Beijing to the near-surface concentrations of O\textsubscript{3} and PM\textsubscript{2.5} in Beijing. Four model simulations are performed, including \( f_{BS} \) with both the anthropogenic emissions from Beijing and outside of Beijing, \( f_{B} \) with the emission from Beijing alone, \( f_{S} \) with only emissions outside of Beijing, and \( f_{0} \) without both the emissions from Beijing and outside of Beijing, representing background concentrations. The air pollutants levels in Beijing are determined by the contribution from pure local emissions \( (f_{B}, f_{B} - f_{0}) \), the trans-boundary transport of pure emissions outside of Beijing \( (f_{S}, f_{S} - f_{0}) \), emission interactions \( (f_{BS}, f_{BS} - f_{B} - f_{S} + f_{0}) \), and the background \( (f_{0}) \).

Figure 10 provides the temporal variation of the average contributions to near-surface O\textsubscript{3} and PM\textsubscript{2.5} concentrations in Beijing from total emissions \( (f_{BS}) \), emissions from Beijing \( (f_{B}) \) and outside of Beijing \( (f_{S}) \) during the simulation episode. Apparently, the emissions outside of Beijing generally play a more important role in the O\textsubscript{3} level of Beijing than the Beijing local emissions. Even when the Beijing local emissions are excluded, the O\textsubscript{3}
concentration in Beijing still remains high level, with an average of 153 µg m$^{-3}$ and ranging from 130 to 180 µg m$^{-3}$ in the afternoon. When only considering the Beijing local emission in simulations, the afternoon average O$_3$ concentration in Beijing is approximately 126.6 µg m$^{-3}$, varying from 80 to 160 µg m$^{-3}$. On July 13, the contribution from Beijing local emissions exceeds that from emissions outside of Beijing because north winds are prevailing, bringing clean air to Beijing (Figure 8f). Table 2 gives the average O$_3$ contributions from 12:00 to 18:00 BJT in Beijing from pure Beijing local emissions, pure emissions outside of Beijing, emission interactions, and background. The pure local emissions contribute about 22.4% on average in the afternoon to the O$_3$ level in Beijing, varying from 15.5% to 35.4%. The pure emissions outside of Beijing contribute more than pure local sources, with an average contribution of 36.6%, ranging from 15.2% to 48.0%. The emission interactions in Beijing decrease the O$_3$ level by 5.1% on average. O$_3$ formation is a nonlinear process, depending on not only the absolute levels of NO$_x$ and VOCs, but also the ratio of VOC/NO$_x$. When the O$_3$ precursors emitted from outside of Beijing are transported to Beijing and mixed with local emissions, the concentrations of O$_3$ precursors are increased and the ratio of VOC$_x$/NO$_x$ is also altered, causing the formed O$_3$ concentration unequal to the simple linear summation of O$_3$ contributions from the local and outside of Beijing emissions. The background O$_3$ in Beijing plays an important role in the O$_3$ level in the afternoon, accounting for 45.6% of the O$_3$ concentration. The O$_3$ contributions in Beijing induced by the trans-boundary transport of emissions outside of Beijing is about 31.5% of the O$_3$ concentration during the study episodes, indicating that the trans-boundary transport constitutes the main reason for the elevated O$_3$ level in Beijing after implementation of the APPCAP.
When the Beijing local emissions are not considered in simulations, Beijing still experiences high PM$_{2.5}$ pollution, with an average PM$_{2.5}$ concentration of 48.3 µg m$^{-3}$ during the simulation episode, and the PM$_{2.5}$ level in Beijing still exceeds 75 µg m$^{-3}$ on several days. However, when only considering the Beijing local emissions, the average PM$_{2.5}$ concentration in Beijing is 19.6 µg m$^{-3}$ during the episode, showing that Beijing’s PM$_{2.5}$ pollution is dominated by the trans-boundary transport. During the study episode, the average PM$_{2.5}$ contribution from the pure local emissions is 13.7%, which is much lower than the contribution of 61.5% from the pure emissions outside of Beijing, further showing the dominant role of the trans-boundary transport in the Beijing PM$_{2.5}$ pollution. The emission interactions enhance the PM$_{2.5}$ level in Beijing on average, with a contribution of 5.9%. The background PM$_{2.5}$ contribution to Beijing is 18.9% on average, lower than those for O$_3$. The PM$_{2.5}$ contributions caused by the trans-boundary transport is about 67.4% of PM$_{2.5}$ concentrations in Beijing, indicating that the cooperation with neighboring provinces to control the PM$_{2.5}$ level is a key for Beijing to improve air quality.

