Response of fine particulate matter to reductions in anthropogenic emissions in Beijing during the 2014 Asia–Pacific Economic Cooperation summit

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\textbf{ABSTRACT}

The nested-grid capability of the global chemical transport model GEOS-Chem, with a horizontal resolution of 1/4° × 5/16° (latitude × longitude), was used to identify the chemical species whose reductions made the largest contributions to decreases in PM\textsubscript{2.5} concentrations (fine particulate matter; diameter ≤ 2.5 μm; defined in this study as the sum of sulfate, nitrate, ammonium, black carbon, and organic carbon aerosols) in Beijing during the 2014 Asia–Pacific Economic Cooperation (APEC) summit. A number of numerical experiments were carried out for the period 15 October–29 November 2014. The model reproduced the observed daily variations of concentrations of PM\textsubscript{2.5} and gas-phase species (carbon monoxide, nitrogen dioxide, and sulfur dioxide). Simulated PM\textsubscript{2.5} concentrations decreased by 55.9%–58.5% during the APEC period, compared to other periods in October and November 2014, which agreed closely with measurements. Sensitivity results showed that emissions control measures regarding nitrogen oxides and organic carbon over North China led to the largest reductions in PM\textsubscript{2.5} concentrations in Beijing during the APEC summit, which led to overall reductions in the PM\textsubscript{2.5} concentration of Beijing by 5.7% and 4.6%, respectively. The control of ammonia emissions was found to be able to greatly reduce PM\textsubscript{2.5} concentrations in the whole of North China during the APEC meeting.

1. Introduction

High PM\textsubscript{2.5} concentrations (fine particulate matter; diameter ≤ 2.5 μm) in China have garnered considerable attention because of their role in the deterioration of air quality and harm to human health (Pui, Chen, and Zuo 2014). Observed wintertime PM\textsubscript{2.5} concentrations can exceed 500 μg m\textsuperscript{-3} on heavily polluted days in Beijing (Wang et al. 2014), which makes it essential to reduce emissions to improve air quality in North China. Emissions reduction measures have proved to be effective in improving air quality in Beijing and its surrounding regions (Wang et al. 2009; Gao et al. 2011). During the 22nd Asia–Pacific Economic Cooperation (APEC) summit, which was hosted in Beijing (5–11 November 2014), tough emissions reduction measures were implemented for the city and its surrounding provinces. The measures resulted in reductions in emissions of sulfur dioxide (SO\textsubscript{2}), nitrogen oxides (NO\textsubscript{x}), PM\textsubscript{10}, PM\textsubscript{2.5}, and non-methane volatile organic compounds (NMVOCs) by 39.2%, 49.6%, 66.6%, 61.6%, and 33.6%, respectively, according to the air quality evaluation report.
of the APEC period released by the Chinese Environmental Protection Administration (CNEPA) (http://www.mep.gov.cn/zhxx/hjyw/201412/t20141218_293140.htm). The summit provided an experimental opportunity to study the impact of emissions reduction measures on the air quality in Beijing and surrounding provinces.

Modeling studies of PM$_{2.5}$ sensitivities to the emissions control measures during the APEC period have been reported in the literature (Liu et al. 2015; Zhai et al. 2016). For example, by using the Nested Air Quality Prediction Model System, Liu et al. (2015) showed that the PM$_{2.5}$ concentrations in Beijing would have increased by 15–30 μg m$^{-3}$ (40%–60%) if no emissions control measures were conducted during the APEC period. Although previous studies provide valuable knowledge regarding the effectiveness of the emissions reduction measures over North China, there is still room for further studies. PM$_{2.5}$ has been proved to change nonlinearly in response to its precursors (Ansari and Pandis 1998; West, Ansari, and Pandis 1999; Holt, Selin, and Solomon 2015), and the control of different precursor emissions in different regions may result in different efficiencies in reducing PM$_{2.5}$ concentrations. For example, NO$_x$ emissions are generally considered to be important for nitrate formation, but nitrate concentrations could be more sensitive to ammonia ($\text{NH}_3$) emissions under NH$_3$-poor conditions when NH$_3$ is insufficient to neutralize nitric acid (Wang et al. 2011). Therefore, it is important to quantify the sensitivities of PM$_{2.5}$ to the changes in emissions of different species in different regions over North China.

