Updated emission inventories of power plants in simulating air quality 
during haze periods over East China

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Abstract. Air pollutant emissions play a determinant role in deteriorating air quality. However, an uncertainty in emission inventories is still the key problem for modeling air pollution. In this study, an updated emission inventory of coal-fired power plants (UEIPP) based on online monitoring data in Jiangsu Province of East China for the year of 2012 was implemented in the widely used Multi-resolution Emission Inventory for China (MEIC). By employing the Weather Research and Forecasting Model with Chemistry (WRF-Chem), two simulation experiments were executed to assess the atmospheric environment change by using the original MEIC emission inventory and the MEIC inventory with the UEIPP. A synthetic analysis shows that power plant emissions of PM$_{2.5}$, PM$_{10}$, SO$_2$ and NO$_x$ were lower, and CO, black carbon (BC), organic carbon (OC) and NMVOCs (Non-methane volatile organic compounds) were higher in UEIPP relatively to those in MEIC, reflecting a large discrepancy in the power plant emissions over East China. In accordance with the changes of UEIPP, the modeled concentrations were reduced for SO$_2$ and NO$_2$, and increased for most areas of primary OC, BC and CO. Interestingly, when the UEIPP was used, the atmospheric oxidizing capacity significantly reinforced, reflecting by increased oxidizing agents, e.g. O$_3$ and OH, thus directly strengthened the chemical
production from SO$_2$ and NO$_x$ to sulfate and nitrate, which offset the reduction of primary PM$_{2.5}$ emissions especially in the haze days. This study indicated the importance of updating air pollutant emission inventories in simulating the complex atmospheric environment changes with the implications on air quality and environmental changes.

**Keyword:** Emission inventory; haze pollution; air quality modeling; secondary aerosols; oxidizing agents

1 Introduction

East China is one of the regions with serious air pollution and frequent haze. In these highly polluted regions, air pollutant emissions play a key role in air quality, and their variations can cause a large uncertainty in air pollution modeling and prediction. It is also crucial for air pollution mitigation to comprehensively understand air pollutant emissions and their impacts on atmospheric environment. Emission inventories are essential for atmospheric environment research, especially for modeling study and air quality policy making.

During past decades, emission inventories for China were established by several groups. These include the global-scale work, such as the Reanalysis of the Tropospheric chemical composition (RETRO) (Schultz, 2007; Zheng et al., 2009), the Hemispheric Transport of Air Pollution (HTAP) (Janssens-Maenhout et al., 2015) and the Emission Database for Global Atmospheric Research (EDGAR), and the national-scale studies including the Transport and Chemical Evolution over the Pacific mission (TRACE-P) (Olivier et al., 2005), the Intercontinental Chemical Transport Experiment-Phase B (INTEX-B) (Zhang et al., 2009), the Regional Emission inventory in Asia (REAS) (Ohara et al., 2007) and the Multi-resolution Emission Inventory for China (MEIC, http://www.meicmodel.org/). Owing to poor measurements, several of these studies were based on “top-down” algorithm, which rendered the uncertainties in estimation of emissions and subsequently decreased the accuracy in modeling atmospheric
environment. For example, previous studies showed a difference of 30% in CO emission among various emission inventories over East Asia, leading to an up to 8% simulated deviation (Amnuaylojaroen et al., 2014). Regional emission inventories were developed recently in China, for the regions of Yangtze River Delta (Huang et al., 2011; Fu et al., 2013), North China Plain (Wang et al., 2010) and Pearl River Delta (Zheng et al., 2009), as well as several provincial and urban areas (Zhao et al., 2015; Jing et al., 2016; He et al., 2016), with more underlying data for activity levels, emission factors, energy combustion and traffic database obtained.

Air pollution in East China is changing from coal-smoke to mix-source polluted type, particularly the secondary aerosols surging in severe haze episodes (Huang et al., 2014a), with more complicated chemical reactions involved in interaction of particle formation and O₃ production. As the largest coal-fired sector of emission framework in China, electric power generation is believed to be the most important source of atmospheric pollutant emissions (Zhao et al., 2010). The power plant emissions accounted for 31-59% of national anthropogenic emissions of SO₂ and 21-44% of NOₓ (Zhao et al., 2008; Wang et al., 2012). The pollutant emissions from coal-fired power plants were usually estimated by the widely adopted “bottom-up” approach (Hao et al., 2002; Zhang et al., 2007b; Zhang et al., 2007a; Ohara et al., 2007; Zhao et al., 2008). However, due to limited access to specific information about power plants, such as the mass of pollutant emitted per unit fuel consumption or per unit industrial production, coal-fired boiler types or accurate location of power plants (Wang et al., 2012), the inaccuracy of emissions from local power plants can be one of uncertainty sources to lead to model bias. The validations of power plant emissions with the impacts on atmospheric environment particularly in haze episodes are still a gap.

Jiangsu Province is one of the most developed areas in East China, providing living place for a
population of 79.2 million with the highest gross domestic production (GDP) per capita in China (NBSC, 2013a; JSNBS, 2013). Severe air pollution episodes of haze and photochemical pollution have shrouded this province in recent years (Fu et al., 2008; Wang et al., 2014; Qi et al., 2015). As elevated emission source, power plants emit air pollutants with longer life cycles in upper air and more efficiently regional transport because of less deposition driving by stronger winds and well organized circulation in upper air, such as by low-level jets (Hu et al., 2013), leading to more significant environmental effect than surface emissions (e.g. on-road emission), reflecting potential importance of accurately estimating the power plant emissions and their influences on air quality. Based on unit-based methodology, Zhao et al. (2008) developed an inventory of coal-consuming power plants for all the provinces in China. The annual SO$_2$ and NO$_x$ emissions were estimated with 1107 kilotons (kt) and 626 kt in 2005, as well as 803 kt and 781 kt in 2010 in Jiangsu Province. Alternately, another study estimated NO$_x$ emission in the province at about 748 kt in 2005 (Wang et al., 2012), reflecting the uncertainties in estimation of NO$_x$ emission from power plants. Source control measures focusing on power generation processes, and facility-related measures to reduce emissions have been widely implemented in China. These measures include flue gas desulfurization (FGD), selective catalytic reduction (SCR) and selective non-catalytic reduction (SNCR), dust collector, etc. It is important to assess the potential air quality changes from implementation of those mitigation measures.

