Main revisions and response to reviewers' comments

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Title: Quantification and evaluation of atmospheric pollutant emissions from open biomass burning with multiple methods: A case study for Yangtze River Delta region, China

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We thank very much for the valuable comments and suggestions from the two reviewers, which help us improve our manuscript. The comments were carefully considered and revisions have been made in response to suggestions. Following is our point-by-point responses to the comments and corresponding revisions.

Reviewer #1

1. *General comments: This manuscript presents a very comprehensive study of historical trend of OBB emissions in YRD. I am very impressed by the large amounts of work done in this study. The presentation is also of high quality, and the structure is well organized. The constraining method is a little bit weak, but makes the story complete. I would suggest the authors improve the constraining method in future studies. The authors have acknowledged the weakness, which is great. I only have very minor comment for improvements. For constraining method, the correction is based on the comparisons of PM10, and the correction factor was applied to all other species. The authors should acknowledge this limitation in the method section.*

Response and revisions:

We appreciate the reviewer's positive remarks on our manuscript. We thank the reviewer's suggestion and will improve the constraining method in future studies from following aspects. The method can be improved incorporating the observed ambient concentrations of multiple pollutants (e.g., PM_{10}, $PM_{2.5}$, OC and EC) if those concentrations with sufficient temporal and spatial resolution get available. Improvement on the results of constraining method can be expected if more reliable emission factors of biomass burning and improved and the emissions of other sources are obtained and applied in the study. For constraining method, the correction of activity level was based on the comparison between simulated and observed PM_{10} concentrations, and the emissions of other species were then revised according to the changed activity level. In this method, the emission estimation of other species depends largely on the reliability of emission factors for PM_{10} and those species. Large uncertainty may exist due to lack of sufficient domestic measurements. We take the reviewer's suggestion and acknowledge this limitation in the method section. **Corresponding revision was shown in lines 258-264 of Page 9 in the revised manuscript.**

Reviewer #2

1. *This manuscript estimates the air pollutant emissions from open biomass burning(OBB) in Yangtze River Delta for 2005-2015 using traditional bottom-up, fire radiative power (FRP)-based, and constraining approaches, and analyzed the differences between those methods and their underlying reasons. The manuscript is generally well written. However, there are still some issues in the manuscript which authors shall pay attention to. So the paper cannot be accepted for publication before authors address the following comments.*

Response and revisions:

We appreciate the reviewer's crucial and important comments. In general, the presentation of the work has been improved, based on specific comments/suggestion from the reviewer. Same emission factors as bottom-up method were applied to estimate the OBB emissions for 2010 based on FRP-based method, and the results were compared with those based on bottom-up method. Both $PM_{2.5}$ and PM_{10} concentrations were used to evaluate the model performance and to analyze the contribution of OBB in June 7-13, 2014. The benchmarks of the evaluation for model performance and meteorological parameters were added in Table 2 and Table S6 in the supplement. We also take the reviewer's suggestion and provide the monthly variations of fire occurrence for other years in Figure S3 in the supplement. Details follow.

2. *As shown in Table S1 and Table S4, the authors use different emission factors for OBB in bottom-up method and FRP-based method. I suggest same emission factors shall be used for both methods. This is why that for most air pollutants, emissions estimated by bottom-up method is higher than that by FRP-based but the emissions of NMVOC and NH3 from bottom-up method is much lower than that by FRP-based method.*

Response and revisions:

We thank the reviewer's comment. In the bottom-up method, the masses of crop residues burned in the field (CRBF) for different crop species could be obtained, therefore the emission factors for different crop types were usually used. However, the masses of CRBF for different crop species in FRP-based method could not be obtained, and the emission factors based on burned area or fire radiative power (BA or FRP method) by other researchers (van der Werf et al., 2010, Kaiser et al., 2012; Liu et al., 2015; Randerson et al., 2018) were applied, ignoring the difference between crop types. In order to know the differences between the OBB emissions based on FRP-based and bottom-up methods with same emission factors, we followed the reviewer's comment and made an extra case: the emission factors applied in the bottom-up method were weighted with the masses of various crop types and used to estimate the OBB emissions for 2010 with the FRP-based method. The estimated OBB emissions (FRP-based (WSE)) were compared with the emissions based on bottom-up method as shown in Table 3. The OBB emissions for all species in FRP-based (WSE) were smaller than those derived by bottom-up method. The differences in OBB emissions between bottom-up and FRP-based (WSE) method were larger than 50% of those between the bottom-up and the original FRP-based method with different emission factors for most species. It indicated that the discrepancy in activity level contributed the most to the difference in OBB emissions between bottom-up and FRP-based method. **Corresponding revision was shown in lines 200-205 of Page 7 and lines 553-559 of Page 18 in the revised manuscript.**

3. The spatial resolutions of the two domains were set at 27 and 9 km respectively. 9km is kind of coarse resolution. How does this spatial resolution affect the CMAQ modeling results? Will you get a better model performance if you use a 3km resolution?

Response and revisions:

We thank the reviewer's comment. The model performance largely depends on the reliability of emission inventories. The emissions of other sources in this study were obtained from the downscaled the Multi resolution Emission Inventory for China (MEIC) with an original spatial resolution of 0.25°×0.25°. The model performance with a finer resolution might not necessarily be better since the emissions were probably not distributed in the correct grids in finer resolution with a simple spatial interpolation (Zheng et al., 2017). Improvement on emission inventory with the underlying data carefully compiled and analyzed is important to achieve better model performance with high-resolution chemistry transport modeling. Our previous study by Zhou et al. (2017) evaluated the downscaled MEIC and improved local emission inventory with CMAQ modeling at a 3 km resolution in southern Jiangsu of Yangtze River Delta (YRD), and found the model performance was better for the latter inventory. Once the emission inventory of all the anthropogenic sources get improved for the whole YRD region, therefore, a better model performance with high-resolution modeling (e.g., 3km) can be expected.

4. Considering that the PM emissions from OBB are mainly $PM_{2.5}$, and the ambient PM_{10} is more affected by the local road dust emissions, it is not appropriate to only use PM_{10} concentration to evaluate the model performance and analyze the contribution of OBB. I think authors shall use both PM_{10}, $PM_{2.5}$, CO, NO_2, SO_2, OC, EC to do the model evaluation. At least $PM_{2.5}$ shall be included considering that most Chinese cities release $PM_{2.5}$ hourly concentrations since 2013. Although authors give a couple of figures in SI, this is not enough. Specifically, the correction based on the comparisons of PM10 cannot be used for all other species.

Response and revisions:

We thank the reviewer's comment. We agree with the reviewer that observation of more relevant species should ideally be included in the constraining method and evaluation of OBB emissions. However, the most and the second most fire counts were found for YRD region in 2012 and 2010 from 2005 to 2015, while the concentrations of $PM_{2.5}$, CO, NO_2, and SO_2 were unavailable before 2013. The largest daily mass ratio of $PM_{2.5}$ to PM_{10} could reach 91.3% in Nanjing during the OBB event of 2012 and 77.2% in Lianyungang during the event of 2014. The contribution of OBB to PM_{10} estimated in this study was 37% in YRD and 55% in Anhui province during OBB period in June 2012. The OBB could thus be identified as an important source of PM_{10} during the OBB event periods as well. Therefore, we used PM_{10} concentration to evaluate the model performance and analyze the contribution of OBB in 2010 and 2012. Compared to $PM_{2.5}$ and PM_{10}, OBB was not a major source of NO_2 and SO_2, and the OC and EC concentrations were still unavailable at present as they were not considered as regulated pollutants in China. In this case, we followed the reviewer's suggestion and applied both $PM_{2.5}$ and PM_{10} concentrations to evaluate the model performance and analyze the contribution of OBB in June 7-13, 2014. Similar to 2010 and 2012, the NMBs and NMEs between observed and simulated particle concentrations with constrained OBB emissions were smaller than most of those without OBB emissions or with OBB emissions based on FRP-based. **Corresponding**

revision was shown in lines 459-475 of Page 15 and 490-498 of Page 16 in the revised manuscript. The average contributions of OBB to $PM_{2.5}$ and PM_{10} during June 7-13, 2014 were estimated at 29% and 23% for 22 cities in YRD. It again suggested that the OBB was an important source of both $PM_{2.5}$ and PM_{10} during OBB event. **Corresponding revision was shown in lines 587-593 and lines 605-607 of Page 19 in the revised manuscript.**

We also admitted the limitation of constrained method, as our response to Question 1 of Reviewer 1. We agree with the reviewer that the concentrations of $PM_{2.5}$ or OC were more suitable for constraining OBB emissions. However, the data were unavailable before 2013, particularly for 2010 and 2012 with the most and the second most fire counts detected by satellite. As OBB was an important source of PM_{10} as well, we had to apply PM_{10} concentrations to constrain the OBB emissions. The activity level was constrained based on the comparisons between simulated and observed PM_{10} concentrations, and the OBB emissions of other species were revised according to the changed activity level. The reliability of emissions for other species depended largely on the accuracy of emission factors for PM_{10} and those species. Uncertainties would be introduced to the emission estimation, resulting from lack of sufficient and qualified domestic field tests on OBB emission factors. We admit this limitation in the method section, and improvement can be expected with more measurements on concentrations of multiple pollutants and local emission factors available in the future. **Corresponding revision was shown in lines 258-264 of Page 9 in the revised manuscript.**

5. The model performance statistics for meteorological parameters shown in Table S6 and that for PM10 concentrations as shown in Table 2 shall include the benchmark of the evaluation.

Response and revisions:

We thank the reviewer’s comment. The benchmarks of the evaluation for meteorological parameters from Emery et al. (2001) and Jiménez et al. (2006) were added in Table S6. The meteorological parameters of this study were basically in compliance with benchmarks. **Corresponding revision was shown in lines 312-317 of Page 11 in the revised manuscript.**

As many factors would influence the model performance of chemistry transport model, no uniform benchmark was obtained for different regions. We selected the results in US (Zhang et al., 2006) as the benchmark for $PM_{2.5}$ and PM_{10} concentrations, as added in Table 2. As can be found in the table, the NMBs and NMEs for most case with the constrained OBB emissions were close to those by Zhang et al. (2006). The NMEs for hourly $PM_{2.5}$ and PM_{10} were slightly larger. Given the larger uncertainty in emission inventory of anthropogenic sources for China and the uncertainty in spatial and temporal distribution of OBB emissions due to satellite detection limit, we believe the model performance with the constrained OBB emissions was improved and acceptable. **Corresponding revision was shown in lines 490-498 of Page 16 in the revised manuscript.**

6. For OBB, temporal allocation is very important. It is good to see the monthly variations of fire occurrence in Figure 1. However, the authors only give information for year 2010 and 2012, I wonder if the authors can provide such information for other years.

Response and revisions:

We thank the reviewer's suggestion and provide the information for other years (2005-2015) in Figure S3 in supplement.

7. Figure 2 shall give the name of each city in the YRD. Otherwise it is difficult for readers to understand when author talk about Lianyungang, Fuyang, Shanghai, Suzhou, Wuxi, Changzhou, etc.

Response and revisions:

We thank the reviewer’s suggestion and provide the name of each city in the YRD in Figure 2.

8. The color in Figure 4 is very difficult to read.

Response and revisions:

We thank the reviewer’s reminder. We applied thicker lines and changed the colors to make the figure easier to read.

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Quantification and evaluation of atmospheric pollutant emissions from open biomass burning with multiple methods: A case study for Yangtze River Delta region, China

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Abstract

Air pollutant emissions from open biomass burning (OBB) in Yangtze River Delta (YRD) were estimated for 2005-2015 using three (traditional bottom-up, fire radiative power (FRP)-based, and constraining) approaches, and the differences between those methods and their sources were analyzed. The species included PM_{10}, $PM_{2.5}$, organic carbon (OC), ~~black~~ elemental carbon (~~BC~~EC), CH_4, non-methane volatile organic compounds (NMVOCs), CO, CO_2, NO_X, SO_2 and NH_3. The inter-annual trends in emissions with FRP-based and constraining methods were similar with the fire counts in 2005-2012, while that with traditional method was not. For most years, emissions of all species estimated with constraining method were smaller than those with traditional method except for NMVOCs, while they were larger than those with FRP-based except for EC, CH_4 and NH_3. Such discrepancies result mainly from different masses of crop residues burned in the field (CRBF) estimated in the three methods. Chemistry transport modeling (CTM) was applied using the three OBB inventories. The simulated PM_{10} concentrations with constrained emissions were closest to available observations, implying constraining method provided the best emission estimates. CO emissions in the three methods were compared with other studies. Similar temporal variations were found for the constrained emissions, FRP-based emissions, GFASv1.0 and GFEDv4.1s, with the largest and the lowest emissions estimated for 2012 and 2006, respectively. The temporal variations of the emissions based on traditional method, GFEDv3.0 and Xia et al. (2016) were different with them. The constrained CO emissions in this study were commonly smaller than those based on traditional bottom-up method and larger than those based on burned area or FRP in other studies. In particular, the constrained emissions were close to GFEDv4.1s that contained emissions from small fires. The contributions of OBB to two particulate pollution events in 2010 and 2012 were analyzed with brute-force method. Attributed to varied OBB emissions and meteorology, the average contribution of OBB to PM_{10} concentrations in June 8-14 2012 was estimated at 37.6% (56.7 μg/m^3), larger than that in June 17-24, 2010 at 21.8 % (24.0 μg/m^3). Influences of diurnal curves of OBB emissions and meteorology on air pollution caused by OBB were evaluated by designing simulation scenarios, and the results suggested that air pollution caused by OBB would become heavier if the meteorological conditions were unfavorable, and that more attention should be

paid to the OBB control at night. Quantified with Monte-Carlo simulation, the uncertainty of traditional bottom-up inventory was smaller than that of FRP-based one. The percentages of CRBF and emission factors were the main source of uncertainty for the two approaches, respectively. Further improvement on CTM for OBB events would help better constraining OBB emissions.

1. Introduction

Open biomass burning (OBB) is an important source of atmospheric particulate matter (PM) and trace gases including methane (CH_4), non-methane volatile organic compounds (NMVOCs), carbon monoxide (CO), carbon dioxide (CO_2), oxides of nitrogen (NO_X), sulfur dioxide (SO_2), and ammonia (NH_3) (Andreae and Merlet, 2001; van der Werf et al., 2010; Wiedinmyer et al., 2011; Kaiser et al., 2012; Giglio et al., 2013, Qiu et al., 2016; Zhou et al., 2017a). As it has significant impacts on air quality and climate (Crutzen and Andreae, 1990; Cheng et al., 2014; Hodzic and Duvel, 2018), it is important to understand the amount, temporal variation and spatial pattern of OBB emissions.