The goal of this work was to identify the chemical species whose reductions made the largest contributions to the decreases in PM$_{2.5}$ concentrations during the APEC period in Beijing. The observational data, GEOS-Chem model, and numerical experiments are described in Section 2. Section 3 presents the model evaluation, as well as the results from sensitivity simulations. Section 4 summarizes the major conclusions of this study.

2. Data and methods

2.1. GEOS-Chem model

We simulated aerosol concentrations using a nested-grid capability of the global chemical transport model GEOS-Chem (version 9-02; http://geos-chem.org) driven by the GEOS-FP assimilated meteorological fields from the Goddard Earth Observing System of the NASA Global Modeling and Assimilation Office. The model has a horizontal resolution of 1/4° × 5/16° (latitude × longitude) for the East Asia domain (15°–55°N, 70°–140°E) and 47 vertical layers extending from the surface to 0.01 hPa. The chemical boundary conditions were provided by a global GEOS-Chem simulation at 4° × 5° (latitude × longitude) horizontal resolution and updated in the nested-grid region every 3 h (Chen et al. 2009; Kim et al. 2015). GEOS-Chem has a fully coupled treatment of tropospheric NO$_x$–carbon monoxide (CO)–hydrocarbon-ozone chemistry and aerosols including sulfate (SO$_4^{2-}$), nitrite (NO$_2$), ammonium (NH$_4^+$), organic carbon (OC), black carbon (BC) (Park et al. 2003, 2004; Pye et al. 2009), mineral dust (Fairlie, Jacob, and Park 2007), and sea salt (Jaeglé et al. 2011). Convective transport in GEOS-Chem mimics that in the parent GEOS general circulation model (Hack 1994; Zhang and McFarlane 1995), accounting for updraft, downdraft, and entrainment mass fluxes archived separately for deep and shallow convection (Wu et al. 2007). The aerosol wet deposition scheme follows that of Liu et al. (2001). For the scavenging of aerosols, SO$_4^{2-}$, NH$_4^+$, nitrate, and hydrophilic OC and BC, aerosols are assumed to be fully soluble. Dry deposition follows the standard resistance-in-series model of Wesely (1989).

2.2. Emissions

Global emissions of aerosols and their precursors in GEOS-Chem follow Park et al. (2003, 2004), with anthropogenic emissions in East Asia overwritten by the MIX Asian anthropogenic emissions inventory for the year 2010 (http://www.meicmodel.org/data-set-mix.html) developed by Li et al. (2015). The diurnal variation of NH$_3$ emission is applied in the model following Zhu et al. (2013). Anthropogenic emissions of SO$_2$, NO$_x$, CO, NH$_3$, BC, and OC are, respectively, 28.6, 29.1, 170.9, 9.8, 1.8, and 3.4 Tg yr$^{-1}$ in China; and 0.15, 0.33, 1.92, 0.07, 0.02, and 0.03 Tg yr$^{-1}$ in Beijing. We defined three regions for the emissions control tests: the North China domain (NC, 31.5°–43.0°N, 110.0°–123.1°E), Beijing (BJ, 39.5°–41.2°N, 115.6°–117.5°E), and the North China domain excluding Beijing (NC-B).

2.3. Numerical experiments

To identify the chemical species whose reductions in emissions made the largest contribution to the decrease in PM$_{2.5}$ concentrations during the APEC period in Beijing, we performed the following numerical experiments:

1. NoCTRL: Simulation for the period 15 October–29 November 2014, with default emissions in the model.

2. CTRL: As in NoCTRL except that, during the APEC summit (3–12 November 2014), emissions of SO$_2$, NO$_x$ (NO$_x$ = NO + NO$_2$), CO, BC, OC, and NMVOCs were reduced, respectively, by 39.2%, 49.6%, 30.0%, 30%, 30%, and 33.6% in Beijing, and by 30% in NC-B, according to the air quality evaluation report of the APEC period released by CNEPA.
were obtained from the China National Environmental Monitoring Center during 15 October–29 November 2014 (Figure 1), as an essential part of model evaluation. Because the GEOS-FP data were available with a temporal resolution of 3 h, the hourly measurements from the NCDC were averaged over the 3 h for comparison. During the APEC period (3–12 November 2014), the wind direction changed frequently and RH was relatively low, with a mean value of 47.3%. As discussed in Liu et al. (2015), such a meteorological condition in Beijing during the APEC period was relatively conducive for the diffusion of air pollutants. For all five of the meteorological parameters shown in Figure 1, the correlation coefficients (R) between the GEOS-FP meteorological data and measurements were in the range of 0.44–0.97 (p = 0.01). At the Beijing Capital International Airport site, the normalized mean bias,