Zhang et al. (2015) established an emission inventory of coal-fired power plants (UEIPP) by collecting the online monitoring data from power plants in atmospheric verifiable accounting tables of Jiangsu Province for 2012. The volumes of flue-gas and pollutant concentrations were measured in-site for each unit, providing the more realistic data for calculating power plant emissions than those used in previous studies. As a major objective of this study, the UEIPP was integrated into MEIC to evaluate the impact of emission update on regional atmospheric environment with an air quality model. We presented
the details of model settings, observational data and emission inventories in Sect. 2 and the modeling evaluation in Sect. 3. The impact of emission change on atmospheric environment and the underlying mechanism were discussed in Sect. 4. The study was summarized in Sect. 5.

2 Data and method

2.1 Model description and configuration

The period from November 29 to December 31, 2013 was chosen as the modeling period, covering a severe haze period (from 3 to 8 December 2013) in Jiangsu Province. The online coupled Weather Research and Forecasting Model with Chemistry (WRF-Chem in version 3.7.1) model was configured in three nesting domains with the horizontal resolutions respectively in 45 km covering most area of East Asia, 15 km covering Eastern China and surrounding areas, and 5 km covering Jiangsu Province and surrounding areas (Fig. 1a). Vertically, there were 35 full eta levels from surface up to 100 hPa with 7 levels below 1 km. The National Center for Environmental Prediction Final Global Forecast System operational analysis data (Kalnay et al., 1996) was utilized for providing the initial and lateral meteorological conditions to WRF-Chem. Grid nudging (Stauffer and Seaman, 1990) was employed for the outmost domain every 6 hours, treating temperature, horizontal wind, and water vapor, to guarantee the precision of large-scale meteorology during the simulations.

The selected physical configurations included Morrison double-moment microphysics scheme (Morrison et al., 2009), RRTMG (Rapid Radiative Transfer Model for GCMs (Global Climate Models)) long and short wave radiation scheme (Iacono et al., 2008), Grell 3D cumulus parameterization, Yonsei University planetary boundary layer scheme (Hong et al., 2006), and Noah land surface model. For chemistry and aerosol mechanism, the CBM-Z (Carbon Bond Mechanism; Zaveri, 1999) coupling with the 8-bin sectional MOSAIC (Model for Simulating Aerosol Interactions and Chemistry) model with
aqueous chemistry (Zaveri et al., 2008) was used. The MOSAIC, treating all the important aerosol components, including nitrate, sulfate, ammonium, black carbon, and primary organic aerosols and other inorganic aerosols, is efficient without compromising accuracy and widely used in air quality and regional/global aerosol models (Zaveri et al., 2008). Since the MOSAIC is incapable of simulating secondary organic aerosols (SOAs), the simulated organic aerosols mentioned hereinafter all refers to primary organic aerosols. The crucial processes of radiation feedback, aerosol and cloud interaction, dry deposition, wet scavenging and cloud chemistry were turned on. Biogenic emissions were calculated online with the MEGAN (Model of Emission of Gases and Aerosol from Nature) (Guenther et al., 2006). The initial and boundary chemistry conditions were based on the vertical profiles of O₃, SO₂, NO₂, VOCs and other air pollutants from the NOAA Aeronomy Lab Regional Oxidant Model (NALROM) (Liu et al., 1996). The first two-day simulation was discarded as the spin-up. The outmost domain of modeling tests was set large enough to cover East Asia to avoid the influence of chemical boundary conditions on simulation. Furthermore, the frequent haze pollution over Eastern China is resulted from the regional pollutant emissions (Wang et al., 2015; Zhang, 2015) with less contribution of foreign emission to haze pollution over Eastern China.

### 2.2 Observational data

Meteorological fields simulated by the model are crucial for the accuracy of air quality modeling. In the south, middle and north parts of Jiangsu Province, we selected three prefecture-level cities of Nanjing, Yancheng and Lianyungang, respectively, to evaluate the overall perspective of meteorological simulation with the available observations. The observed meteorological data, involving 2-m temperature, 2-m relative humidity and 10-m wind speed and direction was collected from the Jiangsu Provincial Meteorological Bureau and Meteorological Information Comprehensive Analysis and Process System.
(MICAPS) of China Meteorological Administration (CMA). Hourly surface concentrations of chemical species in 13 cities of Jiangsu, including SO$_2$, NO$_2$, PM$_{2.5}$, CO and O$_3$, were obtained from Jiangsu Environmental Protection Bureau. Daily secondary inorganic aerosols (SIAs; sulfate, nitrate, ammonium) in PM$_{2.5}$ were measured using MARGA (Online Analyzer of Monitoring of Aerosol and Gases) at Jiangsu Environmental Monitoring Center. MARGA is a fully autonomous sampling and measurement system that continuously measures the gases (HCl, HNO$_3$, HNO$_2$, SO$_2$, and NH$_3$) and aerosol components (CL$^-$, NO$_3^-$, SO$_4^{2-}$, NH$_4^+$, K$^+$, Ca$^{2+}$, and Mg$^{2+}$) by ion chromatography with internal standard eliminates calibration.

2.3 Air pollutant emission inventory

2.3.1 Two inventories for power plant emissions

This study utilized the MEIC inventory as the default anthropogenic emissions including the emissions of sulfur dioxide (SO$_2$), nitrogen oxides (NO$_x$), carbon monoxide (CO), NH$_3$, black carbon (BC), organic carbon (OC), non-methane volatile organic compounds (NMVOCs), PM$_{2.5}$, and PM$_{10}$ by five sectors of power, industry, transportation, residential, and agriculture.

The UEIPP in Jiangsu Province for the year of 2012 consisting of six online species (SO$_2$, NO$_x$, PM$_{2.5}$, PM$_{10}$, BC and OC), was established using the online monitoring data of three pollutants (SO$_2$, NO$_x$ and total suspended particles (TSP)) and volume of flue gases at unit and daily level (Zhang et al., 2015). Atmospheric verifiable accounting tables, comprising accurate locations, boiler type, coal consumption, and control policies for individual plant, were adopted to calculate the CO and NMVOCs emissions in UEIPP (Zhang et al., 2015).

