Reviewer 1:

This study developed the unit-based industrial emission inventory in Beijing-Tianjin-Hebei region, for which configurations and locations of individual industrial emission sources were utilized. Significant differences in horizontal distributions of emissions were seen by comparing with the traditional proxy-based emission inventory. The air quality simulations using this unit-based emission inventory showed better model performance than the proxy-based emission inventory.

I think this is an important progress to get better model performance. It should contribute to developing effective emission controls against heavy air pollution in this region. However, various critical information is missing in the current manuscript. It is necessary to revise it based on the comments described below.

Response: We appreciate the reviewer's valuable comments which help us improve the quality of the manuscript. We have carefully revised the manuscript according to the reviewers' comments. Point-to-point responses are given below. The original comments are in black, while our responses are in blue.

(1) As mentioned in the introduction, previous studies have already developed unit-based emission inventories while their target sectors may be limited. I suppose there should be more papers including Liu et al. (2015) for example. It is necessary to clearly describe what is new in this study. This manuscript says previous studies did not cover all industrial sectors in the BTH region. Then, does this study cover all industrial sectors? Which sectors were newly included? Is the methodology identical for the sectors which have been already included in previous studies? Significance of this study should be described more clearly.

Response: We thank the reviewer for this valuable comment. We searched the papers about unit-based emission inventories again and added more papers in the Introduction section, including Liu et al. (2015) about emission from coal-fired power plants, Chen et al. (2015) about emission from cement industry and Wu et al. (2015) about emission from steel industry. (Page 3, Line 13-16) In the previous studies, they usually focus on one or several sectors such as power plant, cement plant, and iron plant. In this study, we cover most industrial sectors including power plant, industrial boiler, iron and steel production, non-ferrous metal smelter, coking, cement, glass, brick, lime, ceramics, refinery, and chemical industries (Page 4, Line 3-5). Compared with most previous studies, industrial boiler, non-ferrous metal smelter, coking, glass, brick, lime, ceramics, refinery, and chemical industries are newly included. The methodology of calculating the emission of point sources is similar to previous studies, but we calculate the emissions from cement and iron sectors according to specific industrial processes, such as clinker burning and clinker processing stages in the cement sector (Page 4, Line 18 to Page 5, Line 5).

(2) One of difficulties in unit-based emission inventories we often face is consistency of energy consumption against energy statistics. Did this study use energy consumption reported from each emission source? If so, is the sum of the reported energy consumption consistent with that in energy statistics? Usually, it is very hard to collect detailed information of small emission sources. If this is the case, energy consumption should not be consistent, and a hybrid approach in which unit-based and proxy-based information are combined may be necessary for each sector. The unit-based and proxy-based emission inventories were compared in this study. Do energy consumptions used in both inventories match?

Response: Yes, this study calculated emissions using energy consumption or industrial production reported for each emission source.

The plants in this study are from compilation of power industry statistics (China Electricity Council, 2015), China Iron and Steel Industry Association (http://www.chinaisa.org.cn), China Cement Association (http://www.chinacca.org), Chinese environmental statistics (collected from provincial environmental protection bureaus), the first national census of pollution sources (National Bureau of Statistics (NBS), 2010) and bulletin of desulfurization and denitrification facilities from Ministry of Ecology and Environment of China (http://www.mee.gov.cn). (Page 5, Line 6-13)

We compared the sum of the energy consumption or industrial production for each plant with those in official statistics. The sum of individual plants generally accounts for over 90% of the energy consumption or product yield reported in the statistics. For the plants not included in the preceding data sources, we calculate the emission by using "top-down method" and allocate the emission with proxies, such as GDP and population. Therefore, the total energy consumption of both inventories match. (Page 6, Line 14-17; Page 7, Line 25-26)

(3) Although detailed descriptions for vertical distributions are missing in the current manuscript, I agree that reasons of differences in concentrations between the unit-based and proxy-based emission inventories should be horizontal distributions and vertical distributions as mentioned in the second paragraph in the page 9. According to Figures 5 and 7, concentrations simulated with the proxy-based emissions are almost entirely lower throughout the domain. If influences of horizontal distributions are dominant, it is supposed that concentrations in surrounding regions would become higher, but such influences seem to be very limited. Therefore, it might be possible that differences in concentrations between two emission inventories are mainly caused by differences in vertical distributions of emissions. I would strongly recommend conducting an additional simulation to separate influences of horizontal and vertical distributions of emissions by changing only each of them.

Response: We thank the reviewer for this valuable comment. We have conducted an additional simulation in which the unit-based inventory is used but the emission heights are assumed to be the same as the proxy-based inventory. The amount of emission is the same as the other two scenarios. We call the inventory used in this simulation “hypo unit-based inventory”.

Fig. R1 (Fig. 5 in the revised manuscript) shows the distribution of the monthly (January and July) mean concentrations of SO_2, NO_2, ozone, daily maximum 1-h averaged ozone, daily maximum 8-h averaged ozone and $PM_{2.5}$ simulated with the proxy-based inventory, and the differences between the proxy-based simulation and the other two simulations (Diff1: hypo unit-based minus proxy-based; Diff2: unit-based minus proxy-based). For SO_2, NO_2 and $PM_{2.5}$, the concentrations in the urban area are generally higher with the proxy-based inventory than those with the unit-based inventory, especially in winter. In January, large concentration differences between simulations with two inventories are found in urban Tianjin, Tangshan, Baoding and Shijiazhuang, where a large amount of industrial emissions is allocated in the proxy-based inventory due to large population density. The simulation of July follows the same pattern but the concentrations and the difference between the concentrations with two inventories are lower than those of January. In some areas where many factories are located, such as the northern part of Xingtai city, the concentration with unit-based inventory is higher because of a high emission intensity. There are two reasons for the difference between results with proxy-based and unit-based inventories. The first one is the spatial distribution. With detailed information of industrial sectors, more emissions are allocated to certain locations in suburban/rural areas in the unit-based emission inventory. From “Diff1” (hypo unit-based minus proxy-based), we can see that the improved horizontal distribution of the unit-based emission inventory significantly decreases the $PM_{2.5}$, SO_2, and NO_2 concentrations in most urban centers, and significantly increases the concentrations in a large fraction of suburban and rural areas, especially the areas where large industrial plants are located in. The other reason is vertical distribution. Plume rise is calculated in the simulation with the unit-based inventory, which causes the difference of emissions in vertical layers. The higher the pollutants are emitted, the lower the ground concentration becomes. From the differences between Diff1 and Diff2 we can see that the plume rise leads to lower concentrations over the whole region.