Figure 11 shows the temporal variation of the averaged contributions to the near-surface aerosol constituents from total emissions ($f_{BS}$), pure emissions from Beijing ($f_B$), the trans-boundary transport of pure emissions outside of Beijing ($f_S$), emission interactions ($f_{BS}$), and the background ($f_B$) during the simulation episode. The temporal variations of elemental carbon (EC) and POA from pure local emissions and trans-boundary transport exhibit obvious diurnal cycles, e.g., highest during nighttime and lowest in the afternoon, corresponding to the variations of PBL height and anthropogenic emissions. The SOA from pure local emissions reaches its peak in the afternoon when the O$_3$ concentration is high, but
the trans-boundary transport causes the gradual accumulation process of SOA in Beijing from July 5 to 9 and from July 9 to 13. The sulfate temporal profile from the trans-boundary transport is similar to that of SOA, also showing the accumulation process. In addition, the sulfate aerosols from pure local emissions do not vary remarkably. The nitrate aerosols from pure local emissions and the trans-boundary transport generally attain peaks in the morning when the air temperature is not high and the HNO3 concentrations are not low. The ammonium aerosol variations are generally determined by those of sulfate and nitrate aerosols. For example, the variations of ammonium aerosols from the trans-boundary include not only the morning peaks, but also the accumulation processes from July 5 to 9 and from July 9 to 13. Except the sulfate aerosol, the temporal variations of aerosol species from background are not large.

Table 4 presents the average aerosol constituents contributions from pure Beijing local emissions, pure emissions outside of Beijing, emission interactions, and the background, and mass fractions in the total PM$_{2.5}$ in Beijing during the episode. Organic aerosols (POA+SOA) constitute the most important component of PM$_{2.5}$, accounting for 34.8% of PM$_{2.5}$ mass concentration, which is consistent with the ACSM measurement in Beijing (Sun et al., 2014). In addition, SOA contributes more than 70% of organic aerosol mass concentrations, which is due to the high atmospheric oxidation capability caused by elevated O$_3$ concentrations during summertime. Although the SO$_2$ concentrations have been decreased by more than 40% since implementation of the APPCAP, sulfate aerosols still play an important role in the PM$_{2.5}$ level in Beijing and make up 25.1% of the PM$_{2.5}$ mass concentrations, showing high sulfate contributions from the trans-boundary transport and background. The ammonium, nitrate, EC,
and unspecified species account for 13.7%, 14.1%, 5.8%, and 6.5% of the PM$_{2.5}$ mass concentrations, respectively. Secondary aerosol species dominate the PM$_{2.5}$ mass concentration in Beijing, with a contribution of 77.9%, corresponding to the high atmospheric oxidation capability.

The pure local emissions contribute more than 20% of the mass concentrations for the primary aerosol species, but less than 15% for the secondary aerosol species in Beijing (Table 4). The trans-boundary transport of emissions outside of Beijing dominates all the aerosol species levels in Beijing, with contributions exceeding 50%, particularly for SOA and nitrate. In addition, the POA and sulfate background contributions are also high, more than 20%.