The SO$_2$, NO$_x$ and TSP emissions were reckoned directly from online concentrations and volumes of
flue gases as follows:

\[ E_{i,j,k} = A_{i,j,k} \cdot c_{i,j,k}, \]  

where \( A_{i,j,k} \) is the daily emitted volume of flue gas, and \( c_{i,j,k} \) is the daily concentration, with \( i, j, \) and \( k \) representing the pollutant species, individual plant, and day, respectively. The emissions of PM\(_{2.5}\), PM\(_{10}\), BC and OC were then calculated using the online TSP emissions:

\[ E_{i,j,k} = T_{i,j,k} \cdot P_i, \]  

where \( T_{i,j,k} \) stands for the online TSP emissions; \( P_i \) represents the PM\(_{2.5}\), PM\(_{10}\), BC and OC mass ratios to TSP. The online monitoring system is currently incapable of providing the mass ratios at unit level and thus, it’s given as a unified value referring to the work of Zhang et al. (2006) for each the four species, which was 52.7\% (PM\(_{2.5}\)), 80.4\% (PM\(_{10}\)), 8.6\% (BC), and 6.1\% (OC), respectively.

The annual emissions of CO and NMVOCs were calculated using Eq. (3):

\[ E_{i,j,k} = A_{i,j,k} \cdot EF_{i,j,k} \cdot (1-\eta_{i,j,k}), \]  

where \( A_{i,j,k} \) is the activity level, \( EF_{i,j,k} \) is the uncontrolled emission factor, and \( \eta_{i,j,k} \) is the removal efficiency of air pollutant control device. In refer to previous studies (Wang et al., 2005; Streets et al., 2006; Bo et al., 2008; Huang et al., 2011), the \( EF_{i,j,k} \) of CO and NMVOCs was set at 4.03 g kg\(^{-1}\) and 0.12 g kg\(^{-1}\) respectively, and \( \eta_{i,j,k} \) was set to 0.

Following the method used by Li et al., (2014) and the mechanism-dependent mapping tables developed by Cater (2013), the NMVOCs in UEIPP were specified to individual constituent in Regional Acid Deposition Model chemical mechanism (RADM2, Stockwell et al., 1990), which could be adapted to the WRF-Chem/CBMZ mechanism used in this study. The primary distinction, between UEIPP and the
power plant emission inventory estimated in previous China studies, lies in the different data used and subsequently the estimation algorithm as well as the temporal resolution. Previously, power plant emission inventory was mostly estimated using various data such as activity levels, boiler types, fuel types, control policies and emission factors, and the activity levels were usually collected at annually or monthly level. In the UEIPP, the emissions of SO$_2$, NO$_x$, PM$_{2.5}$, PM$_{10}$, BC, and OC were calculated using the online pollutant concentrations and volume of flue gases at daily level. Rejection heights of the UEIPP and the original power plant emission in MEIC were set at about 100 m and 200 m above ground, corresponding to the second and third model levels for this WRF-Chem simulation.

### 2.3.2 Differences between two power plant emission inventories

The UEIPP and MEIC power plant emissions of major air pollutants with their fractions in the total emissions over Jiangsu Province in 2012 were presented in Table 1 for contrastive analysis. Appreciable differences between the two power plant emission inventories were revealed. The power plant emissions of SO$_2$, PM$_{2.5}$, PM$_{10}$ and NO$_x$ in the MEIC are 367.8 kt, 72.2 kt, 103.7 kt, and 733.8 kt with reducing to 105.6 kt, 21.6 kt, 32.6 kt and 277.9 kt respectively in the UEIPP. The notable reductions of SO$_2$ and NO$_x$ may largely due to comprehensive implementation of FGD and SCR/SNCR in Jiangsu Province, which was not captured in national inventory. Application rate and average SO$_2$ removal efficiency of FGD in coal-fired power plants were obviously higher than those in other sectors (Zhou et al., 2016), further confirmed by abrupt decrease of SO$_2$ power plant emission in China since 2006 (Liu et al., 2015). Differences also existed in estimation of NO$_x$ removal efficiency of SCR/SNCR for power plants in Jiangsu in 2012 among different studies, reflecting 37 % (average of SCR/SNCR) calculated by Zhou et al. (2016) while 70 % (SCR) and 25 % (SNCR) calculated by Tian et al. (2013). In addition, due to inconsistent penetration rates and removal efficiencies of dust collectors determined at national and
provincial levels, there also remained discrepancies in estimation of PM$_{2.5}$ and PM$_{10}$ emissions (Xia et al., 2016; Zhou et al., 2016). However, the bias could be avoided in the UEIPP (see Section 2.3.1 for detail).

The UEIPP produced higher CO (582.0 kt), BC (3.6 kt), OC (2.5 kt) and NMVOCs (17.3 kt) emissions compared to the MEIC. The power plants in the MEIC present the very low emissions of BC and OC, particularly for OC with 0.0 kt, resulting largely from the high uncertainties in the emission factor of these species (Zhao et al., 2015; Zhao et al., 2011; Zhou et al., 2016). Furthermore, the MEIC power plant emissions of SO$_2$, PM$_{2.5}$, PM$_{10}$ and NO$_x$ shared with the larger fractions of 28.8 %, 11.3 %, 11.6 % and 35.4 %, relatively to CO, BC, OC and NMVOCs with the fractions of 3.7 %, 0.2 %, 0.0 % and 0.4 %, in the total emissions (Table 1). When the UEIPP was introduced to MEIC by replacing the original power plant emission, the UEIPP contributed 10.4 %, 3.7 %, 4.0 %, 17.2 %, 6.2 %, 4.3 %, 1.7 % and 0.9 %, to the total emissions of SO$_2$, PM$_{2.5}$, PM$_{10}$, NO$_x$, CO, BC, OC, and NMVOCs, respectively. The ratios of PM$_{2.5}$ (3.7 %) and PM$_{10}$ (4.0 %) of the UEIPP were comparable to the ratios of 4 % for PM$_{2.5}$ and 6 % for PM$_{10}$ calculated by Zhou et al. (2016).