The results of the additional simulation have been added to the revised manuscript (Page 11, Line 6 to 26; Page 14, Line 2-4)

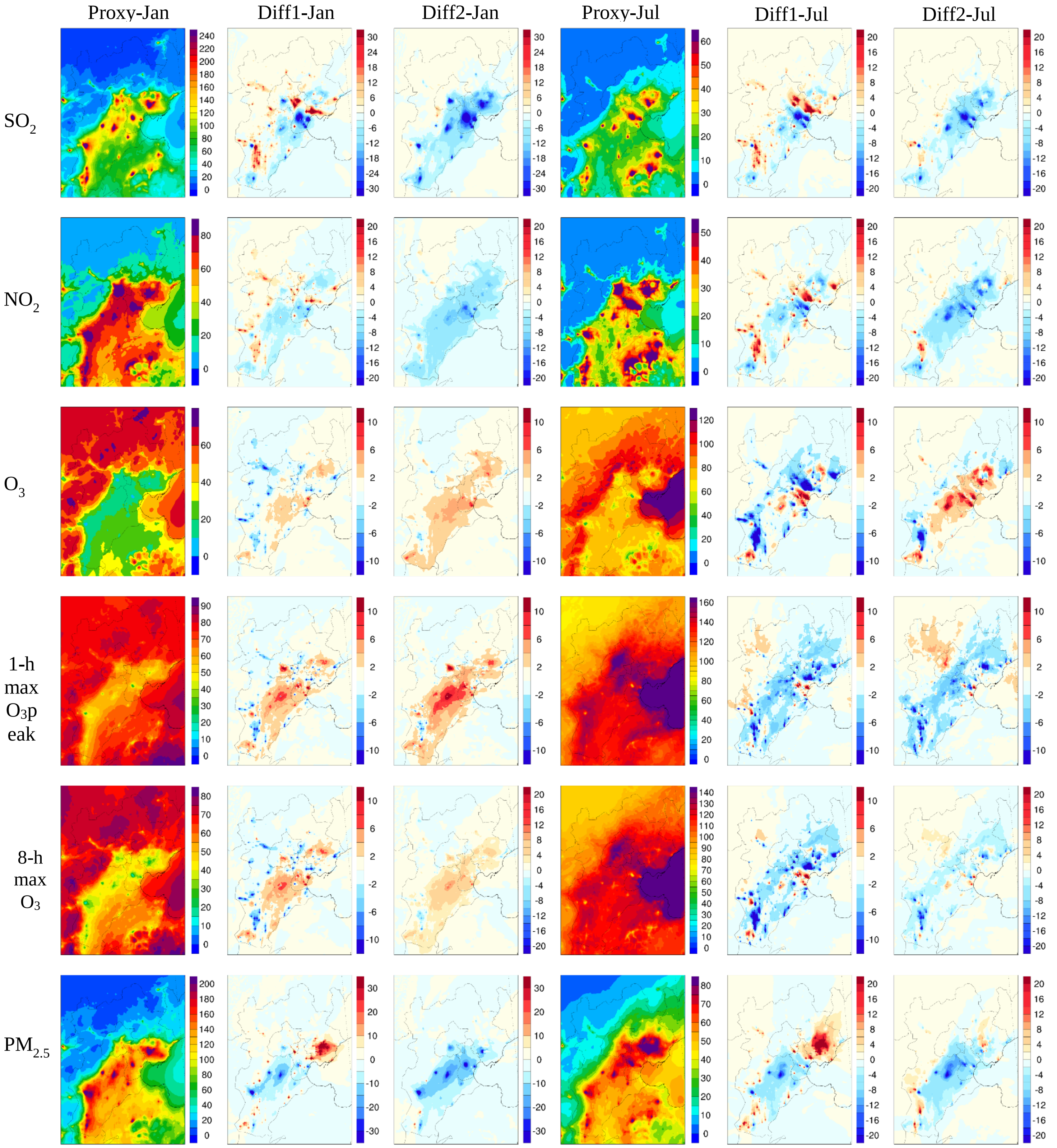

Fig. R1 Spatial distribution of the monthly (January and July) mean concentrations of SO_2, NO_2, ozone, daily maximum 1-h averaged ozone, daily maximum 8-h averaged ozone and $PM_{2.5}$ simulated with the proxy-based inventory, and the differences between the proxy-based simulation and the other two simulations (Diff1: hypo unit-based minus proxy-based; Diff2: unit-based minus proxy-based). The units are $\mathbf{\mu g/m^3}$ for all panels.

(4) This paper shows relative improvements in the unit-based emission inventory by comparing with the proxy-based emission inventory. Therefore, relative changes depend not only on the unit-based inventory but also the proxy-based inventory. If poor proxies are used in the proxy-based inventory, relative improvements could become larger. Therefore, it is important to explicitly show which proxies were used in the proxy-based inventory for each sector (not just "such as population …" at the end of the section 2.2). Use of better proxies should be also one of possible directions to get better model performance.

Response: For the proxies of each sector, we refer to Zhao et al. (2013), Streets et al. (2003) and Woo et al. (2003). We allocate the emissions of each province and each pollutant by two steps. The first step is to allocate the total emission to each county. The second step is to allocate the emission of each county to each grid. The proxies used in this study are shown in Table R1 (Table S2 in the revised manuscript).

