Reviewer #2

0. *In this study, Zhao et al. uses ground-based elemental carbon (EC) measurements from two sites in eastern China to evaluate and constrain black carbon (BC) emissions from two bottom-up inventories: a national/regional inventory for China (MEIC) and a high-resolution inventory for city clusters in southern Jiangsu Province. Both inventories include emissions from transportation, industry, power generation, and the residential sector. The authors show that the posterior emission estimates, constrained by ground measurements, are much smaller than the prior emission estimates, suggesting that pollution control measures by the Jiangsu government have effectively reduced emissions of BC. They also show results from various sensitivity tests, including those on the number of observation sites, spatial representativeness of observation sites, a priori emission inventories, and wet deposition. Overall, this is an interesting study that can be potentially useful for air quality modeling and management, emission inventory development and evaluation, and also studies on regional aerosol effects. Through several fairly detailed sensitivity tests, the authors also demonstrate that the differences between a priori and posteriori emission estimates are robust. However, the paper is overly long (and needs some improvement in presentation quality) and some reorganization may help. And there are also some concerns about the methodology that need to be addressed before this paper can be published in ACP.*

Response and revisions:

We appreciate the reviewer's remarks on the importance of the work. We reorganized Figures and Tables following the reviewer's suggestions (please see our response to Q3 and Q7) and specified the methodology of top-down estimate (please see our response to Q1-2). Please see the details in the following response and revision list to the reviewer's comment.

1. *Major comments: It is not quite clear whether emissions outside of Jiangsu*

Province (but within the model domains) are scaled or not. Given the location of the sites, they could be strongly influenced by emissions from nearby provinces. If different local governments implemented different pollution control measures but the same domain wide scaling factors are used for emissions, that may lead to biases in the final estimated emissions for southern Jiangsu.

Response and revisions:

We appreciate the reviewer's important comment. For MEIC-prior and JS-prior, emissions from different provinces and cities within the modeling domain were scaled based mainly on changes in their respective activity levels from 2012 to 2015, including those outside of Jiangsu Province. However, we did not constrain the emissions outside of Jiangsu Province in the top-down method, and we agree the limitation here. The main reason is that there were very few BC observation data available in the cities outside southern Jiangsu. Using observations at NJU or PAES to constrain emissions from those cities would bring more uncertainty for the cases in which local emissions dominated the air quality. Given this limitation, therefore, more measurements with better spatial coverage were recommended to be conducted and published for constraining BC emissions effectively in the future. We discussed this **in lines 545-547 in the revised manuscript**.

The uncertainty of using observations at two sites to constrain emissions from southern Jiangsu was expected to be insignificant in this work. Located in the downwind of the Yangtze River Delta region (YRD), NJU is more representative for the emissions from western YRD through regional transport. PAES is in urban Nanjing and its air quality is commonly influenced by surrounding transportation and residential sources, thus PAES is representative for the local emissions of Nanjing. We quantified the contribution of Nanjing and Suzhou-Wuxi-Changzhou-Zhenjiang city cluster through the brute-force method **in Sector 4.1 in the revised manuscript**. As can be seen **in Figure S10 in the revised supplement**, the monthly mean contributions of the emissions from the two regions in April were aggregated at 54% and 59% at NJU and PAES respectively. We thus believe it is reasonable to use observations at two sites to constrain emissions from southern Jiangsu.

Regarding the influence of emissions outside southern Jiangsu, the contribution of each sector (c_{power}, $c_{industry}$, $c_{residential}$, and $c_{transportation}$) **in Eq1 in the revised manuscript** was simulated when the emissions from that sector were zeroed out for the whole third domain. It means that the emissions outside southern Jiangsu were also considered in the multiple regression model to obtain scaling factors. We applied the scaling factors to constrain emissions from southern Jiangsu only while remaining emissions outside southern Jiangsu unchanged so that it could better quantify the improvement of modeling performance at two sites due to the top-down estimate in southern Jiangsu. We acknowledge the uncertainty of including emissions of the whole third domain in the multiple regression model, due to different implementation of pollution control measures by city. As shown in Table R1, we compared the reduction rates of monthly BC emissions in the national inventory MEIC from 2012 to 2015 inside and outside southern Jiangsu in the domain. The difference between the two regions was less than 6%, implying the similar progress of pollution control measurements in two regions. Due to limited BC observations, moreover, we also checked the annual reduction rates in $PM_{2.5}$ concentrations from 2013 to 2015 for cities in the third domain based on the observation data from China National Environmental Monitoring Center (http://www.cnemc.cn/). As shown in Table R2, the annual reduction rates were ranged from 10% to 17% by city, reflecting again the similar implementation of air pollution control policies around the regions. Relative statement was added **in lines 222-235 in the revised manuscript**, and Tables R1 and R2 were included as **Tables S2 and S3 in the revised supplement**.

Table R1. Reduction rates in monthly emissions from 2012 to 2015 in MEIC for southern Jiangsu and other regions within the third modeling domain (unit: %).