The spatial difference of two emission inventories over Jiangsu Province was shown in Figure 2, as well as their absolute values in Figure S1. The UEIPP presented the low emissions of SO$_2$, PM$_{2.5}$, PM$_{10}$ and NO$_x$ in the most areas (Fig. 2a-d), and the high emissions of CO, BC, OC and NMVOCs in urban areas over the province (Fig. 2e-h). Two inventories exhibited the similar spatial distribution patterns with large emissions in southland and low emissions in midland and northland (Fig. S1). The power plants around Xuzhou, an industrial city, formed a high emission center over the northwestern Jiangsu (Fig. S1).

To assess the simulation performance with the UEIPP and changes of atmospheric environment over Jiangsu Province under the updated emission condition, two simulations with the original MEIC emission inventory (hereinafter referred as MOD1 simulation) and the updated MEIC emission inventory with the
power generation replaced by UEIPP (hereinafter referred as MOD2 simulation) were carried out. The difference of chemical components between MOD1 and MOD2 simulations were used to assess atmospheric environmental changes in the following sections.

3 Modeling evaluation

3.1 Meteorological evaluation

An evaluation of the meteorological simulations over the domain with 5 km horizontal resolution was carried out in respects of temperature, relative humidity (RH), wind speed and direction in Nanjing, Yancheng and Lianyungang. The evaluation statistical parameters included mean bias (MB), correlation coefficient (R) and root mean square error (RMSE) (Table 2). The R and RMSE of temperature in the three cities ranged from 0.86 to 0.94 (p-values < 0.001) and 2.0 °C to 3.0 °C, showing a close agreement between the simulation and observations. MB values of temperature manifested a slight underestimate in Nanjing (-1.0 °C) and Lianyungang (-0.5 °C), and overestimate in Yancheng (1.1 °C). The R of RH was 0.79, 0.79 and 0.82 (p-values < 0.001), with the RMSE values of 14.6%, 14.4% and 18.2%, respectively in Nanjing, Yancheng and Lianyungang, which were comparable to previous studies (Gao et al., 2016; Liu et al., 2016). The MB of RH was positive in Nanjing and Lianyungang, but negative in Yancheng. The variations of wind speed were generally captured by the model with the R varying from 0.51 to 0.77 (p-values < 0.001). The RMSE ranging from 1.8 m s\(^{-1}\) to 2.1 m s\(^{-1}\), basically conforming to the “good” model performance criteria for wind speed (Less than 2.0 m s\(^{-1}\); Emery et al., 2001). Wind directions were evaluated via Hit Rates (HR; Schlünzen and Sokhi, 2008) with desired accuracy between ±45°. The HR values were 63 %, 64 % and 49 %, respectively in Nanjing, Yancheng and Lianyungang, indicating that variations of wind direction were basically captured. Generally, the meteorological fields in Jiangsu Province were reasonably reproduced by WRF-Chem during the simulation period.
3.2 Chemical evaluation

The surface observations of PM$_{2.5}$, CO, NO$_2$, O$_3$ and SO$_2$ at 13 urban sites in Jiangsu (Fig. 1b) were collected for evaluating the chemical simulation over the domain with 5 km horizontal resolution in MOD1 and MOD2. Three statistical parameters of $R$, mean fractional bias (MFB) and mean fractional error (MFE) were presented in Table 3. MFB and MFE could normalize bias and error for simulated-observed pair ranging from -200% to 200%, and from 0% to 200% respectively, indicating their appropriateness to evaluate performance over a wide range of concentrations (Boylan and Russell, 2006). Normalized mean bias (NMB) and normalized mean error (NME) by individual site and air pollutant were additionally presented in Table S1 in the Supplement. As shown in Table 3, the values of MFB and MFE indicated that the hourly variations of PM$_{2.5}$, CO and NO$_2$ were reasonably captured by both MOD1 and MOD2 simulations, conforming to the “satisfactory” criteria proposed by Morris et al. (2005) that MFB is within ±60 % and MFE is below 75 %. Given a high dependence on emissions, the deviations of CO and NO$_2$ concentrations could be largely resulted from their emission uncertainties. The high $R$ and the negative MFB of O$_3$ indicated the hourly variations were reasonably captured but undervalued systematically, especially at night (Fig. 9b; Fig. S2). The CBM-Z scheme and the outdated land-use data from United States Geological Survey (USGS) were prone to undervalue the surface O$_3$ concentrations in association with producing high NO-titration and dry deposition, respectively (Balzarini et al., 2015; Park et al., 2014). Similar underestimations were previously simulated in Eastern China (Gao et al., 2015; Liao et al., 2015; Wang et al., 2016). The mean NMB and NME of O$_3$ in MOD1, calculated at -53.97 % and 67.00 % respectively (Table S1), were comparable to previous China studies (Li et al., 2012; Tang et al., 2015; Wang et al., 2016; Zhou et al., 2016), while the mean NMB and NME of O$_3$ in MOD2 were ameliorated respectively to -45.83 % and 63.61 %. The SO$_2$ changes were generally captured in the two simulations in terms of MFB and MFE, but with an overestimation and the low $R$. In addition of
emissions, absence of pathways converting SO$_2$ to sulfate in current WRF-Chem model, such as aqueous phase oxidation of dissolved S$_{\text{IV}}$ (the sum of hydrated SO$_2$ (SO$_2$ \cdot H$_2$O), bisulfite (HSO$_3^-$), and sulfite (SO$_3^{2-}$)) by dissolved NO$_2$ under conditions of high ammonia (NH$_3$) and NO$_2$ concentrations (Huang et al., 2014b; Xue et al., 2016; He et al., 2014), was partially responsible for the simulation deviations of SO$_2$ and NO$_2$. Aerosols in East Asia are featured with low acidity due to the high NH$_3$ and mineral dust emissions capturing more acidic gases (SO$_2$ and NO$_x$) under moist air conditions during haze episodes (Cheng et al., 2016; Wang et al., 2016). The modeled SIAx at Nanjing site (Fig. 1b) were assessed in addition. As can be seen from Table 4 and Fig. S3a, the simulated sulfate concentrations were obviously underestimated, providing a further evidence for the above-mentioned speculation. Similar underestimation of sulfate was also found in North China Plain (Gao et al., 2016a, 2016b). The observed nitrate and ammonium concentrations were comparatively well caught, particularly the NMBs of nitrate ranged within ±20 % in the two simulations. In general, the chemical observations were reasonably captured by both simulations of MOD1 and MOD2.