Table R1 Proxies used in the proxy-based inventory for each sector

Sector	Allocate to county	Allocate to grid
Power plant, steel, cement	GDP of secondary industry	Population density
Industrial combustion, other industrial process	GDP of secondary industry	Population density
Domestic fuel	Total GDP	Population density
Domestic biomass	GDP of first industry	Population density
Transportation	GDP of tertiary industry	Road network
Open burning	GDP of first industry	Population density
Livestock	GDP of first industry	Population density
Fertilizer application	GDP of first industry	Population density
Domestic solvent use	Total GDP	Population density
Industrial solvent use	GDP of secondary industry	Population density

(5) Page 3, Line 9-10

I think that Lim et al. (2005) is not related to the description around here.

Response: It is removed from the manuscript.

(6) Page 3, Line 17-18

It is not clear which sectors are considered in previous studies and which sectors newly appear in this study. I would recommend adding a table listing all the industrial sectors considered and which are new in this study.

Response: As is shown in Table R2. The underlined sectors are newly added to this study. This table is added to SI. (Table S3)

Table R2 Comparison of industrial sectors covered in previous studies and this study (the underlined sectors are newly included in this study).

Study	Sector	Region
Zhao et al. (2008), Chen et al. (2014), Liu et al. (2015), Li et al. (2017)	Power plants	China
Wang et al. (2016b), Wu et al. (2015)	Iron plants	China

Lei et al. (2011), Chen et al. (2015)	Cement plants	China
Qi et al. (2017)	Power plants, iron plants, cement factories, coking factories, heating plants, other industries	BTH
This study	Power plants, iron plants, cement factories, coking factories, nonferrous metals, glass factories, brick factories, lime factories, ceramics factories, refinery factories, chemical plants, industrial boilers	BTH

(7) Page 4, Line 6-7

It is not clear what kind of product yields are used for estimating emissions of each sector. I would recommend showing types of products used for each sector in a table I recommended above.

Response: The types of products used for each sector are listed as follows and in Table S4 of the revised manuscript.

Table R3 Types of products or energy consumption used for estimating emissions of each sector.

Industrial sector	**Product or energy consumption**
Power plant	Energy consumption
Industrial boiler	Energy consumption
Iron and steel production	Pig iron, crude steel, rolled steel
Non-ferrous metal smelter	Alumina, aluminum, copper
Coking	Coke
Cement	Cement, clinker
Glass	Glass
Brick	Brick
Lime	Lime
Ceramics	Ceramics
Refinery	Crude oil, ethylene
Chemical industries	Ammonia, caustic soda, soda ash, sulfuric acid, nitric acid

(8) Page 4, Lines 9 and 17

The equation (1) is used to estimate emissions of the pollutant i. The industrial enterprise j and the production process m appear in this equation, but they are summed up. Then, how about the control technology n? It is not summed up, but it does not appear in the left-hand side. Usually fractions of control technologies are inserted, then they are summed up for all of control technologies. This is the same for the control technology k in the equation (2).

Response: Equation (1) and equation (2) are revised as follows:

$$\boldsymbol{E_{i,j}} = \boldsymbol{A_j} \times \boldsymbol{EF_{i,j}} \times \left(\boldsymbol{1} - \boldsymbol{\eta_{i,j}}\right) \qquad (1)$$

where $\boldsymbol{E_{i,j}}$ is emissions of pollutant i from industrial enterprise j, $\boldsymbol{A_j}$ is activity level of industrial enterprise j, $\boldsymbol{EF_{i,j}}$ is uncontrolled emission factor of pollutant i from industrial enterprise j, and $\boldsymbol{\eta_{i,j}}$ is removal efficiency of pollutant i by control technology in enterprise j. $\boldsymbol{\eta_{i,j}}$ is determined by the production process and control technology of the industrial enterprise. The $\boldsymbol{EF_{i,j}}$, which depends on the production process of the industrial enterprise, are calculated according to the sulfur and ash contents of fuels (e.g. coal) used in each province (for PM and SO_2), or obtained from our previous study (Zhao et al., 2013) (for other pollutants).

For those industrial sources with multiple production processes, such as iron and steel production and cement production, emissions are calculated by using the following equation:

$$\boldsymbol{E_{i,j}} = \sum\nolimits_{m} \left(\boldsymbol{AK_{j,m}} \times \boldsymbol{EF_{i,m}} \times \left(\boldsymbol{1} - \boldsymbol{\eta_{i,j,m}}\right)\right) + \left(\boldsymbol{AC_j} \times \boldsymbol{ef_i} \times \left(\boldsymbol{1} - \boldsymbol{\eta_{i,j}}\right)\right) \qquad (2)$$

where $\boldsymbol{E_{i,j}}$ is emissions of pollutant i from industrial enterprise j, $\boldsymbol{AK_{j,m}}$ is the amount of clinker produced by the clinker burning process m of the enterprise j, $\boldsymbol{EF_{i,m}}$ is uncontrolled emission factor for pollutant i from the clinker burning process m, $\boldsymbol{\eta_{i,j,m}}$ is removal efficiency of pollutant i from the clinker burning process m in enterprise j, $\boldsymbol{AC_j}$ is the amount of cement produced by enterprise j, $\boldsymbol{ef_i}$ is uncontrolled emission factors from the clinker processing stage ($\boldsymbol{ef_i}$=0 if i is not particulate matter), $\boldsymbol{\eta_{i,j}}$ is removal efficiency of pollutant i in enterprise j. $\boldsymbol{\eta_{i,j,m}}$ and $\boldsymbol{\eta_{i,j}}$ both depend on the control technology of the industrial enterprise. (Page 4, Line 10 to Page 5, Line 2)

(9) Page 4, Lines 9 and 17

I do not understand why the equations (1) and (2) are separated. It seems the first and second terms of the equation (2) represent clinker and cement production, respectively. However, isn't it possible to treat both as one of production processes m? If not, then what are production processes considered in both equations? Please clarify them. In fact, it is not clear what production processes considered in this study are.