Region	Jan.	Apr.	Jul.	Oct.
Southern Jiangsu (%)	18	18	26	21
Outside southern Jiangsu (%)	12	16	21	15

Table R2. Reduction rates in annual $PM_{2.5}$ concentration for cities within the third modeling domain from 2013 to 2015 (unit: %).

Province	City	Reduction rate (%)
Anhui	Hefei	15.26
Jiangsu	Nantong	15.90
	Taizhou	11.76
	Yangzhou	16.84
	Nanjing	15.58
	Suzhou	12.76
	Wuxi	10.45
	Changzhou	12.31
	Zhenjiang	12.80
Shanghai	Shanghai	10.88

2. *The lack of biomass burning emissions can be concerning. Could the model underestimates of BC in July and particularly October be caused by the biomass burning (particularly agricultural fires)? How does the lack of biomass burning emissions affect the estimated emissions for other sectors?*

Response and revisions:

We thank the reviewer's comment. In both inventories (MEIC and JS), the emissions came from four sectors, including power generation, industry, residential sources and transportation, and the residential sources included fossil fuel and biofuel combustion. However, we did not include emissions from biomass open burning. In another paper of our group (Yang and Zhao, 2019), the emissions from biomass open burning in YRD were thoroughly evaluated with various methods, and the emissions were estimated to decrease by 60% from 2012 to 2015 in southern Jiangsu attributed mainly to the enhanced control of crop burning activities by the local government. With the optimized constrained method, the BC emissions from crop open burning were calculated at 0.83 Gg in southern Jiangsu 2015, contributing small in the JS-prior and JS-posterior at 3% and 6%, respectively. As shown in Table R3, in addition, the most intensive crop burning was found in May and August, indicated by the monthly fire points from satellite dectection. Limited effect of biomass burning was thus expected for the modeling periods in this study.

Table R3. Monthly fire points in southern Jiangsu for 2015, taken from Moderate Resolution Imaging Spectroradiometer (MODIS) Global Monthly Fire Location Product (MCD14ML).

2015	Jan.	Feb.	Mar.	Apr.	May	Jun.	Jul.	Aug.	Sep.	Oct.	Nov.	Dec.
Fire point	9	11	12	58	249	30	96	127	16	9	1	10

In this work, the scaling factor of residential sources in October was estimated at 1.52 in JS-posterior, implying the enhancement in BC emissions in autumn to JS-prior. The result thus implied that there were missing sources likely associated with crop waste burning in autumn, and it was discussed **in lines 420-424 in the revised manuscript**. We also evaluated the sensitivity of the constraining method to the initial emission input **in Section 4.2 in the revised manuscript**, and found the uncertainty from the a priori inventory had limited effects on the top-down estimate. To summarize, therefore, we believe that lack of biomass burning emissions in the initial inventories would not significantly bias the top-down estimation.

3. *The paper is overly long and can be better organized. In particular, if spatial representativeness and wet deposition are important, can the authors focus on the top-down estimates that consider both of these factors? Description of the other sensitivity tests can be brief. Also writing needs to be improved.*

Response and revisions:

We thank the reviewer’s comment. To make the manuscript concise, we moved Figures 9 and 10 in the original manuscript to the revised supplement (Figures S11 and S12) given that the near-linearity was also indicated in previous studies (Wang et al., 2013). We integrated the original Table 8 into Table 3 in the revised manuscript to summarize the modeling performances of different cases. The scaling factors and statistical indicators in Case 7 in the original Table 9 were integrated into Table 5, while emissions by sector in Case 7 and the relative deviations compared to

JS-posterior in Table 9 were integrated into Table 6. We moved the original Figure 3 that presents the seasonal variations in emissions of JS-prior, JS-posterior and MEIC-prior to the revised supplement (the new Figure S8) given the less statistical significant in seasonal patterns of several sectors in JS-posterior. We also moved the original Table 5 that summarizes the emissions from Nanjing and other cities in southern Jiangsu in different cases to Table S9 in the revised supplement. Sections 4.1 and 4.2 in the original manuscript were merged into one section (Section 4.1 in the revised manuscript) to evaluate the effects of number and spatial representativeness of observation sites on the top-down estimate. We believe the analysis on the uncertainty of the a priori inventory was important, as it could help judge the robustness of the constraining method. We found the influence of the a priori emissions was limited, and implied that the method could be potentially applied even if uncertainty existed in the bottom-up inventory. Therefore, we kept this part in the revised manuscript.