The R, MFB and MFE in the MOD1 and MOD2 simulations were presented in Table 3 with an overall assessment of simulation with the UEIPP. A better simulation performance is reflected by higher R, smaller absolute value of MFB and lower MFE, respectively tagged with upward arrows in Table 3. Additionally, significance of the improvements between the statistical indices was checked via using the method of bootstrap confidence interval (DiCiccio and Efron, 1996; He et al., 2017). In response to the introduction of UEIPP, chemical simulation showed a comprehensive improvement in MOD2 (see the upward arrows in Table 3). Although both MOD1 and MOD2 underestimated PM$_{2.5}$, CO, O$_3$ and overestimated NO$_2$ and SO$_2$ as shown in Table 3, the MFBs for those species reduced by 0.07, 0.21, 10.78, 3.6, and 8.26 % respectively from MOD1 to MOD2. The improvements of O$_3$, SO$_2$ and NO$_2$ were statistically significant (Table 3). For the O$_3$ simulation, the improvements of the R, MFB and MFE
significantly passed the confidence level of 99%. For the SO\textsubscript{2} simulation, the improvements of MFB and MFE passed the confidence levels of 99 % and 90 %, respectively. Improvement in MFB of NO\textsubscript{2} was significant at 95 % confidence level. Also, the modeled SIAs were ameliorated in MOD2 simulation (Table 4). Under the unchanged meteorology between two simulations, the reduced deviations of NO\textsubscript{2}, SO\textsubscript{2}, CO, PM\textsubscript{2.5} and O\textsubscript{3} in MOD2 relatively to MOD1 could be attributed to the emission changes with UEIPP. However, PM\textsubscript{2.5} and O\textsubscript{3} are highly dependent on secondary formation, indicating the changes of chemical conversion in the simulations of MOD1 and MOD2, which was more comprehensively investigated in Section 4.

Spatially, overestimates of SO\textsubscript{2} in MOD1 mainly occurred in south urban areas and Xuzhou (Fig. 3a), where the SO\textsubscript{2} overestimates were mostly improved in MOD2 (Fig. 3k, f). For NO\textsubscript{2} simulation, the overestimates lay in the majority cities throughout Jiangsu Province with a few cities underestimated such as Suzhou (Fig. 3b), and were mitigated correspondingly in MOD2 as well (Fig. 3l, g). As a common feature of MOD1 simulation, CO, PM\textsubscript{2.5} and O\textsubscript{3} were undervalued throughout the most city sites (Fig. 3c-e), while in response to the usage of UEIPP, their concentrations in MOD2 were comprehensively improved (Fig. 3m-o, h-j).

As mentioned above, the MOD2 with the introduction of UEIPP, improved the simulation of air pollutants, especially of O\textsubscript{3}, SO\textsubscript{2}, and NO\textsubscript{2} according to model performance evaluation. It is therefore concluded that the power plant emissions of UEIPP is more realistic.

4 Environment changes under updated emission condition

4.1 Influence of emission changes on air pollutant modeling

Aside from introducing the updated emission inventories, another important and meaningful work in
this study is to explore how the emission changes affect the atmospheric environment especially in severe haze episodes for understanding the complexity of atmospheric environment. To this end, we presented the differences of atmospheric compositions simulated with MOD1 and MOD2 in Figure 4. Consistently with the emission changes (Fig. 2), the concentrations were reduced for SO$_2$ and NO$_2$ (Figs. 3k and 3l), and enhanced for BC and OC (Figs. 4c and 4d) in the most areas. However, it’s not clear yet what was responsible for the enhanced O$_3$ concentration in MOD2, and why the PM$_{2.5}$ concentration enhanced when the PM$_{2.5}$ primary emission largely reduced, which were urgently needed to address especially for the abnormal increase of PM$_{2.5}$, since more implemented restrictions on power plants are being executed in East China.

As a secondary air pollutant in the boundary layer, O$_3$ is highly subjected to its precursors (Ou Yang et al., 2012; Gao et al., 2005). The Yangtze River Delta was characterized of VOC-limited, especially in winter, indicating O$_3$ concentrations were depressed by NO$_x$ and sensitive to VOC (Liu et al., 2010; Wang et al., 2008; Tie et al., 2013). The ratio of HCHO/NO$_y$, an indicator to differentiate VOC-limited (HCHO/NO$_y$ < 0.28) and NO$_x$-limited (HCHO/NO$_y$ > 0.28) conditions, verified the Jiangsu Province as the VOC-limited region during the modeling period (Fig. 6). Therefore, either the increase of VOC or the decrease of NO$_x$ could enhance the surface O$_3$ level. Coincidentally, the concentrations of VOC and NO$_2$ increased and decreased respectively (Fig. 4h and 3l), following their emission changes in UEIPP (Table 1). In addition, a high anti-correlation relationship existed between the spatial changes of O$_3$ and NO$_2$ (Figs. 3l and 3o) as well as the diurnal changes (Figs. 9a-b). Therefore, we could partially attribute the underestimated O$_3$ to the emission uncertainties of VOC and NO$_2$ in the original MEIC. Furthermore, as a precursor of O$_3$, high CO concentrations in MOD2 with the updated MEIC emission inventory would contribute to the enhancement of O$_3$ concentration as well.
Quite surprising to us, the surface PM$_{2.5}$ concentrations didn’t follow the reducing emissions of primary PM$_{2.5}$, but increased over almost all the province (Fig. 3n). As PM$_{2.5}$ is highly dependent on three factors of primary emissions, physical processes and chemical reactions in the atmosphere, the latter two factors were more likely to dominate in the simulation of increased PM$_{2.5}$ concentrations. Due to the strong absorbing effects of BC to solar radiation and the higher rejection height of power plants, the enhancement of BC concentrations (Fig. 4c; Table 5) would reduce solar radiation to ground, and thus suppressing vertical diffusion and accumulating more pollutants near surface. The speculation could be verified by reductions of downward short wave flux at ground surface (SWDOWN), 2 m air temperature and boundary layer height (BLH). Regionally averaged over Jiangsu Province during Dec. 1-31, the SWDOWN, 2 m temperature and BLH reduced by 0.65 W m$^{-2}$, 0.005 $^\circ$C and 0.4 m respectively, and the reductions became more significant to 11.8 W m$^{-2}$, 0.3 $^\circ$C and 26.4 m respectively, during the daytime of Dec. 7, 2013 in Wuxi, a haze center, increasing air stability for more air pollutant accumulation. In order to quantify the radiative effects induced by BC emission change, a sensitivity test MODa as same as MOD2 with closing BC emission in UEIPP was performed. Based on the PM$_{2.5}$ differences between MOD2 and MODa regionally averaged over Jiangsu Province, it was estimated that the BC aerosol radiative effect stabilizing boundary layer contributed only about 0.15 µg m$^{-3}$ to the PM$_{2.5}$ enhancements, during the haze episode.