Although the primary aerosol species of EC and unspecified constituents are not involved in the chemical process and also do not participate the gas-particle partitioning, the emission interactions still enhance EC and unspecified constituents concentrations, with contributions of around 1.5%, which is caused by the aerosol radiative effect. Mixing of Beijing local emissions with those outside of Beijing increases the aerosol concentrations in the PBL and decreases the incoming solar radiation down to the surface, cooling the temperature of the low level atmosphere to suppress the development of PBL and hinder the aerosol dispersion in the vertical direction. The emission interactions increase the POA and SOA concentrations, with a POA contribution of 5.3% and a SOA contribution of 5.9%. In the VBS modeling approach, primary organic components are assumed to be semi-volatile and photochemically reactive. Mixing of Beijing local emissions with emissions outside of Beijing enhances the organic condensable gases, and considering that the saturation concentrations of the organic condensable gases do not change, more organic condensable gases participate into the
particle phase, increasing the POA and SOA concentrations. The contributions of emission interactions to inorganic aerosols, including sulfate, nitrate, and ammonium, are more complicated, depending on their particle phase and precursors concentrations. In the present study, ISORROPIA (Version 1.7) is used to calculate the thermodynamic equilibrium between the sulfate-nitrate-ammonium-water aerosols and their gas phase precursors $\text{H}_2\text{SO}_4-\text{HNO}_3-\text{NH}_3$-water vapor. Although mixing of Beijing local emissions with those outside of Beijing increase inorganic aerosols precursors, the inorganic aerosol contributions from emission interactions are still uncertain due to the deliberate thermodynamic equilibrium between inorganic aerosols and their precursors. High atmospheric oxidation capability induced by elevated $\text{O}_3$ concentration facilitates $\text{HNO}_3$ formation through $\text{NO}_2$ reaction with OH during daytime and $\text{N}_2\text{O}_5$ formation through $\text{NO}_2$ reaction with $\text{O}_3$ during nighttime. High $\text{O}_3$ concentrations are produced by Beijing local emissions and those outside of Beijing, accelerating the $\text{HNO}_3$ or $\text{N}_2\text{O}_5$ formation. Hence, mixing of Beijing local emissions with those outside of Beijing considerably increases the $\text{HNO}_3$ or $\text{N}_2\text{O}_5$ levels, pushing more $\text{HNO}_3$ or $\text{N}_2\text{O}_5$ into the particle phase. The nitrate contributions from emission interactions are 18.1%, much more than those for other aerosol constituents. $\text{SO}_2$ gas-phase oxidations by OH and sCl are not as fast as $\text{NO}_2$ reaction with OH, so the formation of sulfuric acid is slow, although the $\text{O}_3$ concentration is high during summertime. Therefore, the sulfate contributions from emission interactions is not significant, only 3.4%. As the ammonium precursor, NH$_3$ is generally from direct emissions. The ammonium contributions from emission interactions are 1.5%, similar to those of primary aerosol species that are caused by aerosol radiative effects, indicating that the NH$_3$ emissions are not sufficiently high.
Summary and Conclusions

In the present study, a persistent air pollution episode with high concentrations of $O_3$ and PM$_{2.5}$ are simulated using the WRF-CHEM model during the period from July 5 to 14, 2015 in BTH, to evaluate the contributions of trans-boundary transport to the air quality in Beijing. Although the APPCAP has been implemented since 2013 September, the average $O_3$ concentration in the afternoon has been increased by 22.8% from 2013 to 2015 in Beijing, and Beijing still has experienced high $O_3$ and/or PM$_{2.5}$ pollutions frequently during summertime of 2015.

In general, the predicted temporal variations of PM$_{2.5}$, $O_3$, and NO$_2$ concentrations agree well with observations in Beijing and BTH, but the model biases still exist, which is perhaps caused by the uncertainties of simulated meteorological conditions and the emission inventory. The model also successfully reproduces the spatial distributions of PM$_{2.5}$, $O_3$, and NO$_2$ concentrations compared with measurements. The model performs reasonably well in modeling the variations of aerosol constituents compared with ACSM measurement at NCNST site in Beijing, but there are considerable biases in POA and sulfate simulations.