Response: Equations (1) and (2) cannot be merged because the production processes represented by the first and second terms of equation (2) are frequently performed in different enterprises. For example, for cement production, clinker may be produced in one enterprise and subsequently processed in another enterprise, which is very common.

Most industrial sources are calculated by equation (1). Only a few industrial sources with multiple processes, such as steel production and cement production, are calculated by equation (2). We have added the preceding descriptions in the revised manuscript (Page 4, Line 18-19; Page 5, Line 3-5).

(10) Page 4, Lines 12-14

EFs depend only on the pollutant i and the production process m. Is there any possibility to use emission factors specific to each industrial enterprise? Is it enough to use identical emission factors for all the industrial enterprises?

Response: For SO_2 and PM, EFs are calculated according to the sulfur and ash contents of fuels (e.g. coal) in each province. For other pollutants, EFs depend only on the pollutant and the production process, and are obtained from our previous studies (Zhao et al., 2013). (Page 4, Line 14-17)

We agree with the reviewer that it is better to use emission factors specific to each individual enterprise. However, such detail offline emission measurements are not yet available in China. The continuous emission monitoring systems (CEMS) data may help to improve the emission esitmates. Cui et al. (2018) estimated the emissions of air pollutants from power plants in China based on the data of CEMS, environmental statistics and the data of pollutant emission permits. (Karplus et al., 2018) evaluated the impact of China’s new air pollution standards on SO_2 emissions by comparing newly available data from CEMS with satellite measurements. We will work on it in the future.

(11) Page 4, Line 25 – Page 5, Line 1

Specific references are not listed here while a lot of specific references for proxy-based emissions are listed in a subsequent paragraph. Specific references should be also listed for unit-based emissions as much as possible.

Response: The references were added in this manuscript.

For all power and industrial sources except industrial boilers, we collect their detailed information, including latitude/longitude, annual product, production technology/process, and pollution control facilities from compilation of power industry statistics (China Electricity Council, 2015), China Iron and Steel Industry Association (http://www.chinaisa.org.cn), China Cement Association (http://www.chinacca.org), Chinese environmental statistics (collected from provincial environmental protection bureaus), the first national census of pollution sources (National Bureau of Statistics (NBS), 2010) and bulletin of desulfurization and denitrification facilities from Ministry of Ecology and Environment of China (http://www.mee.gov.cn). (Page 5, Line 6-13)

(12) Page 5, Lines 2-4

Do these numbers cover all the plants located in the target area?

Response: It's very difficult to cover all the plants located in Beijing-Tianjin-Hebei region because there are some very small factories. The plants in this study are from compilation of power industry statistics (China Electricity Council, 2015), China Iron and Steel Industry Association (http://www.chinaisa.org.cn), China Cement Association (http://www.chinacca.org), Chinese environmental statistics (collected from provincial environmental protection bureaus), the first national census of pollution sources (National Bureau of Statistics (NBS), 2010) and bulletin of desulfurization and denitrification facilities from Ministry of Ecology and Environment of China (http://www.mee.gov.cn). The sum of individual plants generally accounts for over 90% of the energy consumption or product yield reported in the statistics. For the plants not included in the preceding data sources, we calculate the emission by using “top-down method” and allocate the emission with proxies, such as GDP and population. (Page 5, Line 6-13; Page 6, Line 14-18; Page 7, Line 25-26)

(13) Page 5, Line 5

Is there no information on control technologies for boilers?

Response: We do have information on control technologies for boilers. This sentence was revised as: “For industrial boilers, we obtained the location, fuel use amount, and control technologies of over 8 thousand industrial boilers…” (Page 5, line 16-19)

(14) Page 5, Line 9

Is the expression “emission factor method” appropriate? The unit-based approach also uses emission factors. I think it is usually called as “top-down method” (but sometimes confused with top-down estimates utilizing observations including satellites).

Response: “emission factor method” was replaced with “top-down method” in this sentence. (Page 6, Line 7)

(15) Page 5, Line 17

How about speciation of PM2.5 and NMVOCs for unit-based emissions?

Response: As is described in the manuscript, the speciation of $PM_{2.5}$ is from Fu et al. (2013). The $PM_{2.5}$ speciation profile of major sectors is shown in Fig. R2 (Fig. S4 in the manuscript). The speciation of NMVOCs is updated by Wu et al. (2017). The speciation profiles used in the unit-based inventory is the same as those used in the proxy-based inventory. The manuscript is revised accordingly. (Page 6, Line 18-20)