4.2 Reinforcing atmospheric oxidation capacity and enhancing secondary inorganic aerosols

As described in Sect 4.1, the declined emissions of primary PM$_{2.5}$ could not improve the ambient PM$_{2.5}$ concentration and the feedback aerosol radiative effects on aerosol change were weak, implying the contribution of chemical production to the PM$_{2.5}$ enhancement. In this section, the chemical production of SIAs was analyzed, since the CBM-Z/MOSAIC used in the WRF-Chem is incapable of...
simulating SOA formation. Previous studies had revealed that SIAs played an important role in PM$_{2.5}$, particularly in the haze pollution over Eastern China (Huang et al., 2014a; Wang et al., 2014; Gao et al., 2016). Given the reduction of SO$_2$ and NO$_x$ emissions in the UEIPP, the simulated sulfate and nitrate should be lower from the oxidation of SO$_2$ and NO$_2$. However, as shown in Fig. 4, both sulfate and nitrate are increased in MOD2, with more significantly during haze episode (Dec.3-8; Fig. 5). In the atmosphere, sulfate is formed through oxidation of SO$_2$ by gas-phase reactions with OH (Stockwell and Calvert, 1983; Blitz et al., 2003) and stabilized Criegee intermediate formed by O$_3$ and alkenes (Mauldin III et al., 2012) as well as by heterogeneous reactions with H$_2$O$_2$, O$_3$, OH, organic peroxides, and various oxides of nitrogen in clouds (Seinfeld and Pandis, 2012). Nitrate is mostly formed from the gas-phase reactions of NO$_2$ with OH during daylight and heterogeneous reactions of nitrate radical (NO$_3$) at night. Therefore, the formations of secondary sulfate and nitrate depend not only on their precursors, but also on oxidizing capacity of atmosphere.

As major oxidizing agents in atmosphere, O$_3$ and OH were increased from MOD1 to MOD2 (Fig. 3o and 4b), indicating the enhanced oxidizing capacity in MOD2 relatively to MOD1 in Jiangsu region. A WRF-Chem/RADM2 simulation was performed as well because changes in OH and VOC oxidation in the presence of NO$_x$ are sensitive to chemistry mechanisms (Derwent, 2017; Knote et al., 2015; Stockwell et al., 2011; Jimenez et al., 2003). The similar pattern of increasing O$_3$ and OH were found over the province (Fig. S4), which could further indicate the enhanced oxidizing capacity.

To evaluate how the formation of secondary aerosols responded to the enhanced oxidizing capacity, we analyzed the BC-scaled concentrations for sulfate and nitrate. The purpose was to eliminate the influence of air pollutant dilution and mixing in atmospheric physical process. Since BC is quite inertial to chemical reactions, its variations could well reflect the atmospheric physical processes. Thus, the BC-
scaled concentration could better represent the contribution of chemical reaction (Zheng et al., 2015).

Figure 7 presents the daily averaged variation of BC-scaled concentrations for sulfate and nitrate, appending the differences of O\textsubscript{3} as an indicator for the change of atmospheric oxidizing capacity. As revealed in Fig. 7, the enhancements of chemical production simulated in MOD2 were consistent well with the variations of O\textsubscript{3} difference between MOD2 and MOD1. During the haze episode over Dec. 3-8, 2013 the chemical production of sulfate and nitrate enhanced obviously, which in accordance with the rapid build-up of O\textsubscript{3}, indicating the chemical production is intensified by strengthen of oxidizing capacity during the episode. It is worth pointing out that photochemical activity is generally reduced by weakening solar radiation from high aerosol levels during severe haze period in China. Our modelling study also confirmed the relatively low ozone concentrations during the haze period from December 3-8, 2013 in Jiangsu province, and furthermore found the UEIPP led to enhanced atmospheric oxidizing capacity during the haze periods in East China, which could be resulted from changing photochemical oxidation of VOC, NO\textsubscript{x} and SO\textsubscript{2} from regional industrial sources (Guo et al., 2014). Large particle mass growth was observed concurrently with elevated daily ozone concentrations throughout the PM\textsubscript{2.5} episodes in China, indicating the importance of photochemical activity in the secondary aerosol production (Guo et al., 2014).

The SOR (molar ratio of sulfate to sum of sulfate and SO\textsubscript{2}) and NOR (molar ratio of nitrate to sum of nitrate and NO\textsubscript{2}) were used as indicators of secondary transformation (Sun et al., 2006), since the BC-scaled concentrations just represent the intensity of chemical reaction effects with overlooking the precursors for individual compounds. The SOR and NOR would give insights to the chemical transformation of SO\textsubscript{2} and NO\textsubscript{2}. As shown in Figure 8, the chemical transformations from SO\textsubscript{2} and NO\textsubscript{x} to sulfate and nitrate were always strengthened during the whole month in MOD2, that could be why the chemical production of sulfate and nitrate in MOD2 increased (Figs. 7a-b) even with less precursor
concentrations. Additionally, in response to the enhanced atmospheric oxidizing agents, the secondary ammonium was also increased (Fig. 4f) under the same NH₃ emission conditions in MOD1 and MOD2.