Various methods have been used to estimate OBB emissions, including traditional bottom-up method that relied on surveyed amount of biomass burning (traditional bottom-up method), the method based on burned area or fire radiative power (BA or FRP method), and emission constraining with chemistry transport model (CTM) and observation (constraining method). In the traditional bottom-up method that was most frequently used, emissions were calculated as a product of crop production level, the ratio of straw to grain, percentage of dry matter burned in fields, combustion efficiency, and emission factor (Streets et al., 2003; Cao et al., 2007; Wang and Zhang, 2008; Zhao et al., 2012; Xia et al., 2016, Zhou et al., 2017a). The BA or FRP method was developed along with progress of satellite observation technology. BA was detected through remote sensing, and used in OBB emission calculation combined with ground biomass density burned in fields, combustion efficiency and emission factor. As burned area of each agricultural fire was usually small and difficult to be detected, this method could seriously underestimate the emissions (van der Werf et al., 2010; Liu et al., 2015). In FRP-based method, fire radiative energy (FRE) was calculated with FRP at over pass time of satellite and the diurnal cycle of FRP. The mass of crop residues burned in the field (CRBF) were then obtained based on combustion conversion ratio and FRE, and emissions were calculated as a product of the mass of CRBF and emission factor (Kaiser et al., 2012; Liu et al., 2015). In the constraining method, observed concentrations of atmospheric compositions were used to constrain OBB emissions with CTM (Hooghiemstra et al., 2012; Krol et al., 2013; Konovalov et al., 2014). The spatial and temporal distributions of OBB emissions were derived from information of fire points from satellite observation. Although varied methods and data sources might lead to discrepancies in OBB emission estimation, those discrepancies and underlying reasons have seldom been thoroughly analyzed in previous studies. Moreover, few studies applied CTM to evaluate emissions obtained from different methods, thus the uncertainty and reliability in OBB emission estimates remained unclear.

Due to growth of economy and farmers' income, a large number of crop straws were discharged and burned in field, and OBB (which refers to crop straws burned in fields in this paper) became an important source of air pollutants in China (Streets et al., 2003; Shi and Yamaguchi 2014; Qiu et al., 2016; Zhou et al., 2017a). It brings additional pressure to the country, which is suffering poor air quality (Richter et al., 2005; van Donkelaar et al., 2010; Xing et al., 2015; Guo et al., 2017) and making efforts to reduce pollution (Xia et al., 2016; Zheng et al., 2017). Located in the eastern China, Yangtze River Delta (YRD) including the city of Shanghai and the provinces of Anhui, Jiangsu and Zhejiang is one of China’s most developed and heavy-polluted regions (Ran et al., 2009; Xiao et al., 2011; Cheng et al., 2013, Guo et al., 2017). Besides intensive industry and fossil fuel combustion, YRD is also an important area of agriculture production, and frequent OBB events aggravated air pollution in the region (Cheng et al., 2014).

In this study, we chose YRD to develop and evaluate high resolution emission inventories of OBB with different methods. Firstly, we established YRD’s OBB emission inventories for 2005-2012 using the traditional bottom-up method (the percentages of CRBF for 2013-2015 were currently unavailable), and inventories for 2005-2015 using FRP-based and constraining methods. The three inventories were then compared with each other and other available studies, in order to discover the differences and their origins. Meanwhile, the three inventories were evaluated using Models-3 Community Multi-scale Air Quality (CMAQ) system and available ground observations. Contributions of OBB to particulate pollution during ~~two~~ three typical OBB events in 2010~~ and~~, 2012 and 2014 were evaluated through brute-force method.

Influences of meteorology and diurnal curves of OBB emissions on air pollution caused by OBB were also analyzed by designing simulation scenarios. Finally, uncertainties of the three OBB inventories were analyzed and quantified with Monte-Carlo simulation.

2. Data and methods

2.1 Traditional bottom-up method

Annual OBB emissions in YRD were calculated by city from 2005 to 2012 using the traditional bottom-up method with following equations:

$$E_{(i,y),j} = \sum_k \left(M_{(i,y),k} \times EF_{j,k}\right) \quad (1)$$

$$M_{(i,y),k} = P_{(i,y),k} \times R_k \times F_{(i,y)} \times CE_k \quad (2)$$

where *i* and y indicate city and year (2005-2012), respectively; *j* and *k* represent species and crop type, respectively; *E* is the emissions, metric ton (t); *M* is the mass of CRBF, Gg; *EF* is the emission factor, g/kg; *P* is the crop production, Gg; *R* is the ratio of grain to straw (dry matter); *F* is the percentage of CRBF; and *CE* is the combustion efficiency.

As summarized in Table S1 in the supplement, emission factors were obtained based on a comprehensive literature review, and those developed in China were selected preferentially. The mean value was used if various emission factors could be obtained. When the emission factors for one crop straw were not obtained, the mean value of the others was used instead. Annual production of crops at city level was taken from statistical yearbooks (NBS, 2013). The ratios of straw to grain for different crops were obtained from Bi (2010) and Zhang et al. (2008), and the combustion efficiencies for different crop were obtained from Wang et al. (2013), as provided in Table S2 in the supplement. Without officially reported data, the percentages of CRBF were estimated to be half of the percentages of unused crop residues, following Su et al. (2012). In Jiangsu, the percentages of unused crop residues were officially reported for 2008, 2011 and 2012, while data for other years were unavailable. In this work, therefore, the percentages of CRBF were assumed to be constant before 2008 and to decrease by same rate (-15.2%) from 2008 to 2011, since a provincial plan was made in 2009 to increase the utilization of straw (JPDRC and SMAC, 2009). Similarly, the percentages of CRBF for Shanghai were assumed to be constant before 2008 and to decrease by same rate (-16.8%) from 2008 to 2012. Without any official plans released, in contrast, constant percentages of CRBF were assumed for Zhejiang and Anhui before 2011, and that for 2012 was taken from NDRC (2014). We applied uniform percentages of CRBF for cities within a province attributed to lack of detailed information at city level, as summarized in Table S3 in the supplement. OBB emissions after 2012 were not calculated with the traditional bottom-up method, attributed to lack of information on percentages of CRBF and unused crop residues for corresponding years.

2.2 FRP-based method

Similar to traditional bottom-up method, OBB emissions of FRP-based method were calculated by multiplying the mass of CRBF and emission factors of various pollutants, but mass of CRBF were derived from FRP instead of government-reported data. As the burned crop types could not be identified with FRP, uniform emission factors were applied for different crop types (Randerson et al., ~~2015~~2018; Liu et al., 2016; Qiu et al., 2016), as provided in Table S4 in the supplement.

The mass of CRBF was calculated with the following equation:

$$M = FRE \times CR \tag{3}$$

where M represents the mass of CRBF, kg; CR represents the combustion conversion ratio from energy to mass (kg/MJ); and FRE represents the total released radiative energy in an active fire pixel obtained from satellite observation (MJ). We used a combustion ratio (CR) of 0.41 ± 0.04 (kg/MJ) based on the results of Wooster et al. (2005) in the field and Freeborn et al. (2008) in the laboratory. Diurnal cycle of FRP from crop burning was assumed to follow a Gaussian distribution. Following Vermote et al. (2009) and Liu et al. (2015), FRE was calculated using a modified Gaussian function as below:

$$FRE = \int FRP = \int_{0}^{24} FRP_{\mathrm{peak}} \left(\mathrm{b} + \mathrm{e}^{-\frac{(t-h)^2}{2\sigma^2}} \right) dt \tag{4}$$

$$FRE_{\mathrm{peak}} = \frac{FRP_t}{\left[\mathrm{b} + \mathrm{e}^{-\frac{(t-\mathrm{h})^2}{2\sigma^2}} \right]} \tag{5}$$

where FRP_{peak} is the peak fire radiative power in the fire diurnal cycle; t is the overpass time of satellite; and b, σ, and h represent the background level of the diurnal cycle, the width of fire diurnal curve, and the peak hour (local time, LT), respectively.

FRP data were taken from MODIS Global Monthly Fire Location Product (MCD14ML) which provides data from both the Terra and Aqua satellites (Davies et al., 2009). The active fire data in MCD14ML were derived from Terra with overpass times at approximately 10:30 AM and 10:30 PM LT and Aqua satellite with overpass times at 1:30 AM and 1:30 PM LT. The fire products provided the geographic coordinates of fire pixels (also known as fire points), overpass times, satellites and their FRP values. The land cover dataset (GlobCover2009) was used to define croplands (European Space Agency and Université Catholique de Louvain, 2011).

Parameters b, σ, and h from 2005 to 2015 were calculated using the inter-annual Terra to Aqua (T/A) FRP ratios provided in Table S5 in the supplement:

$$b = 0.86r^2 - 0.52r + 0.08 \quad (6)$$

$$\sigma = 3.89r + 1.03 \quad (7)$$

$$h = -1.23r + 14.57 + \varepsilon \quad (8)$$

where r represents the average T/A FRP ratio. Following Liu et al. (2015), we added a parameter ε (4h) to modify FRP_{peak} hour (h) of the diurnal curve, and the modified FRP diurnal curves could better represent observed FRP temporal variability than the original, as shown in Figure S1 in the supplement. As a result, FRE was calculated to range from 1.49×10^6 MJ in 2009 to 1.95×10^6 MJ in 2005, with a mean value of 1.74×10^6 MJ for YRD region (Table S5).

To further understand the sources of discrepancies between bottom-up and FRP-based methods, the emission factors applied in the bottom-up method were weighted with the masses of various crop types and used to estimate the OBB emissions for 2010 with the FRP-based method. The estimated OBB emissions (FRP-based (WSE)) were compared with the emissions based on bottom-up method in section 3.3.

2.3 Constraining method

CTM and observation of ground particle matter (PM) concentrations were applied in constraining OBB emissions given the potentially big contribution of OBB to particle pollution for harvest seasons (Fu et al., 2013; Cheng et al., 2014; Li et al., 2014). To characterize the non-linearity between emissions and concentrations, an initial inventory including OBB and other sources was applied in CTM, and the response of PM concentrations to emissions was calculated by changing OBB emissions by a certain fraction (5% in this study) in the model. We defined a response coefficient as the ratio of relative change in PM concentrations to that in OBB emissions. Simulated PM concentrations were then compared with available observation, and the mass of CRBF and OBB emissions of all species were corrected combining the obtained response coefficient and the discrepancy between observed and simulated PM concentrations. The corrected emissions were further applied in CTM and the process (including recalculation of response coefficient) repeated until the discrepancies between observation and simulation was small enough (the value of I in equation (9) is less than 0.1% in this study). To limit the potential uncertainty in emissions from other sources, the differences between simulated and observed PM concentrations for non-OBB event period were included in the analysis:

$$I = \left| \frac{\sum_{x,i} S_{x,i} - \sum_{x,i} Q_{x,i} \times N_i}{\sum_{x,i} O_{x,i}} - 1 \right| \qquad (9)$$

where x and i stand for the time (time interval of simulation is hour) and city, respectively; O is the observed PM concentration; S and Q are the simulated PM concentration with and without OBB emissions, respectively; and N is the normalized mean bias (NMB) for non-OBB event period.

As primary particles emitted from OBB are almost fine ones, ambient $PM_{2.5}$ concentrations were commonly observed to account for large fractions of PM_{10} during the OBB event. Figure S2 shows the observed concentrations of $PM_{2.5}$ and PM_{10} at Caochangmen station in Nanjing (the capital of Jiangsu) in June 2012, and the average mass ratio of $PM_{2.5}$ to PM_{10} reached 79% during the OBB event in June 8-14, 2012. The ratios might be even higher in northern YRD where most fire points were detected. As ground $PM_{2.5}$ concentrations were unavailable in most cities of northern YRD before 2013, we expected that PM_{10} was an appropriate indicator for OBB pollution, and observed PM_{10} concentrations were used to constrain OBB emissions instead in this study. The daily mean PM_{10} concentrations of all cities were derived from the officially reported Air Pollution Index (API) by China National Environmental Monitoring Center (http://www.cnemc.cn/). The conversion from API scores to PM_{10} concentrations is discussed in the Supplement.

Figure 1 illustrated the ~~monthly variations of fire occurrences in 2010 and 2012 (panels a1 and a2, respectively),~~ spatial patterns of fire points (panels ~~b1~~ a1 and ~~b2~~a2) in June 2010 and 2012, city-level PM_{10} concentrations in YRD region in June 2010 and 2012 (panels ~~c1~~ b1 and ~~c2~~b2), and temporal variations of daily fire occurrences in June 2010 and 2012 (panels ~~d1~~ c1 and ~~d2~~c2). From 2005 to 2012, most OBB activities were found in June 2010 and 2012 and northern YRD was the region with the intensive fire counts. Accordingly PM_{10} concentrations in northern YRD cities were higher than those in more developed and industrialized cities in the eastern YRD (e.g., Shanghai, Suzhou, Wuxi, and Changzhou), because emissions of OBB overwhelmed those from other sources (Li et al., 2014; Huang et al., 2016). Therefore we constrained OBB emissions with observed PM_{10} concentrations in northern YRD cities including Xuzhou, Lianyungang, Fuyang, Bengbu, Huainan, Hefei, Chuzhou and Bozhou. Suggested by the monthly and daily distribution of fire counts (Figures ~~1a~~ S3 and ~~1d~~1c), two strong OBB events were defined for June 17-24, 2010 and June 8-14, 2012, and other days in June of 2010 and 2012 were defined as non-OBB event period. For other years, OBB emissions were first scaled from the constrained emissions in 2010 and 2012 with the ratios of FRE for corresponding year to that for 2010 and 2012 respectively, and then calculated as average of the two. Remarkably, the correction of activity level was based on the comparisons of simulated and observed PM_{10} concentrations~~in constraining method~~, and ~~then~~ the emissions of other species were ~~changed~~ revised ~~based on~~ according to the ~~corrected~~changed activity level. The reliability of emissions estimation for other species ~~based on this method~~ thus depended~~was~~ largely ~~depend~~ on the ~~accuracy~~reliability of emission factors for PM_{10} and those species. Uncertainty ~~It brought~~would be introduced t~~some uncertainties t~~o the ~~emissions of those speci~~e method,s ~~due~~ attributed to ~~the~~lack of sufficient and qualified ~~uncertainties~~ domestic measurements on ~~of~~ emission factors. ~~Moreover, the uncertainty of constrained OBB emission from other reasons was analyzed in section 3.5.~~

Traditional bottom-up method was used to calculate the initial emission input for all species (NMVOCs emission factor was taken from FRP-based method instead as those in bottom-up method (Li et al., 2007) did not contain oxygenated VOCs). In contrast to application of uniform percentage of CRBF within one province, however, percentage of CRBF for each city was calculated based on that in whole YRD and the fraction of FRP in the city to total YRD FRP, to make the spatial distribution of OBB emissions consistent with that of FRP all over YRD region:

$$F_{(i,y)} = \frac{FRP_{(i,y)}}{FRP_{(YRD,y)}} \times \frac{\sum_k P_{(YRD,y),k}}{\sum_k P_{(i,y),k}} \times F_{(YRD,y)} \quad (10)$$

where i and k represent city and crop type, respectively; y indicates the year (2010 and 2012); F, P, and FRP are the percentage of CRBF, crop production, and fire radiative power, respectively. The initial percentage of CRBF for total YRD ($F_{(YRD,y)}$ in eq (10)) was expected to have limited impact on the result and it was set at 10%, smaller than those in previous studies (Streets et al., 2003; Cao et al., 2007; Wang and Zhang, 2008; Zhao et al., 2012; Xia et al., 2016, Zhou et al., 2017a).