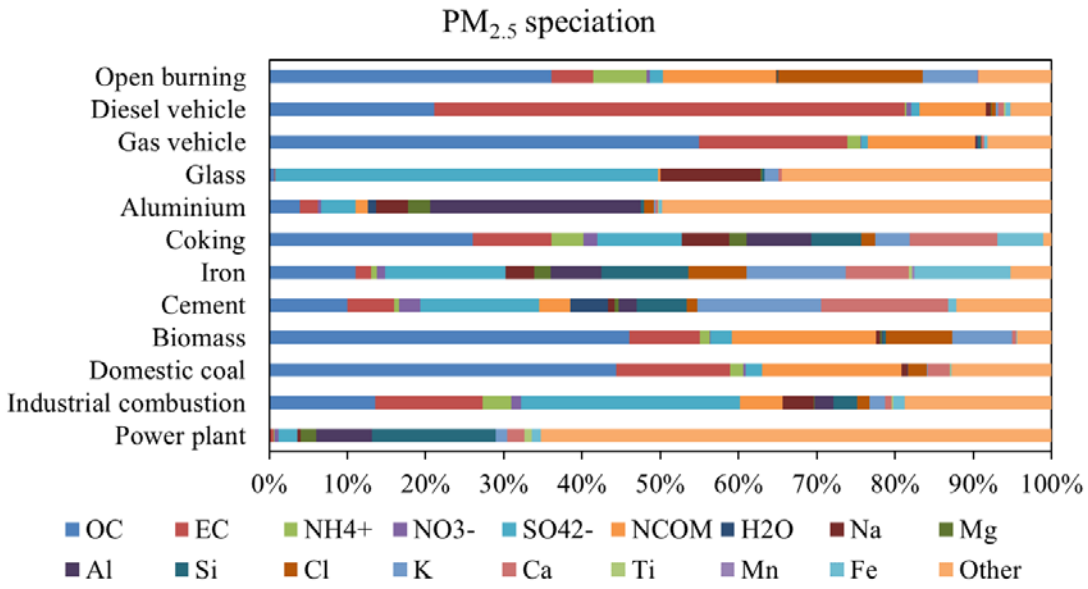

Fig. R2 $PM_{2.5}$ speciation profile of major sectors

(16) Page 5, Line 20 – Page 6, Line 18
References for models and modules are required.
Response: References are added to the manuscript. The revised text is shown as follows: In this work, we use CMAQ version 5.0.2 (EPA, 2014) to simulate the concentration of pollutants. (Page 6, Line 23-24) The Carbon Bond 05 (CB05) and AERO6 (Sarwar et al., 2011) are chosen as the gas-phase and aerosol chemical mechanisms, respectively. (Page 7, Line 5-7)
We use the Weather Research and Forecasting (WRF) model version 3.7.1 (Skamarock et al., 2008) to simulate the meteorological fields. The physics options for the WRF simulation are the Kain-Fritsch cumulus scheme (Kain, 2004), the Morrison double-moment scheme for cloud microphysics (Morrison et al., 2005), the Pleim-Xiu land surface model (Xiu and Pleim, 2001), Pleim-Xiu surface layer scheme (Pleim, 2006), ACM2 (Pleim) boundary layer parameterization (Pleim, 2007), and Rapid Radiative Transfer Model for GCMs radiation scheme (Mlawer et al., 1997). (Page 7, Line 9-14)

(17) Page 6, Line 10
What are “other” configurations? Please show explicitly.
Response: “Other” configurations means the initial and boundary conditions. The meteorological initial and boundary conditions are generated from the Final Operational Global Analysis data (ds083.2) of the National Center for Environmental Prediction (NCEP) at a 1.0° × 1.0° and 6-h resolutions. Default profile data is used for chemical initial and boundary conditions. It is revised accordingly in the manuscript. (Page 7, Line 14-17)

(18) Page 6, Lines 21-23
Is CO not included in this study? Why?
Response: The ambient CO pollution is not a serious issue in China currently. According to China National Environmental Monitoring Centre (data source: http://106.37.208.233:20035/), the daily CO concentration in the BTH region is less than 1.5 mg/m^3, which is much lower than the national ambient air quality standard (4 mg/m^3). In addition, the influence of CO emission on the formation of $PM_{2.5}$ and O_3 is quite small. For these two reasons, we did not include CO emission in this study.
In the model simulations described in this paper, we used CO emissions developed by Janssens-Maenhout et al. (2015).

(19) Page 7 Lines 1-22
Area names are mentioned in these paragraphs. However, horizontal distributions firstly appear later in Fig. 3. Its description should appear before descriptions of areas.
Response: The sequence of these sentences has been adjusted. (Page 8, Line 12-13)

(20) Page 7, Line 6

It is impossible to see many industrial boilers in Fig. 2.
Response: The link was wrong and we revised it to “Fig. 3”. (Page 8, Line 12-13)

(21) Page 8, Line 9
I think that NMB and NME are not appropriate metrics in terms of this study. The target of this study is accurate horizontal distributions. However, overestimation in one areas and underestimation in other areas could be cancelled out in these metrics. It is necessary to appropriate metrics which can properly shows improvements realized in this study.
Response: Thank you for this valuable comment.
While the overestimation and underestimation in different areas could be cancelled out in normalized mean bias (NMB), they cannot be cancelled out in the normalized mean error (NME), which characterizes the absolute difference between observation and simulation. Similarly, mean fractional error (MFE) is also an index that will not cancel out the overestimation and underestimation. The NME and MFE for SO_2, NO_2, $PM_{2.5}$, and O_3 are mostly lower with the unit-based inventory than with the proxy-based inventory, which means that the spatial distributions of these pollutants are better captured using the unit-based inventory. (Page 12, Line 5-8)
A major difference between the proxy-based and unit-based inventories is that the traditional proxy-based inventory allocates more emission to the urban area, whereas the unit-based inventory allocates more emission to suburban area where more factories are located. To quantify the impact of changed emission distribution between urban and suburban areas, we introduced the metric of “concentration gradient”, which is defined as the ratios of urban concentrations to suburban concentrations. The concentration gradients simulated with the unit-based inventory agree much better with observations than those simulated with the proxy-based inventory, implying that the unit-based emission inventory better reproduces the distributions of pollutant emissions between the urban and suburban areas. (Page 12, Line 5-22)
In addition, most of the observational sites (70 out of 80) are located in urban area. (Page 9, Line 19-20) Therefore, the calculated NMB is dominated by the behavior of the urban sites, and is not likely to be significantly cancelled out by the limited suburban sites.
Finally, we have shown the model performance for major air pollutants at each individual site in Beijing in the revised Supplementary Information (Table S6-S9). For the urban sites, the concentrations of $PM_{2.5}$, SO_2 and NO_2 are much lower with the unit-based inventory than with the proxy-based inventory. For the suburban sites, however, the concentrations are either slightly higher or slightly lower with the unit-based inventory than with the proxy-based inventory. The situation for ozone is quite the opposite. The ozone concentration at urban sites is higher with the unit-based inventory than with the proxy-based inventory. In suburban sites, it is lower with the unit-based inventory than with the proxy-based inventory. In addition, for the simulations with the unit-based inventory, the NME and MFE of individual sites are usually lower than those with the proxy-based inventory while the correlation efficient is usually higher, which means that the error is generally smaller and the trend is more

similar to the observation when the unit-based inventory is used.