As shown in Table 5, the total concentration of sulfate, nitrate and ammonium increased by 1.32 µg m⁻³ during the whole month of Dec. 1-31, 2013 and even reached up to 4.77 µg m⁻³ in the haze episode of Dec. 3-8, 2013, higher than the increments of PM₂.₅ as well as the total increments of BC and OC, which could clearly reveal that the enhancement of SIAs in response to the reinforced atmospheric oxidizing capacity contributed the majority to the increased PM₂.₅ concentrations. This conclusion was verified by an emission sensitivity study in the North China Plain performed by Wang et al., (2016), who found that the 30 % emission reduction of NOₓ led to a notable increase in PM₂.₅ concentrations contributing to NH₃-rich and VOC-limited conditions in the winter.

It should interpret the larger enhancement in concentrations of SIAs than the PM₂.₅ in Table 5. Compared to the MOD1, the lower emission of primary PM₂.₅ in UEIPP (Table 1) lead to the less concentrations of primary PM₂.₅ in MOD2, and the enhancement in concentrations of SIAs was partially offset by the lower primary concentrations of PM₂.₅.

5 Conclusions

Power plant, as a major air pollutant source in China, had been imposed restrictions by the government in response to the increasing air pollution, which led power plant emissions to large variations during past few years. Due to various underlying data and approaches, there remained uncertainties in estimating the power plant emissions. In the present study, the UEIPP in Jiangsu Province for 2012 was introduced in the MEIC emission inventory as the major point sources. The variation and complexity of atmospheric environment in response to the change of power plant emissions over Jiangsu were studied, by executing the WRF-Chem simulations using the original emissions of MEIC and the MEIC with its
The study focused on the uncertainty in estimating the power plant emissions. In the UEIPP, the emission amounts of $SO_2$, $PM_{2.5}$, $PM_{10}$, NO$_x$, CO, BC, OC and NMVOCs were 105.6 kt, 21.6 kt, 32.6 kt, 277.9 kt, 582.0 kt, 3.6 kt, 2.5 kt, and 17.3 kt, respectively, manifesting obvious difference with the MEIC emission inventory. Uncertainties in meteorology (MET) and chemistry (CHEM), another two major uncertainties in numerical simulations of fine urban PM were excluded with optimally selected meteorological and chemical settings, based on the meteorological and chemical validations of the MOD1 simulation comparing to the observations. There are the challenges for the development of atmospheric chemical models with the MET, especially in PBL-parameterizations, such as the key role of aging of BC in the interaction between pollution and PBL (Peng et al., 2016) and CHEM including enhanced aqueous chemistry during the development of severe haze in China (Zhang et al., 2015). Further modeling work could consider these MET and CHEM issues based on the new understandings of atmospheric physical and chemical processes.

The UEIPP drove the simulation performance superior to the original power plant emission of MEIC inventory in terms of the proximity between simulated and observed air pollutant concentrations, suggesting a more realistic power plant emission inventory. The complexity of atmospheric environment was revealed as well, through comparing the changes of various primary and secondary compositions in atmosphere. Atmospheric oxidizing capacity was reinforced in response to the enhancement of O$_3$ and OH, which was largely due to higher VOC emissions and lower NO$_x$ in the UEIPP. $PM_{2.5}$ increased almost all over the Jiangsu Province even the primary emission reduced by 7.6%. This anomaly phenomenon was mainly attributed to the enhanced formation of SIAs, induced by the enhanced atmospheric oxidizing capacity, revealing the complex mechanism of air pollution from fine particulate
matter to atmospheric oxidants. Reduction of SO\(_2\) may free NH\(_3\) to react instead with NO\(_x\) creating ammonium nitrate particles, which would need further studying. Our study also quantified the PM\(_{2.5}\) enhancement in response to BC radiative effect stabilizing boundary layer. The chemical reaction was more dominant in PM\(_{2.5}\) enhancement than the BC radiative effect.

Given the complicated processes in environmental change, the restrictions of emissions should be comprehensively considered rather than one single factor. Furthermore, the effects of emission inventories on air quality variations could be assessed based on long-term observation and simulation studies, and formation of SOAs would be also enhance due to the reinforced atmospheric oxidizing capacity and higher VOC emissions.

6 Acknowledgements

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Table 1 The UEIPP and MEIC power plant emissions of major air pollutants as well as the ratios in total emission inventory over Jiangsu Province in 2012.

<table>
<thead>
<tr>
<th></th>
<th>UEIPP Emission (kt year$^{-1}$)</th>
<th>Ratio in total (%)</th>
<th>MEIC power plant emission Emission (kt year$^{-1}$)</th>
<th>Ratio in total (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>SO$_2$</td>
<td>105.6</td>
<td>10.4</td>
<td>367.8</td>
<td>28.8</td>
</tr>
<tr>
<td>PM$_{2.5}$</td>
<td>21.6</td>
<td>3.7</td>
<td>72.2</td>
<td>11.3</td>
</tr>
<tr>
<td>PM$_{10}$</td>
<td>32.6</td>
<td>4.0</td>
<td>103.7</td>
<td>11.6</td>
</tr>
<tr>
<td>NO$_x$</td>
<td>277.9</td>
<td>17.2</td>
<td>733.8</td>
<td>35.4</td>
</tr>
<tr>
<td>CO</td>
<td>582.0</td>
<td>6.2</td>
<td>343.5</td>
<td>3.7</td>
</tr>
<tr>
<td>BC</td>
<td>3.6</td>
<td>0.1</td>
<td>0.6</td>
<td>0.2</td>
</tr>
<tr>
<td>OC</td>
<td>2.5</td>
<td>1.7</td>
<td>0.0</td>
<td>0.0</td>
</tr>
<tr>
<td>NMVOCs</td>
<td>17.3</td>
<td>0.9</td>
<td>7.2</td>
<td>0.4</td>
</tr>
</tbody>
</table>

Table 2 Statistics between observed and simulated meteorology.