2.4 Temporal and spatial distributions

The spatial and temporal patterns of OBB emissions in the three inventories were determined according to the FRP of agricultural fire points. The emissions of m-th grid in region u on n-th day in year y were calculated using equation (11):

$$E_{(m,n),j} = \frac{FRP_{(m,n)}}{FRP_{(u,y)}} \times E_{(u,y),j} \quad (11)$$

where $FRP_{(m,n)}$ is the FRP of m-th grid on n-th day; $FRP_{(u,y)}$ and $E_{(u,y),j}$ are the total FRP and OBB emissions of species j for region u in year y, respectively. The region u indicates city for FRP-based and constraining method, while it indicates province for traditional bottom-up method since uniform percentages of CRBF was applied within the same province in the method.

2.5 Configuration of air quality modeling

The Models-3 Community Multi-scale Air Quality (CMAQ) version 4.7.1 was applied to constrain OBB emissions and to evaluate OBB inventories with different methods. As shown in Figure 2, one-way nested domain modeling was conducted, and the spatial resolutions of the two domains were set at 27 and 9 km respectively in Lambert Conformal Conic projection, centered at (110^{o}E, 34^{o}N) with two true latitudes 25 and 40° N. The mother domain (D1, 180×130 cells) covered most parts of China, Japan, North and South Korea, while the second domain (D2, 118×97 cells) covered the whole YRD region. OBB inventories developed in this work were applied in D2. Emissions from other anthropogenic sources in D1 and D2 were obtained from the downscaled the Multi resolution Emission Inventory for China (MEIC, http://www.meicmodel.org/) with an original spatial resolution of 0.25°×0.25°. Population density was applied to relocate MEIC to each modeling domain. Biogenic emission inventory was from the Model Emissions of Gases and Aerosols from Nature developed under the Monitoring Atmospheric Composition and Climate project (MEGAN MACC, Sindelarova et al., 2014), and the emission inventories of Cl, HCl and lightning NO_X were from the Global Emissions Initiative (GEIA, Price et al., 1997). Meteorological fields were provided by the Weather Research and Forecasting Model (WRF) version 3.4, and the carbon bond gas-phase mechanism (CB05) and AERO5 aerosol module were adopted. Other details on model configuration and parameters were given in Zhou et al. (2017b).

Meteorological parameters of WRF model were compared with the observation dataset of US National Climate Data Center (NCDC), as summarized in Table S6 in the Supplement. For June 2010, the average biases between the two datasets were 0.06 m/s for wind speed, 9.84 degree for wind direction, 0.64 K for temperature and 2.99% for relative humidity.For June 2012, the average biases between the two datasets were 0.01 m/s for wind speed, 7 degree for wind direction, 0.91 K for temperature and 3.1% for relative humidity. The analogue numbers were 0.01 and 0.67 m/s, 7 and 18.22 degree, 0.91 and 0.43 K and 3.1 and 0.07% respectively for June 2012 and 2014, respectively.The analogue numbers were 0.06 m/s, 9.84 degree, 0.64 K and 2.99% respectively for June 2010. The meteorological parameters of this study were basicallyin compliance with –conformity to the benchmarks derived from Emery et al. (2001) and Jiménez et al. (2006). Simulated daily PM_{10} concentrations were compared with observation for non-OBB event period in June 2010 and 2012 in Table S7 in the supplement. The average of normalized mean biases (NMB) and normalized mean errors (NME) were -19.92% and 38.9% for 17 YRD cities in June 2010, and -2220.98% and 33.9% for 22 cities in June 2012, respectively. Simulated daily and hourly PM_{10} and $PM_{2.5}$ concentrations were compared with the observation for non-OBB event period in June 2014 in Table S8 in the supplement. The hourly NMB of $PM_{2.5}$ and PM_{10} were -29.9% and -39.8%, and the hourly NME of $PM_{2.5}$ and PM_{10} were 49.8% and 54.7%. The model performance was similar with that derived by Zhang et al. (2006) in US in general. As shown in Figure S3 S4 in the supplement, moreover, simulated hourly PM_{10} and $PM_{2.5}$ concentrations were in good agreement with observations at four air quality monitoring sites in YRD during non-OBB event period in June 2012. The comparison thus implied the reliability of emission inventory of anthropogenic origin used in this work, while underestimation might occur indicated by the negative NMB.

3. Results and discussions

3.1 OBB emissions estimated with the three methods

OBB emissions estimated with the traditional bottom-up method for 2005-2012 were shown in Table S8 S9 in the supplement. As emission factors were assumed unchanged during the period, similar inter-annual trends were found for all species and CO_2 was selected as a representative species for further discussion. As shown in Figure 3, CO_2 emissions from traditional bottom-up method were estimated to decrease from 23000 in 2005 to 19973 Gg in 2012, with a peak value of 27061 Gg in 2008. In contrast, the number of fire points in YRD farmland increased from 7158 in 2005 to 17074 in 2012. The fire counts detected from satellite thus did not support the effectiveness of OBB restriction by government in YRD before 2013. Table S9 S10 in the supplement presents the annual OBB emissions derived from FRP-based method for 2005-2015 in YRD region. Associated with fire counts, CO_2 emissions were estimated to grow by 119.7% from 2005 to 2012, with the largest and the second largest annual emissions calculated at 19977 and 12718 Gg for 2012 and 2010, respectively (Figure 3). Similar temporal variability was found for fire counts, which increased by 138.5% from 2005 to 2012, with the most and the second most counts found at 17074 and 12322 for 2012 and 2010, respectively.

With the constraining method, as shown in Figure S54 in the supplement, the ratio of constrained mass of CRBF for 2012 to 2010 was 1.51, clearly lower than the ratios of original FRE (1.75) but close to the ratio of modified FRE for 2012 to 2010 (1.57). The comparison suggested that modified FRE better reflect the OBB activity in YRD than original FRE. In order to make the ratio of FRE for the two years be closer to the ratio of constrained mass of CRBF, an improved method was developed for calculating the FRE. Given the possible variation of FRP_{peak} hour between years, we obtained the diurnal cycle of total FRP of YRD for 2005-2015 based on Gaussian fitting as shown in Figure S65 in the supplement. The ratio of FRE for 2012 to 2010 was recalculated at 1.54, further closer to the ratio of constrained mass of CRBF. Therefore the ratios of FRE for another given year to 2012 and 2010 were calculated with this improved method, and were then applied to emission scaling for that year. The constrained OBB emissions from 2005 to 2015 were summarized in Table 1. The inter-annual trend in constrained emissions was similar with those in fire counts and

FRP-based emissions but different from that in emissions with traditional bottom-up method, as shown in Figure 3. It is usually difficult to collect accurate percentages of CRBF from bottom-up method, as it demands intensive investigation in the rural areas. In addition, the percentages of CRBF were not updated for each year, and same percentages were commonly applied for years without sufficient data support from local surveys.

The constrained CO_2 emissions for Jiangsu, Anhui, Zhejiang and Shanghai were calculated at 5790, 4699, 1104 and 419 Gg in 2005, accounting for 48.2%, 39.1%, 9.2% and 3.5% of total OBB emissions in YRD, respectively. The analogue numbers for 2012 were 7345, 16159, 2574 and 394 Gg, and 27.7%, 61.0%, 9.7% and 1.5%, respectively. Jiangsu and Anhui were found to contribute largest to OBB emissions in YRD for 2005 and 2012, respectively. In the traditional bottom-up method, however, Anhui was estimated to contribute largest for both years. City-level OBB emissions estimated with the three methods were summarized in Table S10S11-S12 S13 in the supplement. With the constraining method, in particular, largest CO_2 emissions were found in Suzhou (1708 Gg) of Anhui, Lianyungang (1578 Gg) and Xuzhou (1401 Gg) of Jiangsu in 2005, accounting for 14.2%, 13.1% and 11.7% of the total emissions, respectively. In 2012, Suzhou, Bozhou of Anhui, and Xuzhou of Jiangsu were identified as the cities with the largest emissions, with the values estimated at 5007, 2433, and 2109 Gg, respectively. Depending on distribution of fire points, the shares of OBB emissions by city were close between constraining and FRP-based method, and large emissions concentrated in the north of YRD. Based on surveyed percentages of CRBF and crop production, in contrast, the emission shares by city in traditional bottom-up method were clearly different from the other two, and emissions concentrated in Anhui cities with high crop production level.

The average annual emissions of CO_2 for 2005-2011 with traditional bottom-up method were 87.0% larger than those in constraining method and the emissions for 2012 was 24.6% times smaller than those in constraining method. Given the same sources of emission factors for all species except NMVOCs, the discrepancies of OBB emissions for most species between constraining and traditional bottom-up methods come from the activity levels (i.e., percentages of CRBF and crop production). The average annual constrained emissions from 2005 to 2015 were larger than those derived by FRP-based method for all species except EC, CH_4 and NH_3, since the average annual mass of CRBF from constraining method were 36.9% larger than those from FRP-based method for these years, as shown in Figure S7~~6~~.

The percentage of CRBF is an important parameter to judge OBB activity and to estimate emissions. Besides the investigated values applied in traditional bottom-up approach, the percentages of CRBF were recalculated based on the constrained emissions at provincial level and were shown in Figure ~~S7~~ S8 in the supplement. The largest and smallest percentages of CRBF in the whole YRD region were estimated at 18.3% in 2012 and 8.1% in 2006, respectively. The inter-annual trend in percentages of CRBF for YRD was closest to that for Anhui province, as the province dominated the crop burning in the region. The different inter-annual trends by province were strongly influenced by agricultural practice and government management. Agricultural practice could be associated with income level and mechanization level. Increased income, would lead to more crop residues discarded and burned in the field, while development of mechanization would lead to less. The constrained percentages of CRBF for Shanghai increased from 2005 to 2007 and declined after 2007, while those for Jiangsu decreased from 2005 to 2008 and increased after 2008. Increasing trends were found for the percentages of CRBF for Anhui and Zhejiang from 2005 to 2012, and they might result largely from growth of farmers' income. Note that percentages of CRBF for all provinces except Zhejiang decreased significantly in 2008, attributed largely to the measures of air quality improvement for Beijing Olympic Games. Shanghai was the only one with its percentage of CRBF significantly reduced in 2010, resulting mainly from the air pollution control for Shanghai World Expo in that year. Compare to the percentages of CRBF used in bottom-up method, the constrained ones of Anhui and Jiangsu for all the years except 2012 were smaller, leading to lower constrained OBB emissions than bottom-up ones in those years.

The constrained percentages of CRBF and straw yields for 2012 were shown by city in Figure ~~S8~~ S9 in the supplement, and clear inconsistency in spatial distributions can be found. The percentage of CRBF was not necessarily high for a city with large straw production. For instance, straw production of Yancheng was higher than most other cities, but its percentage of CRBF was 5.7% and lower than most other cities. Through linear regression, correlation coefficient was calculated at only 0.06 between constrained percentage of CRBF and straw yield at city level. The poor correlation between them thus suggested that large uncertainty could be derived if uniform percentage of CRBF was applied to calculate OBB emissions for cities within given province, as what we did in the traditional bottom-up methodology.

3.2 Evaluation of the three OBB inventories with CMAQ

Figures 4 and 5 illustrate the observed 24-hour averaged and simulated hourly PM_{10} concentrations for selected YRD cities in June 17-25, 2010 and June 8-14, 2012, respectively. Four emission cases, i.e., inventory without and with OBB emissions estimated using the three methods, were included. The simulated PM_{10} concentrations without OBB emissions were significantly lower than observation for all cities, implying that OBB was an important source of airborne particulates during the two periods. Simulations with OBB emissions derived from the three methods performed better than those without OBB emissions for most cities during June17-25, 2010 and all cities during June 8-14, 2012. The best performance was found for simulations with constrained OBB emissions in most cities during the two periods, and the high PM_{10} concentrations were generally caught by CTM for the concerned OBB events. In 2010, the observed high concentrations were simulated with constrained emissions in Lianyungang during June 21-23, and Fuyang and Huainan during June 19-21. In 2012, the observed high concentrations were caught with constrained emissions in Xuzhou during June 12-14, Lianyungang during June 13-14, Fuyang during June 11-12, Bozhou during June 10 and Chuzhou during June 11-12. The results thus indicated that fire points could principally capture the temporal and spatial distribution of OBB emissions. Overestimation still existed with constrained OBB emissions for the cities with intensive fire points (e.g., Xuzhou, Bozhou and Fuyang in 2012 and Bengbu in 2010), while underestimation commonly existed for cities with fewer fire points (e.g., Hefei, Chuzhou and Huainan in 2010 and 2012). Due to limitation of MODIS observation, fires at moderate to small scales could not be fully detected (Giglio et al., 2003; Schroeder et al., 2008), thus the spatial allocation of OBB emissions based on FRP could possibly result in more emissions than actual in areas with intensive fire points. ~~In order to further evaluate the OBB emissions, ~~Moreover, we ~~use~~used both $PM_{2.5}$ and PM_{10} concentrations (which were available since 2013) to evaluate the model performances when~~ith~~ the constrained, FRP-based or no OBB emissions ~~based on constraining and FRP-based methods and without OBB emissions~~were applied in CTM for an~~during~~ OBB event ~~of~~ during June 7-13, 2014. Figures S10 and S11 in the supplement illustrate the observed and simulated hourly concentrations for $PM_{2.5}$ and

PM_{10} ~~concentrations for~~in selected YRD cities~~ ion June 7-13, 2014~~, respectively. The best performance was found for simulations with the constrained OBB emissions in most cities during the periods, and the~~ high $PM_{2.5}$ and PM_1~~ peak particle~~0~~ concentrations were generally caught by CTM~~ for the concerned OBB events~~. The observed high concentrations were simulated with the constrained emissions in Lianyungang and Suqian~~during~~on June 12 and Huaian and Yancheng ~~during ~~on June 13.