(22) Page 8, Lines 15-17

What is a possible reason for the poor model performance on SO2?

Response: The overestimation of SO_2 concentrations may be due to the lack of several SO_2 reaction mechanisms in CMAQ, such as heterogeneous reactions of SO_2 on the surface of dust particles (Fu et al., 2016), the oxidation of SO_2 by NOx in aerosol liquid water (Cheng et al., 2016;Wang et al., 2016a), the effects of SO_2 and NH_3 on secondary organic aerosol formation (Chu et al., 2016), etc.

The biased spatial distribution of SO_2 emissions from residential combustion may also contribute to the overestimation. A large fraction of residential combustion takes place in the rural areas. In this work, however, the emission of residential combustion is allocated by GDP and population, which leads to an overestimation of SO_2 emission in urban area and hence an overestimation of SO_2 concentration. (Page 10, Line 4-11)

(23) Page 9, Lines 19-20

I cannot find any descriptions on plume rise before here. How to gather stack information? How to calculate plume rise? These descriptions are required in the method section.

Response: The stack information required for plume rise calculation includes stack height, flue gas temperature, chimney diameter and flue gas velocity. For power plants, we get the stack height from Compilation of power industry statistics (China Electricity Council, 2015). For the stack height of cement factories, we refer to the emission standard of air pollutants for cement industry (Ministry of Environmental Protection of China, 2013). For the stack height of glass, brick, lime and ceramics industries, we refer to emission standard of air pollutants for industrial kiln and furnace (Ministry of Environmental Protection of China, 1997). For the stack height of non-ferrous metal smelter, coking, refinery and chemical industries, as well as the flue gas temperature, chimney diameter and flue gas velocity for all industrial sectors, we refer to the national information platform of pollutant discharge permit (http://114.251.10.126/permitExt/outside/default.jsp), where we can find very detailed information of the plants with the pollutant discharge permit. For the sources without the pollutant discharge permit, we use the parameters of the plant with a similar production output or coal consumption. (Page 5, Line 23 to Page 6, Line 5) The data source of stack information is shown in Table R4 (Table S5 in the manuscript).

Table R4 Data source of stack information

Sector	Stack height	Flue gas temperature, Chimney diameter, Flue gas velocity
Power plant	Compilation of power industry statistics	National information platform of pollutant discharge permit
Cement plant	Emission standard of air pollutants for cement industry	

Glass, brick, lime and ceramics industries	Emission standard of air pollutants for industrial kiln and furnace
Non-ferrous metal smelter, coking, refinery and chemical industries	National information platform of pollutant discharge permit

Plume rise is calculated with a built-in algorithm of CMAQ based on the Briggs's scheme (Briggs, 1982). In this algorithm, plume rise is estimated by simulating the buoyancy effect and momentum rise, using hourly and gridded meteorological data. Then, the plume is distributed into the vertical layers that the plume intersects based on the pressure in each layer. (Page 5, Line 20-22; Page 8, Line 3-5)

(24) Page 10, Line 1
Details of "concentration gradient" are necessary. How to select urban and suburban locations? Are monthly mean concentrations used?
Response: For Beijing, the suburban areas refer to the districts that are far from the center of the city (the red star in Fig. 2). From Fig. 2 we can see that there are 8 sites located in the urban districts in Beijing. In the north, there are four sites far away from the city center and close to the city border. We treat the four sites in the north as suburban sites and the others as urban sites. For Tianjin, as shown in Fig. 3, there are two city centers. Ten sites are located in urban area and 5 sites are located in suburban area. In the calculation of the concentration gradient, monthly mean concentrations are used (Page 12, Line 10). These figures are added to the Supplementary Information. (Fig. S4-S5)

Fig. R3 The observational sites in Beijing

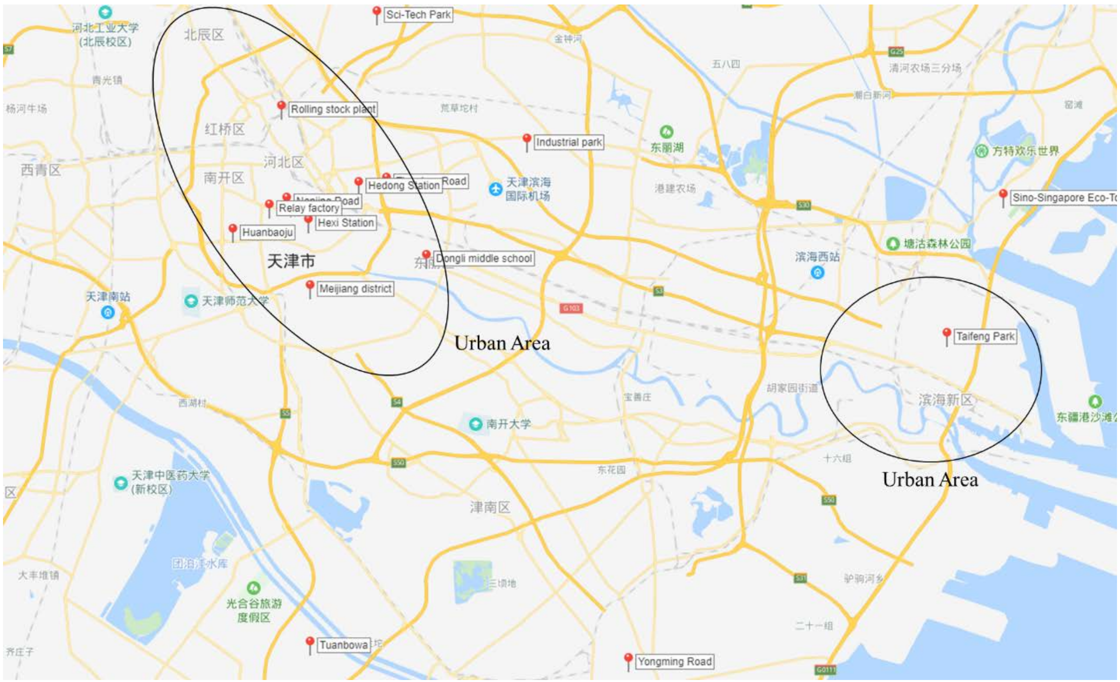

Fig. R4 The observational sites in Tianjin

(25) Page 10, Lines 24-27

I think it is not enough to explain changes of NO3- only by NOx sensitivities. I do not think they are main reasons. SO42- concentrations in the unit-based approach are much lower than the proxy-based approach whereas NH4+ is almost constant as shown in Fig.