<table>
<thead>
<tr>
<th></th>
<th>Nanjing</th>
<th>Yancheng</th>
<th>Lianyungang</th>
</tr>
</thead>
<tbody>
<tr>
<td>T (°C)</td>
<td>Obs.</td>
<td>Mod.</td>
<td>MB</td>
</tr>
<tr>
<td>4.8</td>
<td>3.8</td>
<td>-1.0</td>
<td>0.86</td>
</tr>
<tr>
<td>RH (%)</td>
<td>63.0</td>
<td>63.6</td>
<td>0.6</td>
</tr>
<tr>
<td>WS (m s$^{-1}$)</td>
<td>2.0</td>
<td>3.1</td>
<td>1.1</td>
</tr>
</tbody>
</table>

* T: temperature at 2m; RH: relatively humidity at 2m; WS: wind speed at 10m; MB: mean bias; R: correlation coefficient; RMSE: root mean square error; all R values passed $p < 0.001$.

Table 3 Statistics variables between observed and simulated PM$_{2.5}$, CO, NO$_2$, O$_3$ and SO$_2$.

<table>
<thead>
<tr>
<th></th>
<th>MOD1</th>
<th>MOD2</th>
<th>MOD1</th>
<th>MOD2</th>
<th>MOD1</th>
<th>MOD2</th>
</tr>
</thead>
<tbody>
<tr>
<td>R</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>PM$_{2.5}$</td>
<td>0.568</td>
<td>0.571 ↑</td>
<td>-8.31</td>
<td>-8.24 ↑</td>
<td>45.40</td>
<td>45.65</td>
</tr>
<tr>
<td>CO</td>
<td>0.516</td>
<td>0.515 ↑</td>
<td>-36.05</td>
<td>-35.84 ↑</td>
<td>52.05</td>
<td>52.10</td>
</tr>
<tr>
<td>NO$_2$</td>
<td>0.456</td>
<td>0.466 ↑</td>
<td>14.08</td>
<td>10.48 ↑ **</td>
<td>39.37</td>
<td>38.77 ↑</td>
</tr>
<tr>
<td>O$_3$</td>
<td>0.600</td>
<td>0.625 ↑ ***</td>
<td>-121.46</td>
<td>-110.68 ↑ ***</td>
<td>131.74</td>
<td>124.67 ↑ ***</td>
</tr>
<tr>
<td>SO$_2$</td>
<td>0.260</td>
<td>0.261 ↑</td>
<td>24.88</td>
<td>16.62 ↑ ***</td>
<td>65.18</td>
<td>63.20 ↑ *</td>
</tr>
</tbody>
</table>

* all R values passed $p < 0.001$; Up arrows indicate the chemical simulation results in MOD2 are improved; "**", "***" and "****" indicate the improvements are statistically significant with confidence level at 99 %, 95 % and 90%, respectively. Equations of R, MFB and MFE were provided in Supplement.

Table 4 Validation statistics of SIAs in PM$_{2.5}$ at Nanjing.

<table>
<thead>
<tr>
<th></th>
<th>NMB (%)</th>
<th>NME (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>MOD1</td>
<td>MOD2</td>
<td>MOD1</td>
</tr>
<tr>
<td>Sulfate</td>
<td>-87.61</td>
<td>87.61</td>
</tr>
<tr>
<td>Nitrate</td>
<td>-19.47</td>
<td>29.06</td>
</tr>
<tr>
<td>Ammonium</td>
<td>-55.12</td>
<td>55.12</td>
</tr>
</tbody>
</table>

* Up arrows indicate the same meaning as in Table 3. Equations of NMB and NME were provided in Supplement.
Table 5 Increased concentrations (µg m⁻³) of PM₂.₅, total of BC and OC, and total of SOA (sulfate, nitrate and ammonium) from MOD1 to MOD2 averaged over Jiangsu Province with the percentage differences between MOD1 and MOD2 compared with MOD1.

<table>
<thead>
<tr>
<th></th>
<th>PM₂.₅</th>
<th>BC+OC</th>
<th>Sulfate+Nitrate+Ammonium</th>
</tr>
</thead>
<tbody>
<tr>
<td>Hazy days (Dec.3-8)</td>
<td>3.38 (1.81 %)</td>
<td>1.04 (3.06 %)</td>
<td>4.77 (5.11 %)</td>
</tr>
<tr>
<td>The whole (Dec.1-31)</td>
<td>1.03 (0.83 %)</td>
<td>0.58 (2.38 %)</td>
<td>1.32 (3.96 %)</td>
</tr>
</tbody>
</table>
Figure 1 Model domains and the locations of 13 cities in Jiangsu Province.

Figure 2 Differences of power plant emission between MEIC and UEIPP in 2012 (units: tons).

Figure 3 The spatial distributions of near-surface SO$_2$, NO$_x$, CO, PM$_{2.5}$ and O$_3$ concentrations ($\mu$g m$^{-3}$) from (a-e) MOD1, (f-j) MOD2 and (k-o) the differences between MOD2 and MOD1 averaged over December 2013. The observed concentrations were indicated by shaded circles.
**Figure 4** Differences of chemical species between MOD2 and MOD1 in December 2013. Unit: “pptv” for OH; “ppmv” for VOC; “µg m^{-3}” for the others.

**Figure 5** Chemical species of PM$_{2.5}$ simulated in MOD1 (left bar of each bar pair) and MOD2 (right bar of each bar pair) in December 2013.

**Figure 6** Ratios of HCHO/NO$_y$ simulated in MOD1 (blue) and MOD2 (red).
Figure 7 Daily variations of (a) sulfate/BC, (b) nitrate/BC and (c) difference of O$_3$ (MOD2 - MOD1) averaged over Jiangsu with the red rectangular column marking the severe haze episode (Dec. 3-8); the increase of sulfate/BC and nitrate/BC suggests enhanced chemical productions.
Figure 8 Daily variations of (a) SOR and (b) NOR.

Figure 9 Diurnal variations of (a) NO$_2$ and (b) O$_3$ averaged over 13 cities in Jiangsu Province (Fig. 1b).