The NMB and NME between observed and simulated $PM_{2.5}$ and PM_{10} concentrations are shown in Table 2. ~~Among all~~In most~~ the~~ cases, the NMB and NME with constrained OBB emissions were smaller than ~~most of ~~those with other OBB emissions, implying the best guess of OBB emissions obtained through the constraining method combining CTM and ground observation. The simulated $PM_{2.5}$ and PM_{10} concentrations using FRP-based OBB emissions were smaller than observation for the three~~two~~ periods, due mainly to the mass of CRBF were underestimated. The results thus indicated that OBB emissions might be underestimated in FRP-based method in 2010, 2012 and 201~~4~~2, since many small fires in YRD were undetected in MODIS active fire detection products. The probability of MODIS detection was strongly dependent upon the temperature and area of the fire being observed. The average probability of detection for tropical savanna was 33.6% when the temperature of fire was between 600 and 800 ℃ and the area of fire was between 100 and 1000 m^2 (Giglio et al., 2003). In YRD region, on one hand, the fire temperature of crop residue burned in fields was relatively low. On the other hand, nearly 100 farmers were possibly located in a single 1×1 km MODIS pixel (Liu et al., 2015), and a famer commonly owned croplands of several hundred square meters. Therefore many fire pixels in YRD might not be detected, leading to underestimation in the total FRE. The simulated PM_{10} concentrations using traditional bottom-up OBB emissions were higher than observation in 2010 but lower in 2012. The results thus implied the growth in OBB emissions from 2010 to 2012 could not be captured by traditional bottom-up method, attributed partly to application of unreliable percentage of CRBF. We further selected the performance of CMAQ modeling in US (Zhang et al., 2006) as the benchmark for $PM_{2.5}$ and PM_{10} simulation. As can be seen in Table 2, the NMBs and NMEs for most case with the constrained OBB emissions were close to those by Zhang et al. (2006). The NMEs for hourly

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$PM_{2.5}$ and PM_{10} were slightly larger. Given the larger uncertainty in emission inventory of anthropogenic sources for China and the uncertainty in spatial and temporal distribution of OBB emissions due to satellite detection limit, we believe the model performance with the constrained OBB emissions was improved and acceptable.

~~The NMB of hourly $PM_{2.5}$ and PM_{10} during OBB event of June 2014 with constrained OBB emissions were smaller than those of Zhang (2006), and the NME of hourly $PM_{2.5}$ and PM_{10} with constrained OBB emissions were larger than those of Zhang (2006). Considering that the accuracy of emissions of other sources for US might be higher than that used in this study and there were uncertainties in the temporal and spatial distribution of OBB emissions derived from satellite due to insufficient satellite detection capability, the model performance with constrained OBB emissions was acceptable. However, the NMB and NME of hourly $PM_{2.5}$ and PM_{10} without OBB emissions and with FRP-based emissions were higher than those of Zhang (2006).~~

3.3 Comparisons of different methods and studies

We selected CO to compare emissions in this work and other inventories for YRD, given the similar emission factors of CO applied in different studies. CO emissions from the three methods in this study were compared with GFASv1.0 (Kaiser et al., 2012), GFEDv3.0 (van der Werf et al., 2010), GFEDv4.1 (Randerson et al. ~~2015~~2018), Wang and Zhang (2008), Huang et al. (2012), Xia et al. (2016) and Zhou et al. (2017a), as shown in Figure 6. The emissions from Wang and Zhang (2008), Huang et al. (2012), Xia et al. (2016) and Zhou et al. (2017a) were derived by traditional bottom-up method, while GFASv1.0, GFEDv3.0 and GFEDv4.1 were based on FRP and BA methods. In particular, emissions from small fires were included in GFEDv4.1. Similar inter-annual variations were found for emissions derived from FRP measurement including the constrained and FRP-based emissions in this work, GFAS v1.0, and GFED v4.1, while those of GFEDv3.0 and Xia et al. (2016) were different. The percentages of CRBF were assumed unchanged during the studying period in Xia et al. (2016), thus the temporal variation of OBB emissions were associated with the change in annual straw production.

The constrained CO emissions in this work were lower than other studies using traditional bottom-up method (Wang and Zhang, 2008; Huang et al., 2012; Xia et al., 2016) and higher than those based on burned area and FRP derived from satellite (GFEDv3.0; GFASv1.0; GFEDv4.1). In particular, the average annual constrained emissions from 2005 to 2012 were 3.9, 0.5 and 15.0 times larger than those in GFASv1.0, GFEDv4.1s and GFEDv3.0, respectively. The constrained emissions were closest to GFED v4.1s that included small fires. Since the area of farmland belonging to individual farmers was usually small, small fires were expected to be important sources of OBB emissions in YRD. GFEDv4.1s might still underestimate OBB emissions due to the omission errors for the small fires in MODIS active fire detection products (Schroeder et al., 2008). In addition, the constrained CO emission for 2013 was 31.5% larger than those by Qiu et al. (2016) calculated based on burned area from satellite observations. The average annual CO emissions from 2005 to 2012 by the constraining method were 57.2% smaller than Xia et al. (2016), and the constrained emissions for 2006 were respectively 27.6% and 56.9% lower than those by Huang et al. (2012) and Wang and Zhang (2008). It implied again that the emissions derived from traditional bottom-up method might be overestimated. Moreover, discrepancy in estimations for the same year between Huang et al. (2012) and Wang and Zhang (2008) with traditional bottom-up resulted mainly from application of different percentages of CRBF, implying that calculation of OBB emissions was sensitive to the parameter with the bottom-up approach.

The spatial distribution of constrained emissions in this work and those in GFASv1.0, GFEDv3.0 and GFEDv4.1s were illustrated in Figure 7. Intensive OBB emissions in GFEDv3.0 were mainly found in parts of Anhui, Jiangsu and Shanghai, while the constrained emissions, GFEDv4.1s and GFASv1.0 emissions occurred in most YRD regions in accordance with the distribution of fire points. Therefore, GFEDv3.0 might miss a large number of burned areas, leading to underestimation in emissions and bias in spatial distribution.

In order to understand the discrepancies of emissions for different species in this work and other inventories, the emissions of 2010 derived from the three methods in this study, GFASv1.0, GFEDv3.0, GFEDv4.1s and Xia et al. (2016) were summarized in Table 3. Similar to CO, the constrained emissions for all species in this work were lower than Xia et al. (2016) and OBB emissions of this study based on traditional bottom-up method. The constrained emissions for all species in this work were larger than GFASv1.0 and those for all species except NH_3 were larger than GFEDv3.0 and GFEDv4.1s. In addition, the constrained emissions for most species were lower than the emissions from Huang et al. (2012), Wang and Zhang (2008) and Xia et al. (2016) using traditional bottom-up method in 2006. In most cases, the discrepancy in activity levels between studies was larger than that in emission factors. Specifically, the OBB emissions for all species– in FRP-based (WSE)with same emission factors as bottom-up method based on FRP-based method were lower smaller than those derived by bottom-up method. The differences in OBB emissions between bottom-up and FRP-based (WSE) method were larger than 50% of those between the bottom-up and the original FRP-based method with different emission factors for most species.The differences ofin OBB emissions for most species between bottom-up and FRP-based method with same emission factors (WSE) were larger than or half more than those between bottom-up and FRP-based method with different emission factors. It indicated that the discrepancy in activity levels contributed the most of to the difference ofin OBB emissions between bottom-up and FRP-basedthe two methods.

Resulting from the different sources of emission factors, the discrepancies between studies or methods varied greatly by species. For PM_{10} and $PM_{2.5,}$ as an example, the emissions by Xia et al. (2016) were respectively 35.8% and 50.3% higher than constrained emissions in 2010. The discrepancies for SO_2 and NO_X were larger: the emissions by Xia et al. (2016) were 4.7 and 3.1 times larger than our constrained emissions, respectively. Moreover, the constrained NMVOCs emission was 152.5 and 10.7 times larger than that of GFEDv3.0 and GFEDv4.1s in 2010, as the emission factors of GFEDv3.0 and GFEDv4.1s did not contain oxygenated VOCs. In contrast, the constrained NH_3 emission was 4.7% and 47.9% smaller than that of GFEDv3.0 and GFEDv4.1s. The comparisons indicated that emission factors were important sources of uncertainties in estimation of OBB emissions with different methods.

3.4 Contribution of OBB to particulate pollution and its influencing factors

The brute-force method (BFM, Dunker et al., 1996) was used to analyze the contributions of OBB to particulate PM_{10} pollution for the two OBB events, June 17-24, 2010 and June 8-14, 2012. Simulated PM_{10} concentrations with and without constrained OBB emissions were compared, and the difference indicated the contribution from OBB as shown by city in Figure 8. The average contribution in June

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8-14, 2012 was estimated at 37.6% (56.7 μg/m^3) for 22 cities in YRD, and the contribution for June 17-24, 2010 was smaller at 21.8 % (24.0 μg/m^3) for 17 cities. Our result for 2012 was nearly the same as that for 5 YRD cities in 2011 (37.0%) by Cheng et al. (2014). Using the BFM method, the contribution of OBB emissions to PM_{10} concentrations were estimated to increase by 136.3% from 2010 to 2012 in this work, and the growth rate was larger than that of OBB emissions (50.8%). Therefore, factors other than emissions (e.g., meteorology) could also play an important role in elevating the contribution of OBB to ambient particle pollution. For example, the average precipitation in June 8-14, 2012 was 36% lower than that in June 17-24, 2010, exaggerating the particle pollution during OBB event. For the OBB event during June 7-13, 2014, In order to know the contributions of OBB to both $PM_{2.5}$ and PM_{10} pollution during OBB event, the brute-force method was used to analyze the contributions of OBB to $PM_{2.5}$ and PM_{10} in June 7-13, 2014, and the contributions fromof OBB to both $PM_{2.5}$ and PM_{10} concentrations were shown by city in Figure 9. The average contributions of $PM_{2.5}$ and PM_{10} in June 7-13, 2014 were estimated at 29.3% (20.0 μg/m^3) and 23.1% (17.3 μg/m^3) for 22 cities in YRD, indicating again. Therefore, the contribution of OBB to PM_{10} was up to 78.8% of the contribution of OBB to $PM_{2.5}$. It suggested that the OBB was an important source of $PM_{2.5}$ and PM_{10}ambient particles simultaneously. TheO-BB contributions of to PM_{10} for 2014 was obviously smaller than that offor 2012 and close to that of 2010, attributed mainly due mainly to the reduced straw burning in crop land. difference of OBB emissions.

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The average contributions of OBB for 2012 were estimated at 55.0% (98.4 μg/m^3), 36.4% (58.0 μg/m^3), 23.6% (12.9 μg/m^3), and 14.4% (11.2 μg/m^3) for 6 cities of Anhui, 10 cities of Jiangsu, 5 cities of Zhejiang and Shanghai, respectively. For individual cities, large contributions of OBB for 2012 were found in Xuzhou, Bozhou, Fuyang, and Lianyungang located in the north YRD, reaching 82.3% (284.3 μg/m^3), 75.2% (207.5 μg/m^3), 71.9% (134.7 μg/m^3) and 63.5% (96.2 μg/m^3), respectively. Similarly, large contributions for 2010 were found in Lianyungang, Fuyang and Bozhou reaching 63.3% (69.8 μg/m^3), 58.2% (71.9 μg/m^3) and 78.8% (53.6 μg/m^3), respectively. In general the spatial distribution of contributions to PM_{10} mass concentrations was similar with that of fire points, confirming the rationality of constraining OBB emissions with observed PM_{10} concentration in cities in north Anhui and Jiangsu. The similar result was found in For contributions of OBB to $PM_{2.5}$ during the OBB event in 2014, the large contributions of OBB were found in

Xuzhou, Huaian and Suqian ~~located in the in the north YRD~~during the event in 2014, reaching 67.5% (111.7 μg/m^3), 60.7% (50.6 μg/m^3) and 53.2% (49.6 μg/m^3), respectively.

To explore the influence of meteorology on air pollution caused by OBB, we simulated PM_{10} concentrations for June 8-14 (PE1) and June 22-28 2012 (PE2) with varied meteorology conditions but fixed OBB emissions (i.e., constrained emissions for June 8-14, 2012). Poorer meteorology conditions during PE1 were found than PE2. The average wind speed in PE1 was 2.4 m/s, 17% lower than that in PE2. The average wind direction in PE1 was 168.3°, close to south with polluted air in land. In contrast, the average wind direction in PE2 was 118.3°, close to east with clean air from the ocean. The average precipitation in PE2 was 6.8mm, 28% higher than that in PE1. As shown in Figure ~~9~~10, the average contribution of OBB to PM_{10} concentrations for 22 cities in YRD region was estimated at 56.7 μg/m^3 for PE1, 23% larger than that for PE2, and the contributions in most cities were much larger for PE1 than those for PE2, except for Bozhou and Fuyang. The comparisons thus suggest that air pollution caused by OBB would exaggerate under poorer meteorology conditions. To reduce air pollution caused by OBB in harvest season in YRD, therefore, more attention should be paid to the OBB restriction on those days with unfavorable meteorology conditions such as calm wind and rainless period.

To further analyze the influence of diurnal variation of emissions on air pollution caused by OBB, we simulated PM_{10} concentrations of June 17-24 2010 with various diurnal curves of OBB emissions (i.e., those for 2010 and 2012). Constrained emissions were applied in the simulation. As shown in Figure ~~10~~11, the contributions of OBB to PM_{10} concentrations based on diurnal curve of 2012 were larger than those based on 2010 for almost all YRD cities, and the average contribution for the 17 cities was calculated at 28.6 μg/m^3 based on diurnal curve of 2012, 10% larger than that based on 2010. The contribution in Bozhou changed most (1.37 times larger with 2012 curve), while those in Shanghai, Huzhou and Shaoxing changed least. The time of peak value for OBB emissions in 2012 was 2.5 hours later than 2010, indicating that the fraction of OBB emissions at night for 2012 would be larger than that for 2010. As the diffusion condition for air pollutants at night was usually worse than that during daytime, more OBB emissions at night would elevate its contribution to particle pollution. In the actual fact, the supervision of OBB prohibition was usually conducted by government during daytime, thus some farmers burned more crop residues at night to avoid the punishment. To improve the air quality in harvest season in YRD, more attention should be paid to the OBB restriction at night.