7. In this case, more HNO3 is converted to NO3- with excess NH4+ whereas these processes depend on abundance of HNO3 or NH3.

Response: Thank you for the valuable idea. We have added this reason to explain the changes of NO_3^- as follows:

As for nitrate, concentration of nitrate in the simulation with unit-based inventory is much higher than that with proxy-based inventory in winter while the differences between the results with two inventories vary with location in summer. Sulfate concentrations in the unit-based approach are much lower than the proxy-based approach. In this case, more abundant NH_3 is available to react with HNO_3, leading to enhanced formation of NO_3^-. (Page 13, Line 11-14)

References:

Briggs, G. A.: Plume Rise Predictions, in: Lectures on Air Pollution and Environmental Impact Analyses, edited by: Haugen, D. A., American Meteorological Society, Boston, MA, 59-111, 1982.

Chen, L., Sun, Y., Wu, X., Zhang, Y., Zheng, C., Gao, X., and Cen, K.: Unit-based emission inventory and uncertainty assessment of coal-fired power plants, Atmos Environ, 99, 527-535, 10.1016/j.atmosenv.2014.10.023, 2014.

Chen, W., Hong, J., and Xu, C.: Pollutants generated by cement production in China, their impacts, and the potential for environmental improvement, Journal of Cleaner Production, 103, 61-69, 10.1016/j.jclepro.2014.04.048, 2015.

Cheng, Y., Zheng, G., Wei, C., Mu, Q., Zheng, B., Wang, Z., Gao, M., Zhang, Q., He, K., Carmichael, G., Poschl, U., and Su, H.: Reactive nitrogen chemistry in aerosol water as a source of sulfate during haze events in China, Science Advances, 2, 10.1126/sciadv.1601530, 2016.

China Electricity Council: Compilation of power industry statistics 2014, China Electricity Council, Beijing, 2015.

Chu, B., Zhang, X., Liu, Y., He, H., Sun, Y., Jiang, J., Li, J., and Hao, J.: Synergetic formation of secondary inorganic and organic aerosol: effect of SO₂ and NH₃ on particle formation and growth, Atmos Chem Phys, 16, 14219-14230, 10.5194/acp-16-14219-2016, 2016.

Cui, J., Qu, J., Bo, X., Chang, X., Feng, X., Mo, H., Li, S., Zhao, Y., Zhu, F., and Ren, Z.: High resolution power emission inventory for China based on CEMS in 2015, China Environmental Science, 38, 2062~2074, 2018.

CMAQv5.0.2 (Version 5.0.2), 2014.

Fu, X., Wang, S. X., Zhao, B., Xing, J., Cheng, Z., Liu, H., and Hao, J. M.: Emission inventory of primary pollutants and chemical speciation in 2010 for the Yangtze River Delta region, China, Atmos Environ, 70, 39-50, 10.1016/j.atmosenv.2012.12.034, 2013.

Fu, X., Wang, S., Chang, X., Cai, S., Xing, J., and Hao, J.: Modeling analysis of secondary inorganic aerosols over China: pollution characteristics, and meteorological and dust impacts, Sci Rep, 6, 35992, 10.1038/srep35992, 2016.

Janssens-Maenhout, G., Crippa, M., Guizzardi, D., Dentener, F., Muntean, M., Pouliot, G., Keating, T., Zhang, Q., Kurokawa, J., Wankmüller, R., Denier van der Gon, H., Kuenen, J. J. P., Klimont, Z., Frost, G., Darras, S., Koffi, B., and Li, M.: HTAP_v2.2: a mosaic of regional and global emission grid maps for 2008 and 2010 to study hemispheric transport of air pollution, Atmos Chem Phys, 15, 11411-11432, 10.5194/acp-15-11411-2015, 2015.

Kain, J. S.: The Kain-Fritsch convective parameterization: An update, Journal of Applied Meteorology, 43, 170-181, 10.1175/1520-0450(2004)043<0170:tkcpau>2.0.co;2, 2004.

Karplus, V. J., Zhang, S., and Almond, D.: Quantifying coal power plant responses to tighter SO2 emissions standards in China, Proc Natl Acad Sci U S A, 115, 7004-7009, 10.1073/pnas.1800605115, 2018.

Lei, Y., Zhang, Q., Nielsen, C., and He, K.: An inventory of primary air pollutants and CO2 emissions from cement production in China, 1990-2020, Atmos Environ, 45, 147-154, 10.1016/j.atmosenv.2010.09.034, 2011.

Li, M., Zhang, Q., Kurokawa, J., Woo, J. H., He, K. B., Lu, Z. F., Ohara, T., Song, Y., Streets, D. G., Carmichael, G. R., Cheng, Y. F., Hong, C. P., Huo, H., Jiang, X. J., Kang, S. C., Liu, F., Su, H., and Zheng, B.: MIX: a mosaic Asian anthropogenic emission inventory under the international collaboration framework of the MICS-Asia and HTAP, Atmos Chem Phys, 17, 935-963, 10.5194/acp-17-935-2017, 2017.

Liu, F., Zhang, Q., Tong, D., Zheng, B., Li, M., Huo, H., and He, K. B.: High-resolution inventory of technologies, activities, and emissions of coal-fired power plants in China from 1990 to 2010, Atmos Chem Phys, 15, 13299-13317, 2015.

Ministry of Environmental Protection of China: Emission standard of air pollutants for industrial kiln and furnace, Ministry of Environmental Protection of China (MEP), Beijing, 1997.