The FSA is used to investigate the contribution of trans-boundary transport of emissions outside of Beijing to the air quality in Beijing. If the Beijing local emissions are not considered in model simulations, the $O_3$ and PM$_{2.5}$ concentrations in Beijing still remain high levels, showing that the trans-boundary transport of emissions outside of Beijing plays a more important role in the air quality in Beijing than the Beijing local emissions. On average,
the pure Beijing local emissions contribute 22.4% of O$_3$ in the afternoon and 13.7% of PM$_{2.5}$ mass concentrations in Beijing during the episode. The O$_3$ contribution in the afternoon and PM$_{2.5}$ contribution from the trans-boundary transport of emissions outside of Beijing are 36.6% and 61.5%, respectively, far exceeding those from the pure local emissions. The interactions of Beijing local emissions with those outside of Beijing generally decrease the O$_3$ level in the afternoon and increase the PM$_{2.5}$ level in Beijing during the episode, with contributions of -5.1% and +4.4%, respectively. In addition, the trans-boundary transport dominates all the aerosol species levels in Beijing, with contributions exceeding 50% on average, particularly for SOA and nitrate. The emission interactions in general increase all the aerosol species levels due to the aerosol radiative effect and the enhancement of precursors of secondary aerosols. Hence, the air quality in Beijing during summertime is principally determined by the trans-boundary transport of emissions outside of Beijing. The cooperation with neighboring provinces to decrease pollutant emissions is the optimum approach to mitigate the air pollution in Beijing.

It is worth noting that, although the WRF-CHEM model well captures the spatial distributions and temporal variations of pollutants, the model biases still exist. The discrepancies between the predictions and observations are possibly caused by the uncertainties in the emission inventory and the meteorological fields simulations. Future studies need to be conducted to improve the WRF-CHEM model simulations, and further to assess the contributions of trans-boundary transport of emissions outside of Beijing to the air quality in Beijing, considering the rapid changes in anthropogenic emissions since implementation of the APPCAP.
Data availability: The real-time $O_3$ and PM$_{2.5}$ are accessible for the public on the website http://106.37.208.233:20035/. One can also access the historic profile of observed ambient pollutants through visiting http://www.aqistudy.cn/.

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Table 1 Hourly mass concentrations of pollutants averaged in the afternoon at 12 monitoring sites in Beijing during summertime of 2013 and 2015.

<table>
<thead>
<tr>
<th>Pollutants</th>
<th>CO (mg m⁻³)</th>
<th>SO₂ (µg m⁻³)</th>
<th>NO₂ (µg m⁻³)</th>
<th>O₃ (µg m⁻³)</th>
<th>PM₂.₅ (µg m⁻³)</th>
</tr>
</thead>
<tbody>
<tr>
<td>2013</td>
<td>1.09</td>
<td>9.85</td>
<td>31.6</td>
<td>133.0</td>
<td>81.4</td>
</tr>
<tr>
<td>2015</td>
<td>0.88</td>
<td>5.71</td>
<td>23.6</td>
<td>163.2</td>
<td>61.9</td>
</tr>
<tr>
<td>Change (%)</td>
<td>-20.0</td>
<td>-42.0</td>
<td>-25.1</td>
<td>+22.8</td>
<td>-24.0</td>
</tr>
</tbody>
</table>
Table 2 Average O3 contributions (%) from 12:00 to 18:00 BJT in Beijing from pure Beijing local emissions, pure emissions outside of Beijing, the interactions of both emissions, and background from 5 to 14 July 2015.

<table>
<thead>
<tr>
<th>Date</th>
<th>Beijing</th>
<th>Surroundings</th>
<th>Interactions</th>
<th>Background</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$f_B$</td>
<td>$f_S$</td>
<td>$f_{BS}$</td>
<td>$f_0$</td>
</tr>
<tr>
<td>5</td>
<td>15.5</td>
<td>26.1</td>
<td>-2.4</td>
<td>60.8</td>
</tr>
<tr>
<td>6</td>
<td>19.8</td>
<td>30.9</td>
<td>-3.0</td>
<td>52.3</td>
</tr>
<tr>
<td>7</td>
<td>25.5</td>
<td>36.0</td>
<td>-3.6</td>
<td>42.1</td>
</tr>
<tr>
<td>8</td>
<td>27.0</td>
<td>36.9</td>
<td>-5.9</td>
<td>42.0</td>
</tr>
<tr>
<td>9</td>
<td>23.2</td>
<td>35.3</td>
<td>-4.6</td>
<td>46.1</td>
</tr>
<tr>
<td>10</td>
<td>18.6</td>
<td>39.9</td>
<td>-2.6</td>
<td>44.1</td>
</tr>
<tr>
<td>11</td>
<td>29.4</td>
<td>48.0</td>
<td>-10.0</td>
<td>32.6</td>
</tr>
<tr>
<td>12</td>
<td>35.4</td>
<td>40.6</td>
<td>-11.4</td>
<td>35.4</td>
</tr>
<tr>
<td>13</td>
<td>23.4</td>
<td>15.2</td>
<td>-1.5</td>
<td>62.9</td>
</tr>
<tr>
<td>14</td>
<td>20.3</td>
<td>32.2</td>
<td>-3.3</td>
<td>50.8</td>
</tr>
<tr>
<td>Average</td>
<td>22.4</td>
<td>36.6</td>
<td>-5.1</td>
<td>46.1</td>
</tr>
</tbody>
</table>
Table 3 Average PM$_{2.5}$ contributions (%) in Beijing from pure Beijing local emissions, pure emissions outside of Beijing, the interactions of both emissions, and background from 5 to 14 July 2015.