3.5 Uncertainty analysis

The uncertainties of OBB emissions estimated with bottom-up and FRP-based methods were quantified by species using a Monte-Carlo simulation for 2012. A total of 20,000 simulations were performed and the uncertainties were expressed as 95% confidence intervals (CIs) around the central estimates. The parameters contributing most to OBB emission uncertainty were also identified according to their contribution to the variance in Monte-Carlo simulation.

For traditional bottom-up method, parameters included crop productions, percentages of CRBF, straw to grain ratios, combustion efficiencies, and emission factors. Crop production was directly taken from official statistical yearbooks (NBS, 2013) and its uncertainty was expected to be limited and not included in the analysis. As the percentage of CRBF was determined at half of the percentage of unused crop residues, its uncertainty was set at -100% to +100%. The combustion efficiencies were assumed within an uncertainty range of 10% around the mean value according to de Zarate (2005) and Zhang et al. (2008). Uncertainties of emission factors were obtained from original literatures where they were derived. If emission factor was derived from a single measurement, normal distribution was applied with standard deviation directly taken from that work. If emission factor was derived from multiple measurements and the samples were insufficient for data fitting, uniform distribution was tentatively applied with a conservative strategy to avoid possible underestimation of uncertainty: The uncertain range of given emission factor would be expanded according to Li et al. (2007) if the range originally from multiple studies was smaller than that in Li et al. (2007). Summarized in Table ~~S13~~ S14 in the supplement was a database for emission factors and percentages of CRBF, with their uncertainties indicated by probability distribution function (PDF). As shown in Table 4, the uncertainties of OBB emissions with traditional bottom-up method for PM_{10}, $PM_{2.5}$, EC, OC, CH_4, NMVOCs, CO, CO_2, NO_X, SO_2 and NH_3 in 2012 were estimated at -56% to +70%, -56% to +70%, -50% to +54%, -54% to +73%, -49% to +58%, -48% to +59%, -46% to +73%, -48% to +60%, -47% to +87%, -59% to +138% and -51% to +67%, respectively. For most species, the percentages of CRBF contributed largest to the uncertainties of OBB emissions, while emission factors were more significant to SO_2 uncertainty.

For FRP-based method, parameters included total FRE, combustion conversion ratio and emission factors. Uncertainty of total FRE was associated with FRP value, MODIS detection resolution, and the methodology used to calculate FRE per fire pixel. Indicated by Freeborn et al. (2014), the coefficient of variation of MODIS FRP for a fire pixel was 50%, but it declined to smaller than 5% for the aggregation of over 50 MODIS active fire pixels. Give the large number of fire pixels for in YRD (more than 17000 in 2012), FRP was expected to contribute little to uncertainty of total FRE and could thus be ignored. Due to limitation of MODIS resolution, small fires could not be fully detected and the number of fire pixel could be underestimated by 300% on crop-dominant areas (Schroeder et al., 2008), therefore the uncertainty of number of fire pixel was assumed to be 0 to +300%. The method used to calculate FRE based on single fire pixel assumed that fire lasted one day. Given the small cropland owned by one farmer in YRD, individual fire normally lasted several hours, and FRE could be overestimated. As the total FRE in FRP-based method was estimated 2.6 times larger than that from constraining method based on the same number of the fire pixel, we tentatively assumed the uncertainty range of FRE for one fire pixel at 0% to -72%. The uncertainty of total FRE was then estimated at -17% to +154% (95% CIs) based on the principle that total FRE was calculated as the number of fire pixel multiplied by average FRE. The uncertainty of combustion conversion ratio was derived from Wooster et al. (2005) and Freeborn et al. (2008), while those of emission factors taken from Akagi et al. (2011). As a result, uncertainties of FRP-based inventory were estimated at -77% to +274%, -63% to +244%, -78% to +281%, -78% to 276%, -83% to +315%, -63% to +243%, -52% to +223%, -21% to +164%, -82% to +303%, -78% to +279%, and -82% to +302% for PM_{10}, $PM_{2.5}$, EC, OC, CH_4, NMVOCs, CO, CO_2, NO_X, SO_2 and NH_3 in 2012, respectively. Emission factors contributed most to the uncertainties of emissions for all species except CO_2.

The uncertainty of constrained emissions could hardly be provided by Monte-Carlo simulation, as the results were associated with CTM performance. In general, CTM performance could be influenced by emission estimates for sources other than OBB, chemistry mechanism of CTM and temporal and spatial distribution of OBB emissions. Emission inventory of anthropogenic sources that incorporates the best available information of individual plants was expected to improve the CTM performance at the regional or local scale (Zhou et al., 2017b). The influence of chemistry mechanism came mainly from secondary organic carbon (SOC) modeling. According to the Cheng et al. (2014) and Chen et al. (2017), the mass fraction of SOC to PM_{10} could reach 10% during the OBB event in YRD, and that part might not be well constrained with the approach we applied in this work. Similar to FRP-based method, moreover, temporal and spatial distribution of OBB emissions based on FRP might not be entirely consistent with the reality, due to omission errors in the MODIS active fire detection products and limited times of satellite overpass as discussed earlier. Due to data limitation, finally, we relied on available PM_{10} concentrations in current method. More data of multi pollutant concentrations (e.g., $PM_{2.5}$, OC and EC) with sufficient temporal and spatial resolution are in great need to better constrain the OBB emissions.

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In general, uncertainties of OBB emissions with traditional bottom-up method were estimated smaller than those with FRP-based method, and uncertainties for CO_2 and CO were usually smaller than other species in both methods attributed mainly to fewer variations in their emission factors. OBB emission estimation with traditional bottom-up method could be improved if more accurate percentages of CRBF are obtained, and that with FRP-based method could be improved when the omission error of satellite and the uncertainties of emission factors are reduced. Efforts should also be recommended on improvement of CTM for better constraining the OBB emissions.

4. Conclusions

Taking YRD in China as an example, we have thoroughly analyzed the discrepancies and their sources of OBB emissions estimated with traditional bottom-up, FRP-based and constraining methods. The simulated PM_{10} concentrations through CMAQ with constrained emissions were closest to available observation, implying the improvement of emission estimation with this method. The inter-annual variations in emissions with FRP-based and constraining methods were similar with the fire counts, while that with traditional bottom-up method was not. It indicated that emissions with traditional bottom-up method could not capture the real inter-annual trend of OBB emissions. The emissions of all species except NMVOCs based on traditional bottom-up method might be overestimated in most years, attributed mainly to the elevated percentages of CRBF used in the method. The emissions with FRP-based method might be underestimated in 2005-2015, attributed to the omission errors in the MODIS active fire detection products and thereby to the underestimation in mass of CRBF. The CO emissions with traditional bottom-up, FRP-based and constraining methods were compared with other studies. Similar temporal variations were found for the constrained emissions, emissions based on FRP-based, and emissions in GFASv1.0 and GFEDv4.1s. CO emissions based on traditional bottom-up method both in this work and other studies were usually higher than those derived by constraining method, and the CO emissions based on FRP-based method both in this work and other studies usually were lower than those derived by constraining method. It again demonstrated that traditional bottom-up method might overestimate OBB emissions in YRD and FRP-based method might underestimate them. The contributions of OBB to particulate pollution in typical episodes were analyzed using the Brute-force method in CMAQ modeling. The OBB emissions in 2012 were 51% larger than those in 2010, while its contribution to average PM_{10} mass concentrations was estimated to increase by 136% from 2010 to 2012. It indicated that the elevated contribution of OBB was not attributed only to growth in OBB emissions but was also influenced by the meteorology. Quantified with a Monte-Carlo framework, the uncertainties of OBB emissions with traditional bottom-up method were smaller than those with FRP-based method. The uncertainties of emissions based on traditional bottom-up and FRP-based were mainly from the percentages of CRBF and emission factors, respectively. Further improvement on CTM for OBB events would help better constraining OBB emissions.

Limitations remained in this study. Given the difficulty in field investigation, annual CRBF used in traditional bottom-up method was obtained from limited studies and it could not correctly reflect the real OBB activity. The reliability of OBB emissions with FRP-based method depended largely on the detection resolution of the satellite. In YRD where the burned areas of individual fires were small, many fires could not be detected by MODIS. The accuracy of constrained emissions depended largely on model performance and spatial and temporal distributions of OBB emissions derived from satellite-observed FRP. Therefore FRP-based and constraining method may be improved if more reliable fire information is obtained. In addition, more measurements on local emission factors for OBB are suggested in the future to reduce the uncertainty of emissions.

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FIGURE CAPTIONS

Figure 1. ~~(a) Monthly variations of fire occurrences in 2010 and 2012,~~ (~~b~~a) spatial patterns of fire points in June 2010 and June 2012, (~~c~~b) PM_{10} concentrations for city-level in YRD in June 2010 and June 2012, and (~~d~~c) temporal variations of daily fire occurrences in June 2010 and 2012. City abbreviations FY, BZ, BB, HN, HF, CZ(a), XZ, LYG, NJ, YZ, ZJ, TZ, NT, CZ, WX, SZ, HZ(a), JX, HZ, SX, NB, SH indicate is Fuyang, Bozhou, Bengbu, Huainan, Hefei, Chuzhou, Xuzhou, Lianyungang, Nanjing, Yangzhou, Zhenjiang, Taizhou, Nantong, Changzhou, Wuxi, Suzhou, Huzhou, Jiaxing, Hangzhou, Shaoxing, Ningbo, and Shanghai~~)~~.

Figure 2. Model domain and locations of 43 meteorological monitoring sites. The numbers of 1-41 represent the cities of Fuyang, Bozhou, Huaibei, Suzhou, Huainan, Bengbu, Luan, Hefei, Chuzhou, Anqing, Chaohu, Maanshan, Chizhou, Tongling, Wuhu, Huangshan, Xuancheng, Xuzhou, Lianyungang, Suqian, Huaian, Yancheng, Yangzhou, Taizhou, Nanjing, Zhenjiang, Nantong, Changzhou, Wuxi, Suzhou; Huzhou, Jiaxing, Hangzhou, Shaoxing, Ningbo, Zhoushan, Quzhou, Jinhua, Taizhou, Lishui and Wenzhou, respectively.~~Model domain and locations of 43 meteorological monitoring sites.~~

Figure 3. Fire counts and CO_2 emissions estimated with traditional bottom-up, FRP-based and constraining methods for YRD 2005-2012.

Figure 4. Observed 24-hour averaged PM_{10} concentrations and simulated hourly PM_{10} concentrations without OBB emissions (No_OBB) and with OBB emissions based on traditional bottom-up (Traditional_OBB), FRP-based (FRP_OBB) and constraining (Constrained_OBB) methods in Lianyungang, Fuyang, Bozhou, Bengbu, Huainan, Hefei, and Chuzhou during June 17-25, 2010.

Figure 5. Observed 24-hour averaged PM_{10} concentrations and simulated hourly PM_{10} concentrations without OBB emissions (No_OBB) and with OBB emissions based on traditional bottom-up (Traditional_OBB), FRP-based (FRP_OBB) and constraining (Constrained_OBB) methods in Xuzhou, Lianyungang, Fuyang, Bozhou, Bengbu, Huainan, Hefei, and Chuzhou during June 8-14, 2012.

Figure 6. Annual CO emissions from OBB in YRD obtained in this work and other studies from 2005 to 2012.

Figure 7. Spatial distributions of CO emissions from OBB obtained in this work (constraining method), GFAS v1.0, GFED v3.0 and GFED v4.1s in 2010.

Figure 8. The contribution of OBB to PM_{10} concentrations for different YRD cities during OBB events in June 2010 and 2012.

Figure 9. The contribution of OBB to $PM_{2.5}$ and PM_{10} concentrations for different YRD cities during OBB event in June 2014.

Figure ~~9~~10. PM_{10} concentrations contributed by OBB for different YRD cities in Jun 8-14 (PE1) and June 22-28 (PE2), 2012.

Figure ~~10~~11. PM_{10} concentrations contributed by OBB for different YRD cities based on the diurnal variations of 2010 and 2012 in Jun 8-14, 2010.

TABLES

Table 1. Constrained OBB emissions from 2005 to 2015 in YRD (Unit: Gg).

	PM_{10}	$PM_{2.5}$	EC	OC	CH_4	NMVOCs	CO	CO_2	NOx	SO_2	NH_3
2005	175.7	153.7	4.4	38.7	32.1	420.3	670.2	12011.2	22.2	2.7	4.1
2006	171.3	149.9	4.3	37.8	31.3	409.9	653.7	11716.7	21.7	2.6	4.0
2007	219.1	191.7	5.5	48.3	40.0	524.2	835.9	14981.9	27.7	3.4	5.1
2008	176.7	154.6	4.4	39.0	32.3	422.8	674.3	12085.2	22.3	2.7	4.1
2009	178.8	156.4	4.5	39.4	32.6	427.7	682.0	12223.3	22.6	2.8	4.2
2010	257.9	225.7	6.5	58.3	47.6	624.5	987.7	17720.3	33.0	4.0	6.1
2011	188.9	165.3	4.7	41.7	34.5	452.0	720.7	12917.7	23.9	2.9	4.4
2012	389.0	340.4	9.6	83.6	70.2	919.4	1478.6	26473.6	48.6	6.0	9.0
2013	260.7	228.1	6.5	57.5	47.6	623.8	994.7	17828.1	33.0	4.0	6.1
2014	332.4	290.8	8.3	73.3	60.7	795.2	1268.1	22729.0	42.0	5.1	7.8
2015	109.9	96.1	2.8	24.2	20.1	262.9	419.3	7514.6	13.9	1.7	2.6

Table 2. Model performance statistics for concentrations of $PM_{2.5}$ and PM_{10} from observation and CMAQ simulation without OBB emissions (No_OBB) and with OBB emissions based on traditional bottom-up (Traditional_OBB), FRP-based (FRP_OBB) and constraining methods (Constrained_OBB) for the ~~two~~ three OBB events of June 2010 ~~and~~ , 2012 and 2014.