Ministry of Environmental Protection of China: Emission standard of air pollutants for cement industry, Ministry of Environmental Protection of China (MEP), Beijing, 2013.

Mlawer, E. J., Taubman, S. J., Brown, P. D., Iacono, M. J., and Clough, S. A.: Radiative transfer for inhomogeneous atmospheres: RRTM, a validated correlated-k model for the longwave, J Geophys Res-Atmos, 102, 16663-16682, 10.1029/97jd00237, 1997.

Morrison, H., Curry, J. A., and Khvorostyanov, V. I.: A new double-moment microphysics parameterization for application in cloud and climate models. Part I: Description, Journal of the Atmospheric Sciences, 62, 1665-1677, 10.1175/jas3446.1, 2005.

National Bureau of Statistics (NBS): Report of the first national census of pollution sources, China Statistics Press, Beijing, 2010.

Pleim, J. E.: A simple, efficient solution of flux-profile relationships in the atmospheric surface layer, Journal of Applied Meteorology and Climatology, 45, 341-347, 10.1175/jam2339.1, 2006.

Pleim, J. E.: A Combined Local and Nonlocal Closure Model for the Atmospheric Boundary Layer. Part II: Application and Evaluation in a Mesoscale Meteorological Model, Journal of Applied Meteorology and Climatology, 46, 1396-1409, 10.1175/jam2534.1, 2007.

Qi, J., Zheng, B., Li, M., Yu, F., Chen, C., Liu, F., Zhou, X., Yuan, J., Zhang, Q., and He, K.: A high-resolution air pollutants emission inventory in 2013 for the Beijing-Tianjin-Hebei region, China, Atmos Environ, 170, 156-168, 10.1016/j.atmosenv.2017.09.039, 2017.

Sarwar, G., Appel, K. W., Carlton, A. G., Mathur, R., Schere, K., Zhang, R., and Majeed, M. A.: Impact of a new condensed toluene mechanism on air quality model predictions in the US, Geoscientific Model Development, 4, 183-193, 10.5194/gmd-4-183-2011, 2011.

Skamarock, W. C., Dudhia, J. B. K. J., Gill, D. O., Barker, D., Wang, W., and Powers, J. G.: A Description of the Advanced Research WRF Version 3, NCAR Technical Note NCAR/TN-475+STR, 10.5065/D68S4MVH, 2008.

Streets, D. G., Bond, T. C., Carmichael, G. R., Fernandes, S. D., Fu, Q., He, D., Klimont, Z., Nelson, S. M., Tsai, N. Y., Wang, M. Q., Woo, J. H., and Yarber, K. F.: An inventory of gaseous and primary aerosol emissions in Asia in the year 2000, Journal of Geophysical Research: Atmospheres, 108, 10.1029/2002jd003093, 2003.

Wang, G., Zhang, R., Gomez, M. E., Yang, L., Levy Zamora, M., Hu, M., Lin, Y., Peng, J., Guo, S., Meng, J., Li, J., Cheng, C., Hu, T., Ren, Y., Wang, Y., Gao, J., Cao, J., An, Z., Zhou, W., Li, G., Wang, J., Tian, P., Marrero-Ortiz, W., Secrest, J., Du, Z., Zheng, J., Shang, D., Zeng, L., Shao, M., Wang, W., Huang, Y., Wang, Y., Zhu, Y., Li, Y., Hu, J., Pan, B., Cai, L., Cheng, Y., Ji, Y., Zhang, F., Rosenfeld, D., Liss, P. S., Duce, R. A., Kolb, C. E., and Molina, M. J.: Persistent sulfate formation from London Fog to Chinese haze, Proc Natl Acad Sci U S A, 10.1073/pnas.1616540113, 2016a.

Wang, K., Tian, H., Hua, S., Zhu, C., Gao, J., Xue, Y., Hao, J., Wang, Y., and Zhou, J.: A comprehensive emission inventory of multiple air pollutants from iron and steel industry in China: Temporal trends and spatial variation characteristics, Sci Total Environ, 559, 7-14, 10.1016/j.scitotenv.2016.03.125, 2016b.

Woo, J. H., Baek, J. M., Kim, J. W., Carmichael, G. R., Thongboonchoo, N., Kim, S. T., and An, J. H.: Development of a multi-resolution emission inventory and its impact on sulfur distribution for Northeast Asia, Water Air and Soil Pollution, 148, 259-278, 10.1023/a:1025493321901, 2003.

Wu, W., Zhao, B., Wang, S., and Hao, J.: Ozone and secondary organic aerosol formation potential from anthropogenic volatile organic compounds emissions in China, J Environ Sci (China), 53, 224-237, 10.1016/j.jes.2016.03.025, 2017.

Wu, X., Zhao, L., Zhang, Y., Zheng, C., Gao, X., and Cen, K.: Primary Air Pollutant Emissions and Future Prediction of Iron and Steel Industry in China, Aerosol and Air Quality Research, 15, 1422-1432, 10.4209/aaqr.2015.01.0029, 2015.

Xiu, A. J., and Pleim, J. E.: Development of a land surface model. Part I: Application in a mesoscale meteorological model, Journal of Applied Meteorology, 40, 192-209, 10.1175/1520-0450(2001)040<0192:doalsm>2.0.co;2, 2001.

Zhao, B., Wang, S. X., Wang, J. D., Fu, J. S., Liu, T. H., Xu, J. Y., Fu, X., and Hao, J. M.: Impact of national NOx and SO2 control policies on particulate matter pollution in China, Atmos Environ, 77, 453-463, 10.1016/j.atmosenv.2013.05.012, 2013.

Zhao, Y., Wang, S. X., Duan, L., Lei, Y., Cao, P. F., and Hao, J. M.: Primary air pollutant emissions of coal-fired power plants in China: Current status and future prediction, Atmos Environ, 42, 8442-8452, 10.1016/j.atmosenv.2008.08.021, 2008.