

Interactive comment on “Quantification and evaluation of atmospheric pollutant emissions from open biomass burning with multiple methods: A case study for Yangtze River Delta region, China” by Yang Yang and Yu Zhao

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Received and published: 10 November 2018

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.

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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 PM₁₀ 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

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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 $0.25^{\circ} \times 0.25^{\circ}$. 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.

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4. Considering that the PM emissions from OBB are mainly PM_{2.5}, and the ambient PM₁₀ is more affected by the local road dust emissions, it is not appropriate to only use PM₁₀ concentration to evaluate the model performance and analyze the contribution of OBB. I think authors shall use both PM₁₀, PM_{2.5}, CO, NO₂, SO₂, 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, NO₂, and SO₂ were unavailable before 2013. The largest daily mass ratio of PM_{2.5} to PM₁₀ 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 PM₁₀ 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 PM₁₀ during the OBB event periods as well. Therefore, we used PM₁₀ concentration to evaluate the model performance and analyze the contribution of OBB in 2010 and 2012. Compared to PM_{2.5} and PM₁₀, OBB was not a major source of NO₂ and SO₂, 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 PM₁₀ 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 PM₁₀ during June

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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 PM₁₀ 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 PM₁₀ as well, we had to apply PM₁₀ concentrations to constrain the OBB emissions. The activity level was constrained based on the comparisons between simulated and observed PM₁₀ 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 PM₁₀ 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

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different regions. We selected the results in US (Zhang et al., 2006) as the benchmark for PM_{2.5} and PM₁₀ 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 PM₁₀ 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.

Interactive comment on Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2018-701>, 2018.

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