	June 2010		June 2012	
	~~NMB~~	~~NME~~	~~NMB~~	~~NME~~
~~No_OBB~~	~~-47%~~	~~50%~~	~~-60%~~	~~68%~~
~~Traditional_OBB~~	~~11%~~	~~44%~~	~~-16%~~	~~45%~~
~~FRP_OBB~~	~~-33%~~	~~41%~~	~~-45%~~	~~52%~~
~~Constrained_OBB~~	~~-16%~~	~~37%~~	~~-10%~~	~~45%~~

			No_OBB		Traditional_OBB		FRP_OBB		Constrained_OBB	
			NMB	NME	NMB	NME	NMB	NME	NMB	NME
2010	PM_{10}	Daily	-47%	50%	11%	44%	-33%	41%	-16%	37%
2012	PM_{10}	Daily	-60%	68%	-16%	45%	-45%	52%	-10%	45%
2014	PM_{10}	Daily	-59%	59%			-54%	54%	-37%	42%
		Hourly	-59%	60%			-54%	57%	-37%	52%
	$PM_{2.5}$	Daily	-52%	52%			-41%	42%	-12%	39%
		Hourly	-52%	56%			-41%	51%	-13%	54%
Bench-mark	PM_{10}								-45%	49%
	$PM_{2.5}$								-33%	43%

Note: [a] from Zhang et al. (2006). NMB and NME were calculated using following equations (P and O indicate the results from modeling prediction and observation, respectively):

$$NMB = \frac{\sum_{i=1}^{n}(P_i - O_i)}{\sum_{i=1}^{n}(O_i)} \times 100\%;\quad NME = \frac{\sum_{i=1}^{n}|P_i - O_i|}{\sum_{i=1}^{n}(O_i)} \times 100\% \; .$$

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Table 3. OBB emissions in YRD derived from this work and other studies in 2010 (Unit: Gg).

	PM_{10}	$PM_{2.5}$	EC	OC	CH_4	NMVOCs	CO	CO_2	NOx	SO_2	NH_3
Traditional (this work)	362.4	317.1	9.3	85.7	67.9	154.9	1391.8	24978.0	47.0	5.4	8.7
FRP-based (this work)	57.8	50.6	6.4	18.5	46.5	412.5	820.1	12718.0	24.9	3.2	17.7
FRP-based (WSE)[1]	158.6	139.1	4.1	38.5	30.1	68.7	612.8	11004.3	20.9	2.4	
Constrained (this work)	257.9	225.7	6.5	58.3	47.6	624.5	987.7	17720.3	33.0	4.0	6.1
GFASv1.0	-	17.8	1.0	9.5	15.6	88.7	196.3	3097.8	5.1	1.0	3.1
GFEDv3.0	-	3.5	0.2	1.7	3.2	4.1	39.4	701.6	1.1	0.2	6.4
GFEDv4.1s	-	33.6	4.0	12.4	31.3	53.2	548.3	8519.7	16.7	2.2	11.7
Xia et al, (2016)	350.2	339.3	14.8	137.8	-	-	1989.9	49835.1	134.3	22.6	-

[1] FRP-based (WSE)~~,~~: the OBB emissions were ~~derived~~estimated with FRP-based method, applying ~~–~~the same emission factors ~~as~~ used in the bottom-up method ~~in FRP-based method and t~~. The emission factors were obtained by weighting emission factors in the bottom-up method with the masses of various crop ~~straws of different crop species~~ types.

带格式表格

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Table 4. The uncertainties of OBB emissions in YRD indicated as 95% CIs and the top two parameters contributing most to emission uncertainties based on traditional bottom-up and FRP-based methods for 2012. The percentages in the parentheses indicate the contributions of the parameters to the variances of emissions.

	Traditional bottom-up method		FRP-based method	
PM_{10}	-56%, +70%	PCRBF[1] $_{Anhui}$（42%） EF_{wheat} (41%)	-77%, +274%	EF (76%) AF[2] (11%)
$PM_{2.5}$	-56%, +70%	$PCRBF_{Anhui}$（43%） EF_{wheat} (41%)	-63%, +244%	EF (65%) NFP[3] (16%)
EC	-50%, +54%	$PCRBF_{Anhui}$（69%） $PCRBF_{Jiangsu}$（11%）	-78%, +281%	EF (75%) NFP (11%)
OC	-54%, +73%	$PCRBF_{Anhui}$（42%） EF_{rice} (37%)	-78%, +276%	EF (75%) NFP (11%)
CH_4	-49%, +58%	$PCRBF_{Anhui}$（65%） $PCRBF_{Jiangsu}$（11%）	-83%, +315%	EF (79%) NFP (9%)
NMVOCs	-48%, +59%	$PCRBF_{Anhui}$（64%） $PCRBF_{Jiangsu}$（10%）	-63%, +243%	EF (65%) NFP (16%)
CO	-46%, +73%	$PCRBF_{Anhui}$（62%） $PCRBF_{Jiangsu}$（10%）	-52%, +223%	EF (57%) NFP (19%)
CO_2	-48%, +60%	$PCRBF_{Anhui}$（69%） $PCRBF_{Jiangsu}$（10%）	-21%, +164%	NFP (44%) AF (42%)
NO_X	-47%, +87%	$PCRBF_{Anhui}$（51%） EF_{wheat} (23%)	-82%, +303%	EF (78%) NFP (10%)
SO_2	-59%, +138%	EF_{wheat} (35%) $PCRBF_{Anhui}$（27%）	-78%, +279%	EF (74%) NFP (12%)
NH_3	-51%, +67%	$PCRBF_{Anhui}$（55%） EF_{wheat} (12%)	-82%, +302%	EF (79%) NFP (10%)

[1] PCRBF, the percentage of crop residues burned in the field (the subscript indicates province); [2] AF, the average FRE of fire pixels; [3] NFP, the number of fire pixels; [4] MCRBF, the mass of crop residues burned in the field.

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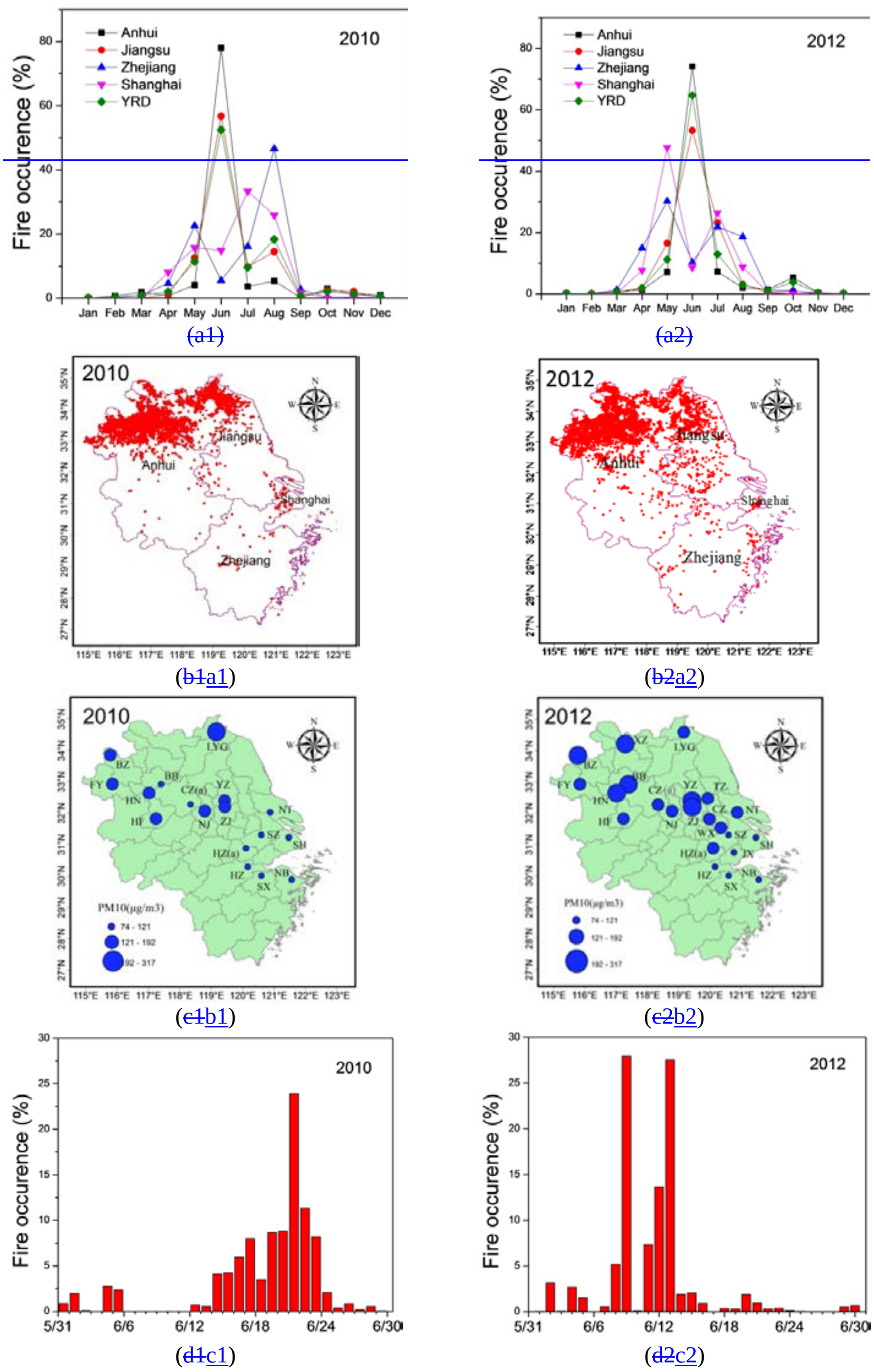
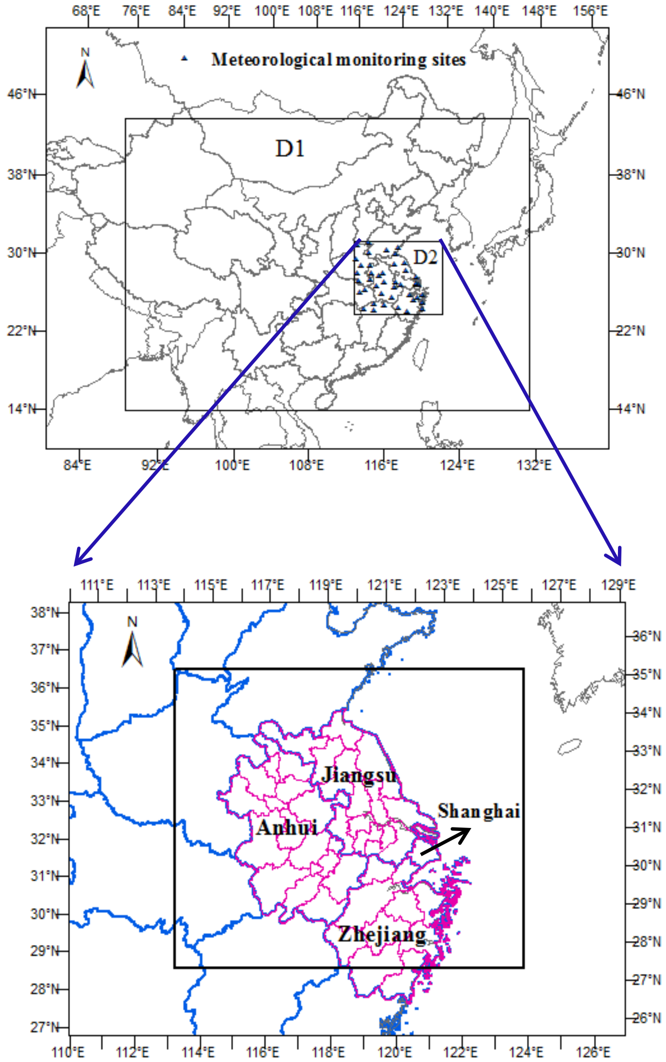
Figure 1.



2010
2012
Anhui
Jiangsu
Zhejiang
Shanghai
YRD
Fire occurence (%)
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec
(a1)
(a2)
Anhui
Jiangsu
Shanghai
Zhejiang
(b1a1)
(b2a2)
PM10(μg/m3)
74 - 121
121 - 192
192 - 317
(c1b1)
(c2b2)
5/31 6/6 6/12 6/18 6/24 6/30
(d1c1)
(d2c2)

1093 **Figure 2.**



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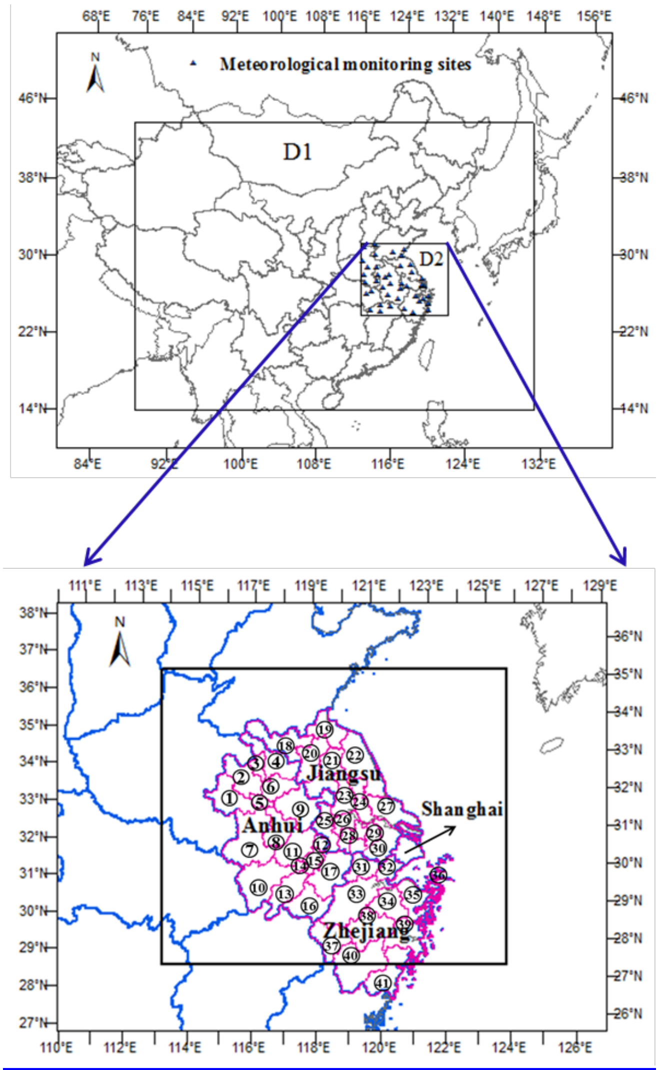
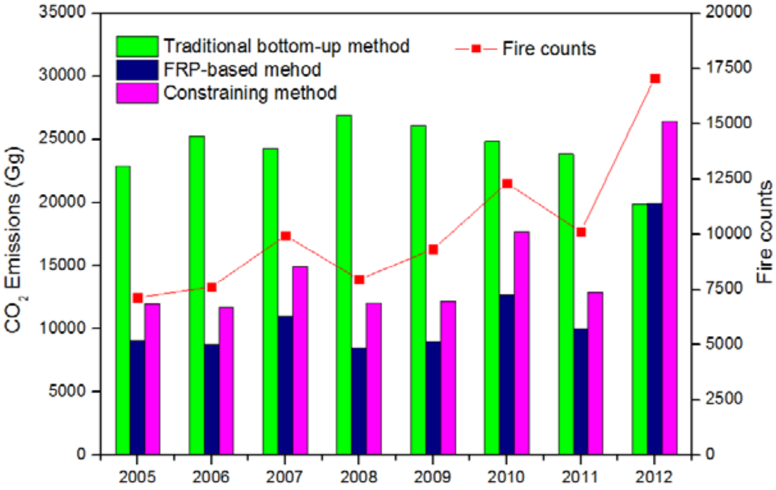