\[
\text{NMB} = \frac{\sum_{i=1}^{n} (A_i - O_i)}{\sum_{i=1}^{n} O_i} \times 100%\]

where \(A_i\) and \(O_i\) are the assimilated GEOS-FP meteorological fields and NCDC observations at time \(i\) for each parameter, respectively, was −12.7%, −0.1%, −42.0%, +48.1% and +32.0% for RH, SLP, temperature, wind speed, and wind direction, respectively, indicating that the GEOS-FP meteorological fields agreed reasonably well with observations.

### 3.2. Concentrations of aerosols and gas-phase species during the APEC summit

Figure 2 compares the daily concentrations from the CTRL and NoCTRL simulations with ground-based measurements in Beijing for PM\(_{2.5}\), CO, NO\(_x\), and SO\(_2\) concentrations during 15 October–29 November 2014. The daily concentrations of PM\(_{2.5}\), CO, and NO\(_x\) were underestimated, and the daily concentrations of SO\(_2\) were overestimated (Figure 2). There are several reasons for the model biases in Beijing. First, the characteristics of local emissions might not have been captured well by the emissions inventories. Chen et al. (2016) indicated that the emissions inventory underestimated the emissions of NO\(_x\) and CO, but overestimated SO\(_2\) emissions, during October 2014, based on a WRF model coupled with online chemistry (WRF-Chem) simulation. With the boundary layer heights and winds reasonably simulated, they reported that the peak concentrations of CO and NO\(_x\) on polluted days were underestimated by 50%–70%. Though WRF-Chem has different chemical schemes to GEOS-Chem, similar biases in simulated concentrations of CO and NO\(_x\) in Beijing can be obtained with a similar emissions inventory. It is likely that the emissions inventory might not have captured the high CO and NO\(_x\) emissions in October in Beijing. With the concentrations of all the other pollutants underestimated, model results (both
in Chen et al. (2016) and this study) show a severe overestimation of SO$_2$ concentrations in Beijing. The simulated SO$_2$ concentrations in other cities over NC were in the range of 30–70 μg m$^{-3}$, showing much smaller model positive biases than those in Beijing, and indicating that SO$_2$ emissions might have been overestimated in Beijing, as new regulations have been applied in recent years. Secondly, we used the anthropogenic emissions for the year 2010, whereas the observed concentrations were taken in 2014, which likely had a considerable impact on the simulated SO$_2$ concentrations. As a result of China’s ‘Twelfth Five-Year’ Plan for National Economic and Social Development (FYP, 2011–2015), national emissions of SO$_2$ were estimated to have decreased by 12.9% within 2010–2014, due to the wide implementation of flue gas desulfurization systems at thermal power units in China (http://www.mep.gov.cn/gkml/hbb/qt/201507/t20150722_307020.htm). Additionally, the model may have missed some chemical mechanisms or oxidants that oxidize SO$_2$ to SO$_4^{2-}$ (Wang et al. 2014), which requires further study.

Figure 1. Comparison of measured (black) and GEOS-FP (red) 3-h RH, SLP, 10-m temperature, 10-m wind speed, and wind direction, in Beijing, during 15 October–29 November 2014. The area between the two central black lines represents the APEC time period (3–12 November). Mean values before, during, and after the APEC summit, and the corresponding correlation coefficients ($R$) between the GEOS-FP meteorological data and measurements, are also shown.
PM$_{2.5}$ concentrations in Beijing in AP in CTRL decreased by 57.4% and 58.5%, respectively. The concentrations of CO, NO$_2$, and SO$_2$ also showed reductions during AP, in both observations and the CTRL results (Figure 2).