<table>
<thead>
<tr>
<th>Date</th>
<th>Emissions</th>
<th>Beijing</th>
<th>Surroundings</th>
<th>Interactions</th>
<th>Background</th>
</tr>
</thead>
<tbody>
<tr>
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<td>$f'_B$</td>
<td>$f'_S$</td>
<td>$f'_{BS}$</td>
<td>$f'_0$</td>
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</tr>
<tr>
<td>5</td>
<td>14.6</td>
<td>55.1</td>
<td>3.3</td>
<td>27.0</td>
<td></td>
</tr>
<tr>
<td>6</td>
<td>14.9</td>
<td>56.3</td>
<td>3.4</td>
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</tr>
<tr>
<td>7</td>
<td>14.2</td>
<td>56.4</td>
<td>8.0</td>
<td>21.4</td>
<td></td>
</tr>
<tr>
<td>8</td>
<td>13.2</td>
<td>61.1</td>
<td>6.4</td>
<td>19.3</td>
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<tr>
<td>9</td>
<td>15.3</td>
<td>61.3</td>
<td>6.3</td>
<td>17.1</td>
<td></td>
</tr>
<tr>
<td>10</td>
<td>11.5</td>
<td>66.5</td>
<td>6.2</td>
<td>15.8</td>
<td></td>
</tr>
<tr>
<td>11</td>
<td>9.7</td>
<td>71.0</td>
<td>8.1</td>
<td>11.2</td>
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</tr>
<tr>
<td>12</td>
<td>14.2</td>
<td>67.6</td>
<td>5.6</td>
<td>12.6</td>
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<tr>
<td>13</td>
<td>19.2</td>
<td>47.2</td>
<td>3.6</td>
<td>30.0</td>
<td></td>
</tr>
<tr>
<td>14</td>
<td>16.6</td>
<td>53.1</td>
<td>6.4</td>
<td>23.9</td>
<td></td>
</tr>
<tr>
<td><strong>Average</strong></td>
<td><strong>13.7</strong></td>
<td><strong>61.5</strong></td>
<td><strong>5.9</strong></td>
<td><strong>18.9</strong></td>
<td></td>
</tr>
</tbody>
</table>
Table 4 Aerosol species’ contributions (%) from pure Beijing local emissions, pure emissions outside Beijing, interactions of both emissions, and background, and mass fraction in the total PM$_{2.5}$ (%) in Beijing averaged during the period from 5 to 14 July 2015.