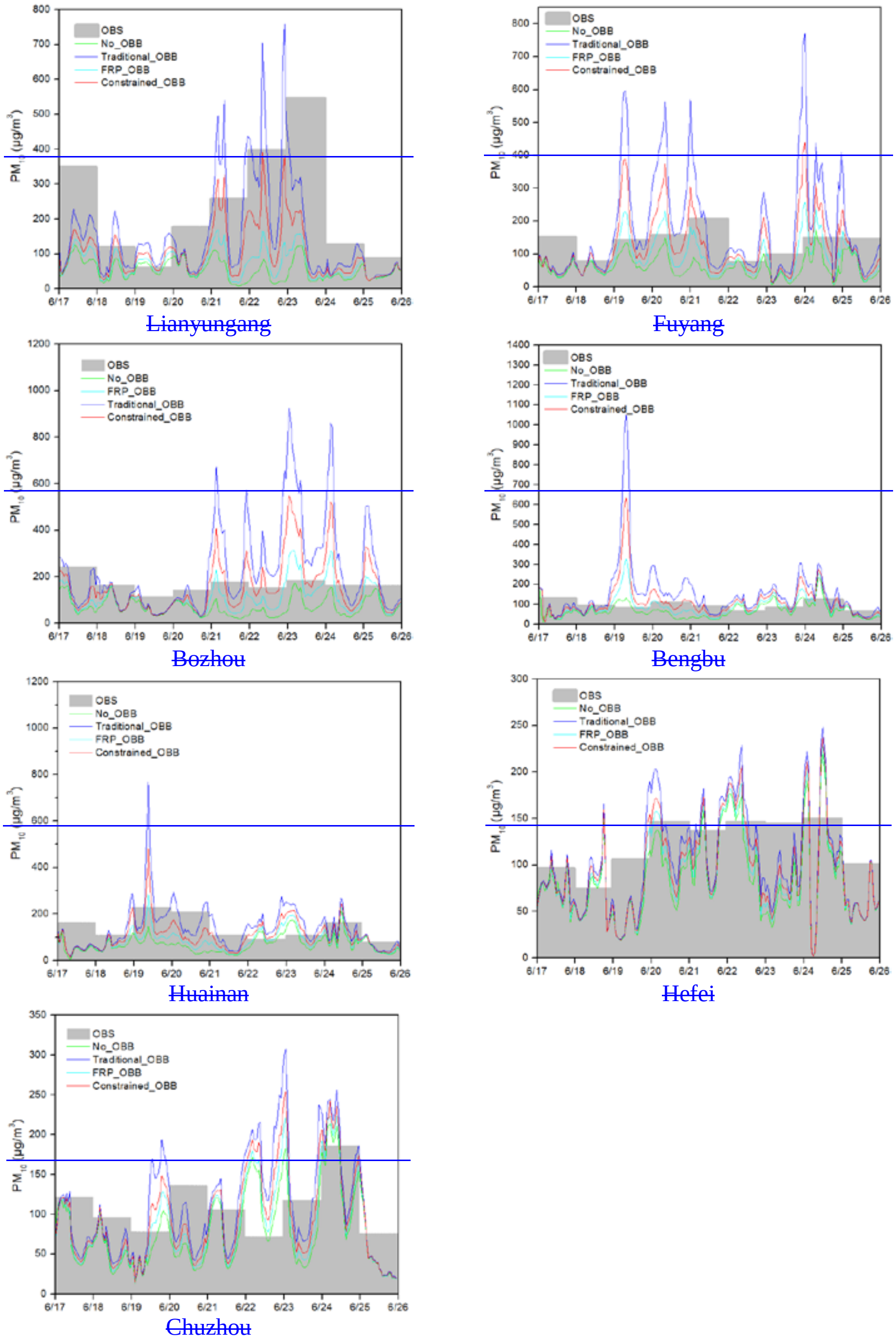
Meteorological monitoring sites
D1
D2
Jiangsu
Anhui
Zhejiang
Shanghai

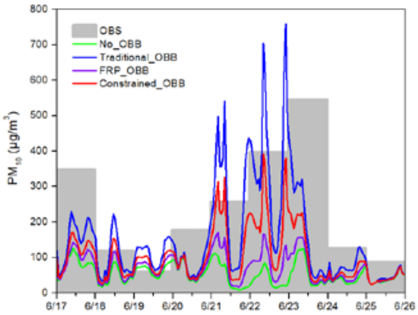
Figure 3.



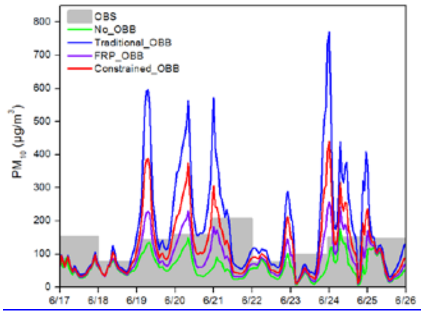
Traditional bottom-up method
FRP-based mehod
Constraining method
Fire counts
CO2 Emissions (Gg)
Fire counts
35000
30000
25000
20000
15000
10000
5000
0
20000
17500
15000
12500
10000
7500
5000
2500
0
2005
2006
2007
2008
2009
2010
2011
2012

Figure 4.

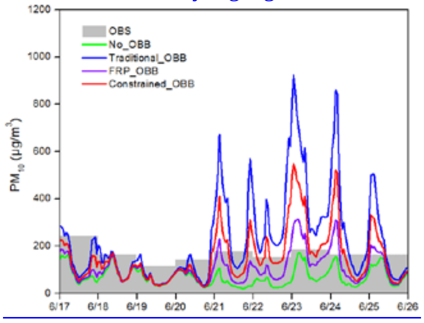

OBS
No_OBB
Traditional_OBB
FRP_OBB
Constrained_OBB
PM10 (μg/m3)
Lianyungang
Fuyang
Bozhou
Bengbu
Huainan
Hefei
Chuzhou



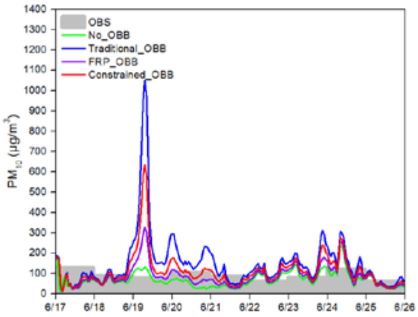
Lianyungang



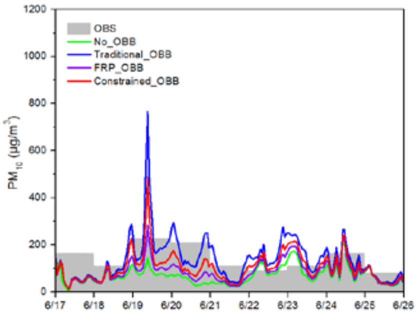
Fuyang



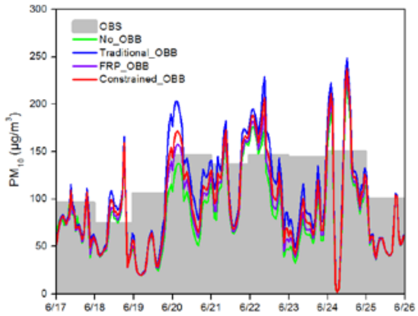
Bozhou



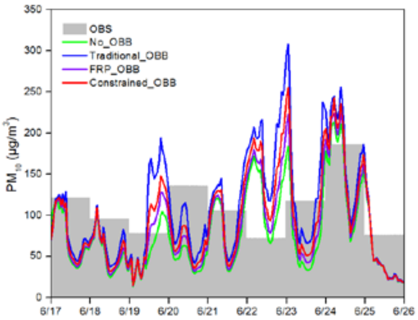
Bengbu



Huainan

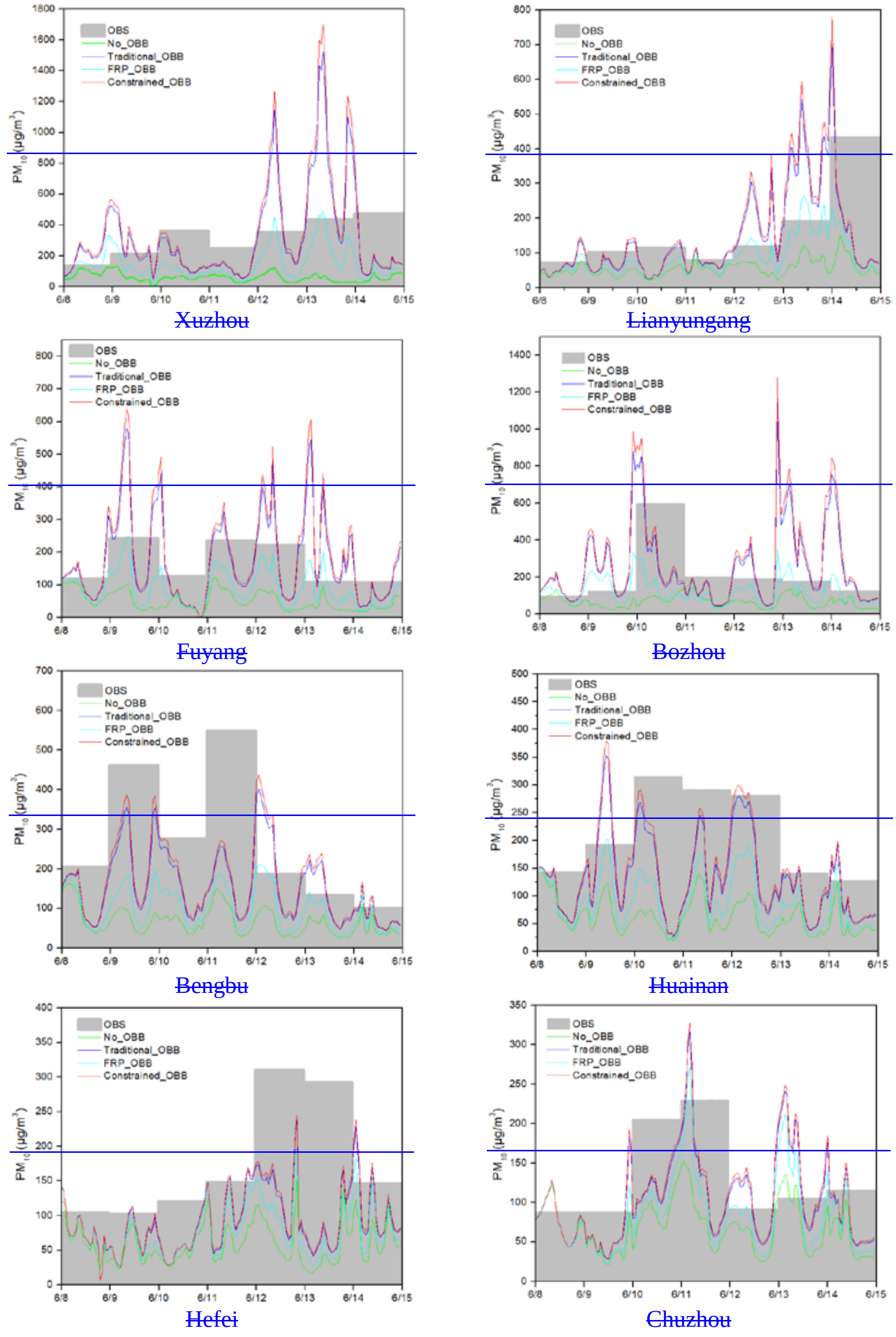


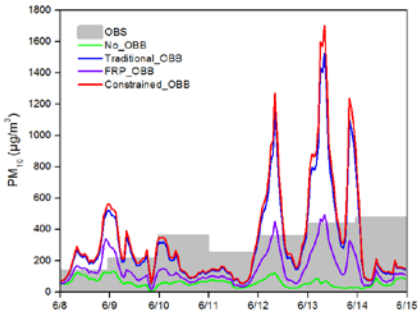
Hefei



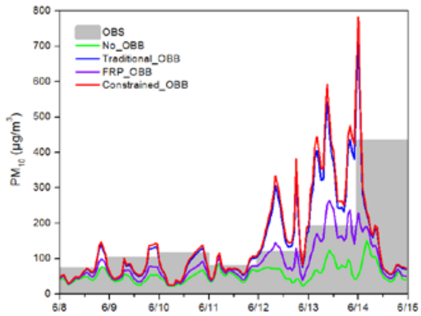
Chuzhou

Figure 5.