The effects of the emissions control measures on PM$_{2.5}$ concentrations can be obtained by examining the differences in concentrations in AP between the CTRL and NoCTRL simulations. The simulated PM$_{2.5}$ concentration was 29.5 μg m$^{-3}$ in AP in CTRL, which was 23.4% lower than the value of 38.6 μg m$^{-3}$ in NoCTRL.

### 3.3. Sensitivity of the PM$_{2.5}$ concentration to emission controls of different species

Sensitivity simulations were performed to identify the chemical species whose reductions in emissions made
the largest contribution to the decrease in PM$_{2.5}$ concentrations during the APEC period in Beijing, as described in Section 2.3. Figure 3 shows the horizontal distributions of reductions in the PM$_{2.5}$ concentration averaged over AP, defined as

\[
\frac{[\text{PM}_{2.5}]_{s} - [\text{PM}_{2.5}]_{\text{NoCTRL}}}{[\text{PM}_{2.5}]_{\text{NoCTRL}}} \times 100\%
\]

where $[\text{PM}_{2.5}]_{s}$ and $[\text{PM}_{2.5}]_{\text{NoCTRL}}$ are the simulated PM$_{2.5}$ concentrations in the sensitivity case and the NoCTRL simulation, respectively.
Emissions of NO$_x$, SO$_2$, OC, and BC were decreased by tough emissions control measures in Beijing and surrounding areas during AP (Liu et al. 2015). When control measures were carried out for the whole of NC (Figure 3(a)–(d)), the controls of OC and NO$_x$ led to the largest reductions in PM$_{2.5}$ concentrations in NC during AP. With emissions reductions of OC and NO$_x$ in NC, the average PM$_{2.5}$ concentration decreased by, respectively, by 5.7% and 4.6% in Beijing, and by 4.6% and 3.7% in NC. The reductions in SO$_2$ emissions in NC resulted in only 1%–4% reductions in the PM$_{2.5}$ concentration in NC, indicative of the success of the control of SO$_2$ emissions in recent years (Zhao, Zhang, and Nielsen 2013). Based on observations during periods when humans use domestic heating (i.e. in winter) of 2013 and 2014, Yang et al. (2016) reported that the concentrations of nitrate and SO$_4^{2-}$ were 9.89 and 4.90 μg m$^{-3}$, respectively, during 1–15 November 2014, and 23.08 and 16.68 μg m$^{-3}$, respectively, during 16–30 November 2014. The model results together with the observations suggest that the control of NO$_x$ emissions was important for reducing PM$_{2.5}$ concentrations in NC.

For emissions control measures conducted in NC-B (Figure 3(e)–(h)), the reductions in NO$_x$ and OC emissions resulted in the largest reductions (2.8%) in the PM$_{2.5}$ concentration in Beijing. With emissions control measures applied locally in Beijing (Figure 3(i)–(l)), the PM$_{2.5}$ concentration in Beijing was most sensitive to the control of OC. With OC and BC emissions reduced by 30% in Beijing, the average PM$_{2.5}$ concentration in Beijing during AP decreased by 2.9% and 2.3%, respectively, relative to the NoCTRL case. The reductions in the PM$_{2.5}$ concentration were within 2% in the cases of NO$_x$ or SO$_2$ control. Since emissions of OC and BC derive mainly from the residential sector (Li et al. 2015), these results from the sensitivity experiments indicate that the control of local residential emissions is important for reducing the PM$_{2.5}$ concentration in Beijing. Overall, the decreases in the PM$_{2.5}$ concentration in Beijing during AP were most sensitive to reductions in NO$_x$ and OC emissions in NC-B and the local reduction in emissions of carbonaceous aerosols.