<table>
<thead>
<tr>
<th>Emissions Species</th>
<th>Mass Fraction In Total PM$_{2.5}$</th>
<th>Beijing $f_B$</th>
<th>Surroundings $f_S$</th>
<th>Interactions $f_{BS}$</th>
<th>Background $f_0$</th>
</tr>
</thead>
<tbody>
<tr>
<td>EC</td>
<td>5.8</td>
<td>27.0</td>
<td>57.9</td>
<td>1.5</td>
<td>13.6</td>
</tr>
<tr>
<td>POA</td>
<td>9.8</td>
<td>20.8</td>
<td>49.0</td>
<td>5.3</td>
<td>24.9</td>
</tr>
<tr>
<td>SOA</td>
<td>25.0</td>
<td>14.6</td>
<td>64.2</td>
<td>5.9</td>
<td>15.3</td>
</tr>
<tr>
<td>Ammonium</td>
<td>13.7</td>
<td>14.5</td>
<td>65.7</td>
<td>1.5</td>
<td>18.3</td>
</tr>
<tr>
<td>Nitrate</td>
<td>14.1</td>
<td>10.1</td>
<td>71.7</td>
<td>18.1</td>
<td>0.1</td>
</tr>
<tr>
<td>Sulfate</td>
<td>25.1</td>
<td>6.5</td>
<td>52.9</td>
<td>3.4</td>
<td>37.2</td>
</tr>
<tr>
<td>Unspecified</td>
<td>6.5</td>
<td>21.2</td>
<td>61.4</td>
<td>1.6</td>
<td>15.8</td>
</tr>
</tbody>
</table>
Figure Captions

Figure 1 WRF-CHEM simulation domain. 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 denotes the number of ambient monitoring sites of cities.

Figure 2 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 Beijing from 5 to 14 July 2015.

Figure 3 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 5 to 14 July 2015.

Figure 4 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 BTH from 5 to 14 July 2015.

Figure 5 Pattern comparison of simulated vs. observed near-surface PM$_{2.5}$ at 10:00 BJT during the selected periods from 5 to 14 July 2015. Colored circles: PM$_{2.5}$ observations; color contour: PM$_{2.5}$ simulations; black arrows: simulated surface winds.

Figure 6 PM$_{2.5}$ pattern variations in Beijing and surrounding areas from 20:00 BJT on July 7 2015 to 18:00 BJT on July 8 2015. Colored circles: PM$_{2.5}$ observations; color contour: PM$_{2.5}$ simulations; black arrows: simulated surface winds.

Figure 7 PM$_{2.5}$ pattern variations in Beijing and surrounding areas from 20:00 BJT on July 10 2015 to 18:00 BJT on July 11 2015. Colored circles: PM$_{2.5}$ observations; color contour: PM$_{2.5}$ simulations; black arrows: simulated surface winds.

Figure 8 Same as Figure 5, but for O$_3$ at 15:00 BJT.

Figure 9 Same as Figure 5, but for NO$_2$ at 08:00 BJT.

Figure 10 Temporal variations of the average contributions to the near-surface O$_3$ and PM$_{2.5}$ concentrations from total emissions (black line, defined as $f_{B5}$), emissions from Beijing (blue line, defined as $f_B$), and emissions outside of Beijing (red line, defined as $f_S$) in Beijing from 5 to 14 July 2015.

Figure 11 Temporal variations of the average contributions to the near-surface aerosol species concentrations from total emissions (black line, defined as $f_{B5}$), pure emissions from Beijing (blue line, $f'_B$, defined as $f_B - f_0$), pure emissions outside of Beijing (red line, $f'_S$, defined as $f_S - f_0$), the emission interactions...
(green line, $f'_{BS}$, defined as $f_{BS} - f_B - f_S + f_0$), and background (black dashed line, defined as $f_0$) in Beijing from 5 to 14 July 2015.
Figure 1
Figure 2
Figure 3
Figure 4

(a) $MB = 6.7 \mu g m^{-3}$  $RMSE = 14.0 \mu g m^{-3}$  $IOA = 0.83$

(b) $MB = 8.3 \mu g m^{-3}$  $RMSE = 19.4 \mu g m^{-3}$  $IOA = 0.95$

(c) $MB = -0.5 \mu g m^{-3}$  $RMSE = 7.5 \mu g m^{-3}$  $IOA = 0.87$
Figure 5
Figure 6
Figure 6 Continued
Figure 7
Figure 7 Continued

(g) 2015-07-11 08:00 BJT

(h) 2015-07-11 10:00 BJT

(i) 2015-07-11 12:00 BJT

(j) 2015-07-11 14:00 BJT

(k) 2015-07-11 16:00 BJT

(l) 2015-07-11 18:00 BJT
Figure 8
Figure 9
Figure 10

(a) Average O₃ concentrations in Beijing

(b) Average PM₂.₅ concentrations in Beijing
Figure 11