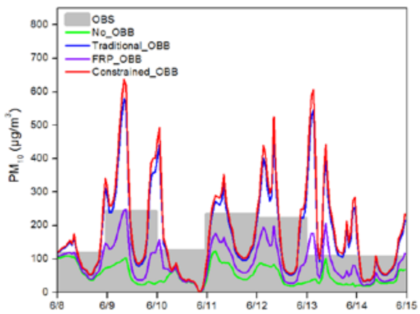

OBS
No_OBB
Traditional_OBB
FRP_OBB
Constrained_OBB
PM10 (μg/m3)
6/8
6/9
6/10
6/11
6/12
6/13
6/14
6/15
Xuzhou
Lianyungang
Fuyang
Bozhou
Bengbu
Huainan
Hefei
Chuzhou



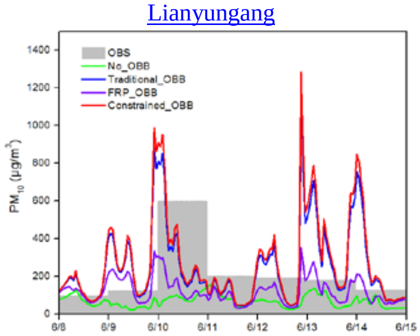
Xuzhou



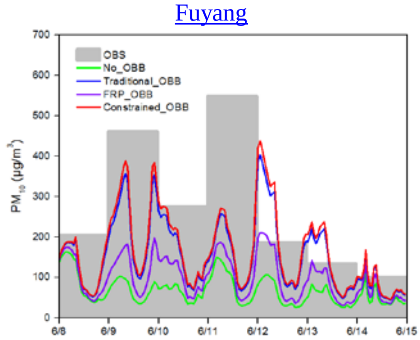
Lianyungang



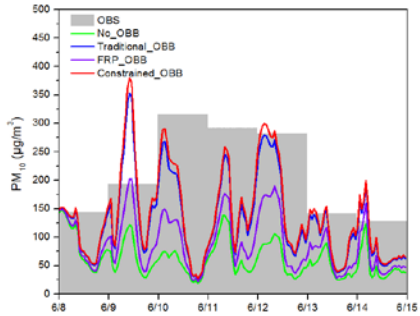
Fuyang



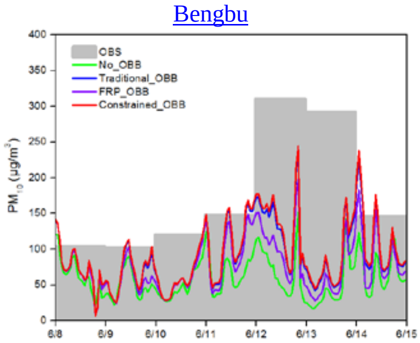
Bozhou



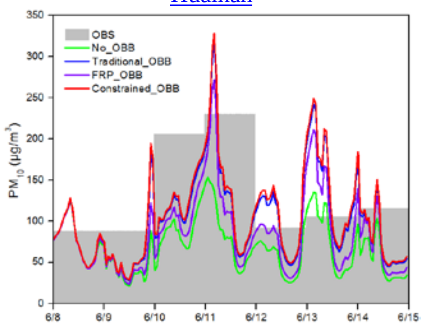
Bengbu



Huainan



Hefei



Chuzhou

Figure 6.

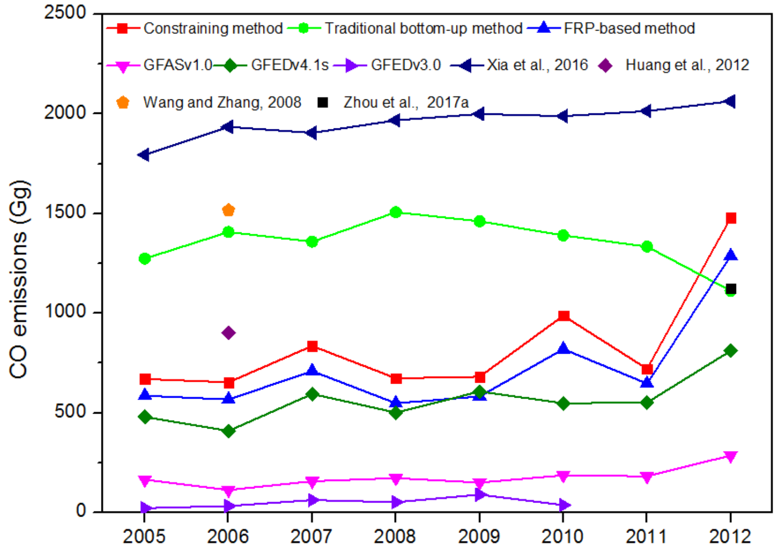

Constraining method
Traditional bottom-up method
FRP-based method
GFASv1.0
GFEDv4.1s
GFEDv3.0
Xia et al., 2016
Huang et al., 2012
Wang and Zhang, 2008
Zhou et al., 2017a
CO emissions (Gg)
0
500
1000
1500
2000
2500
2005
2006
2007
2008
2009
2010
2011
2012

Figure 7.

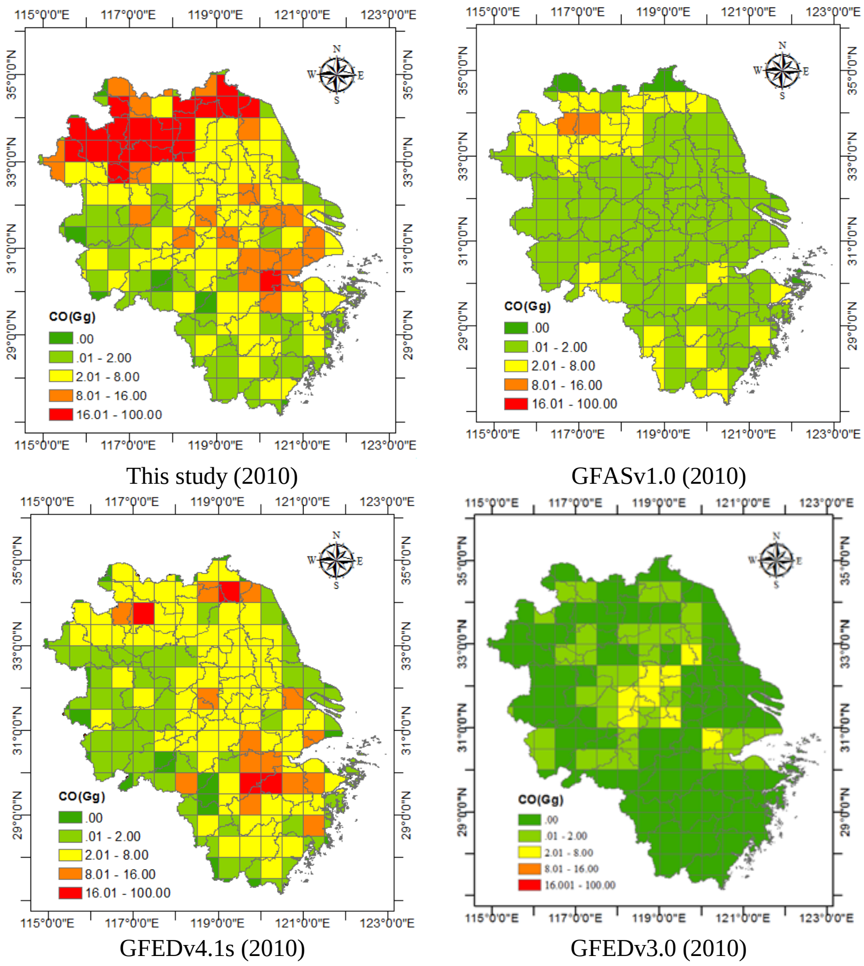
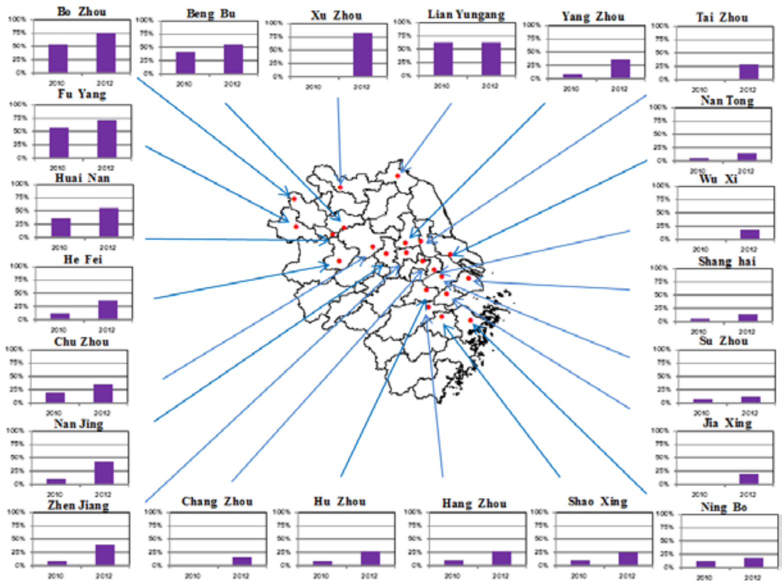
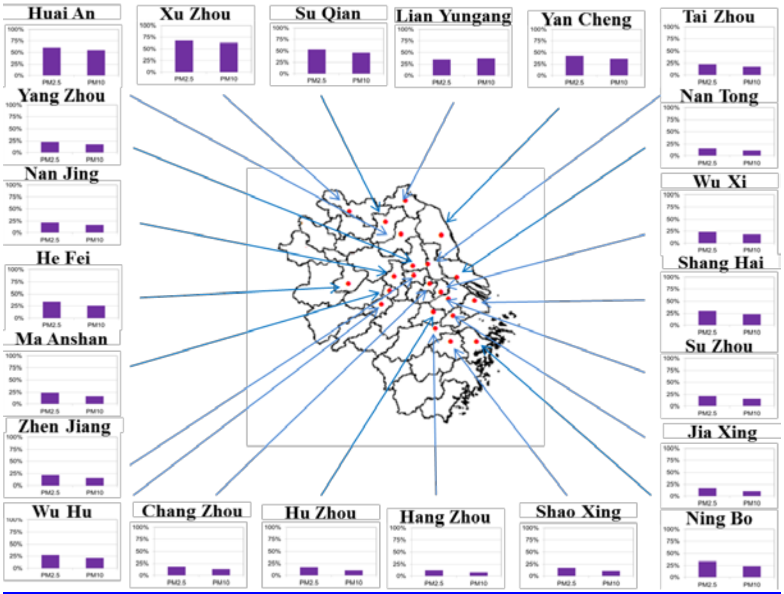

115°0'0"E
117°0'0"E
119°0'0"E
121°0'0"E
123°0'0"E
35°0'0"N
33°0'0"N
31°0'0"N
29°0'0"N
N
S
W
E
CO(Gg)
.00
.01 - 2.00
2.01 - 8.00
8.01 - 16.00
16.01 - 100.00
This study (2010)
GFASv1.0 (2010)
GFEDv4.1s (2010)
16.001 - 100.00
GFEDv3.0 (2010)

Figure 8.


Bo Zhou
Beng Bu
Xu Zhou
Lian Yungang
Yang Zhou
Tai Zhou
Fu Yang
Nan Tong
Huai Nan
Wu Xi
He Fei
Shang hai
Chu Zhou
Su Zhou
Nan Jing
Jia Xing
Zhen Jiang
Chang Zhou
Hu Zhou
Hang Zhou
Shao Xing
Ning Bo
100%
75%
50%
25%
0%
2010
2012

1116 **Figure 9.**



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1118

Figure ~~9~~10.

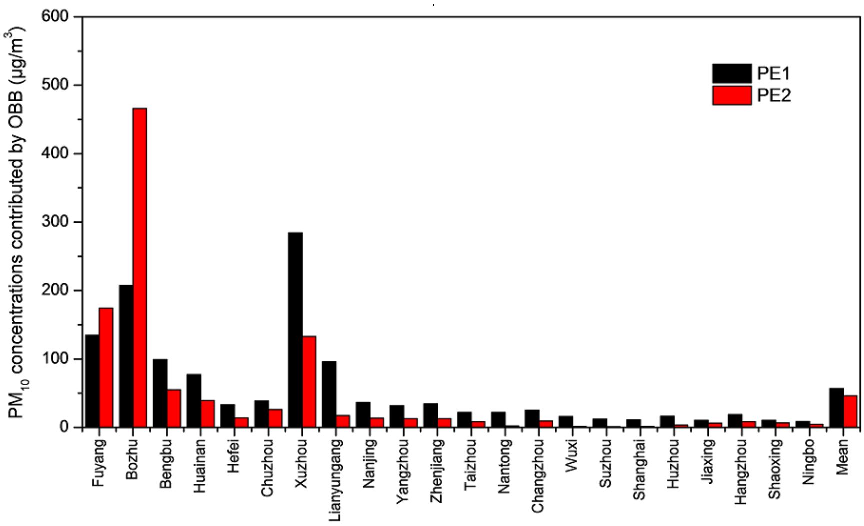
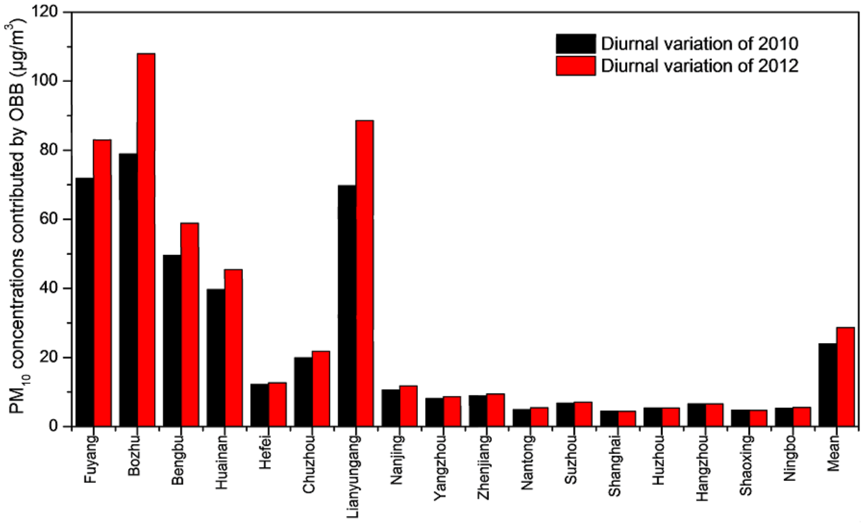

PM10 concentrations contributed by OBB (μg/m3)
600
500
400
300
200
100
0
PE1
PE2
Fuyang
Bozhu
Bengbu
Huainan
Hefei
Chuzhou
Xuzhou
Lianyungang
Nanjing
Yangzhou
Zhenjiang
Taizhou
Nantong
Changzhou
Wuxi
Suzhou
Shanghai
Huzhou
Jiaxing
Hangzhou
Shaoxing
Ningbo
Mean

Figure ~~10~~11.



PM$_{10}$ concentrations contributed by OBB (μg/m^3)
0
20
40
60
80
100
120
Diurnal variation of 2010
Diurnal variation of 2012
Fuyang
Bozhu
Bengbu
Huainan
Hefei
Chuzhou
Lianyungang
Nanjing
Yangzhou
Zhenjiang
Nantong
Suzhou
Shanghai
Huzhou
Hangzhou
Shaoxing
Ningbo
Mean