We also conducted sensitivity experiments to examine the sensitivity of PM$_{2.5}$ concentrations to reductions in NH$_3$ emissions in the same regions (Figure 3(m)–(o)). With NH$_3$ emissions reduced by 30% in NC, the average PM$_{2.5}$ concentration in Beijing and NC decreased, respectively, by 14.8% and 14.7%, compared to the NoCTRL case, suggesting that PM$_{2.5}$ concentrations in Beijing and the surrounding provinces are all very sensitive to NH$_3$ emissions. As a result of the competing system of NH$_3$–nitrate–SO$_4^{2-}$, emissions reductions of SO$_2$ (NO$_x$) can reduce ammonium sulfate (nitrate) concentrations but increase ammonium nitrate (sulfate) concentrations in PM$_{2.5}$. With emission reductions of SO$_2$, NO$_x$, and NH$_3$ in NC, the NH$_3$ concentrations decreased by 2.8%, 6.0%, and 19.8%, respectively, in Beijing, indicating that the emissions control of NH$_3$ in NC is the most efficient in reducing NH$_3^+$ in PM$_{2.5}$. Since 90% of NH$_3$ emissions in China come from the agriculture (Li et al. 2015), NH$_3$ emissions can hardly be reduced with short-term emissions control measures. Therefore, it is essential to reduce NH$_3$ emissions from farmland and livestock wastes in the long term. Technologies, such as using polymer-coated urea products instead of the common use of nitrogen fertilizer, and optimizing feeding to avoid surplus protein in the animal diet, have been considered to be effective in reducing NH$_3$ emissions in western countries (Webb et al. 2008).

The results from the sensitivity simulations provide useful information on emissions reduction strategies in Beijing. In general, cooperating with surrounding provinces and cities could significantly improve the effectiveness of emissions control measures, compared to conducting control measures in Beijing alone. For short-term emissions control during wintertime, like in the APEC case, reducing local emissions of carbonaceous aerosols (OC and BC) and regional emissions of NO$_x$ and OC is the most efficient approach in reducing PM$_{2.5}$ concentrations in Beijing. For long-term emissions control strategies, reducing NH$_3$ emissions from agricultural sources is an effective way to improve the air quality in Beijing. However, decreases in NH$_3$ emissions may result in enhancement of aerosol acidity, since SO$_4^{2-}$ and nitrate may become insufficiently neutralized. Thus, NH$_3$ emissions should be carefully controlled in parallel with appropriate SO$_2$ and NO$_x$ controls in NC.

4. Conclusion

This work used a nested-grid capability of the global chemical transport model GEOS-Chem to simulate gases and aerosols to identify the chemical species whose reductions made the largest contribution to the decrease in PM$_{2.5}$ concentrations during the APEC period in Beijing. A series of numerical experiments were conducted to investigate the sensitivity of surface PM$_{2.5}$ concentrations to the reductions in emissions of SO$_2$, NO$_x$, OC, BC, and NH$_3$ from specific regions.

Results showed that the model successfully captured the daily variations of PM$_{2.5}$, CO, NO$_x$, and SO$_2$ in Beijing, despite the biases in concentrations, with correlation coefficients ranging from 0.44 to 0.83. The discrepancies in peaks between the model results and observations may have arisen from uncertainties in the emissions inventory for local emissions, the mismatch of the emissions base year and model year, and the differences between actual meteorological conditions (such as wind speed) and those in the model. The concentrations of PM$_{2.5}$ showed similar reductions during AP in both observations and the model simulation. The observed and simulated PM$_{2.5}$ concentrations...
during AP were, respectively, 55.9% and 57.4% lower than the values during BAP, and 57.3% and 58.5% lower than the values during AAP. By comparing simulations with and without emissions control measures, it was found that the emissions control measures led to a 23.4% reduction in the PM$_{2.5}$ concentration during the APEC period.

Considering the emissions control measures conducted during AP, the results from the sensitivity simulations showed that the control measures for NO$_x$ and OC over NC led to the largest reductions in PM$_{2.5}$ concentrations in Beijing. Compared to the control of SO$_x$ and NO$_x$, the control of NH$_3$ emissions was found to be able to greatly reduce the NH$_4^+$ concentration in PM$_{2.5}$ in the whole of NC. To examine the effectiveness of emissions control measures of various species for the APEC case, we set the emission reduction rates for NO$_x$, SO$_x$, OC, and BC according to the air quality evaluation report of the APEC period released by CNPEA. We also conducted sensitivity tests with same reduction percentages (−30%) for all the species in BJ, NC-B, and NC, respectively. The model results of the two designs of sensitivity tests produced the same major conclusions as described above.

Finally, it is important to highlight that this study focused mainly on the emissions control of primary organic carbon. In fact, secondary organic aerosols were also significantly reduced in AP due to the emissions control of NMVOCs (Sun et al. 2016). This aspect should be investigated in future studies.

**Disclosure statement**

No potential conflict of interest was reported by the authors.

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