4. *Specific comments: Figure 3 and the paragraph starting from line 389: given that the scaling factor for April and Oct. are more uncertain (in terms of their statistical significance), are the seasonal patterns in the posterior emission estimates significant?*

Response and revisions:

We thank and agree with the reviewer's comment. Though the multiple regression model was statistically significant as a whole indicated by 0.00 of the overall significance in four months, the estimates for certain sources including industry in April and October and residential in April and July were more uncertain to some extent, as illustrated **in Table 1 in the revised manuscript**. It implied that the constrained emissions for those months/sources need to be cautiously applied in CTM and the seasonal patterns in those sectors could be less significant. Relevant discussion was **in lines 383-386 in the revised manuscript** and we moved original Figure 3 that presents the seasonal variations in emissions of JS-prior, JS-posterior and MEIC-prior to **Figure S8 in the revised supplement**.

5. *Figure 5a – what may have caused the model overestimates in mid-January at PAES? How does this period affect emission estimates? Can the authors exclude this period and compare the top-down estimates?*

Response and revisions:

We thank the reviewer's comment. The overestimation in January at PAES (especially in middle and late January, 16th–26th) may result from the emission control policy implemented for the National Memorial Day of Nanjing Massacre Victims in December 13th in 2014. During the period, Nanjing was undertaking series of stringent restrictions on air pollutant emissions. For example, key petrochemical and steel industries were shut down, and all the high-pollution vehicles were forbidden to drive in Nanjing. Those restrictions had large impacts on emissions and thereby air quality in the following month at PAES, but have not been fully considered in current emission inventories. Beside the emission control measures implemented in Nanjing, we evaluated the effect of planetary boundary layer (PBL) height on the modeling performance at PAES, as illustrated in Figure R1. Higher daily average PBL height was found for periods when the simulated concentrations were relatively lower (e.g., 6th -7th, 12th-15th and 28th-31st), resulting in smaller bias between simulations and observations. In contrast, the lower PBL height found in other periods would exaggerate the overestimation in simulated concentrations, given the elevated emissions in JS-prior. We added the analysis **in lines 454-468 in the revised manuscript** and included Figure R1 as **Figure S9 in the revised supplement**. Attributed to the instrument maintenance, moreover, the observation data in January at PAES were relatively insufficient, and the data points were 70% less than those at NJU. Therefore, the contribution of observation at PAES was limited in the multiple regression model.

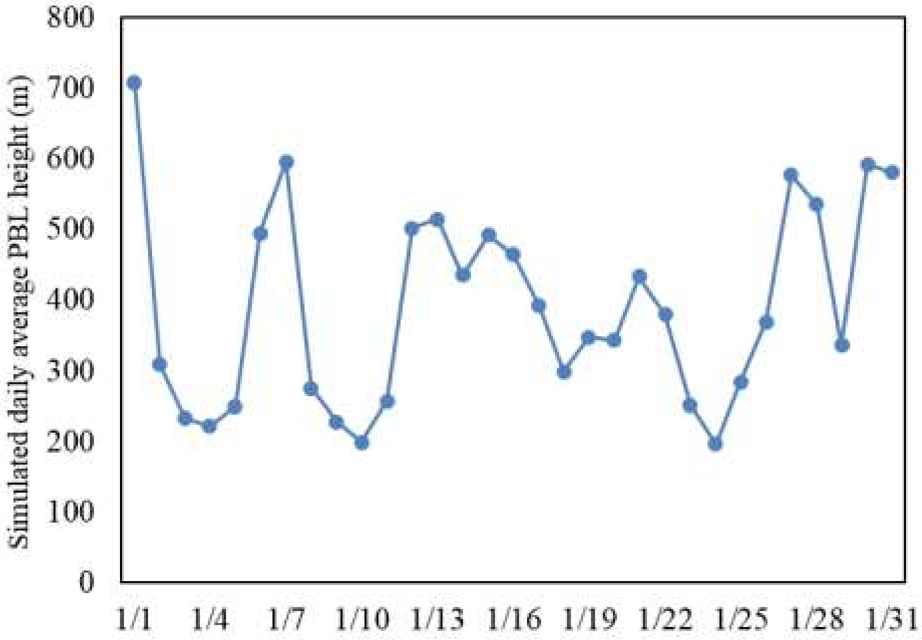

Figure R1. The simulated daily average PBL heights in January 2015 at PAES.

Following the reviewer's suggestion, we excluded the data points in middle and late January (16th -26th) at PAES and re-compared the observed and simulated BC concentrations. As shown in Table R4, the overestimation in CTM was largely reduced when the data were excluded, and the top-down estimate corrected the bias moderately at PAES. We added the discussions **in lines 499-503 in the revised manuscript** and added Table R4 as **Table S6 in the revised supplement**.

Table R4. Statistical indicators for observed and simulated BC concentrations using JS-prior and JS-posterior in January excluding data from 16th to 26th at PAES.

Site	Parameter	JS-prior	JS-posterior
PAES	Average SIM (μg/m^3)	2.86	2.68
	Average OBS (μg/m^3)	2.15	2.15
	NMB (%)	32.95	24.65
	NME (%)	52.61	49.63
	R	0.72	0.74

6. *Lines 456-461: again, could the model bias be due to the lack of biomass burning emissions?*

Response and revisions:

We thank the reviewer's comment. The bigger bias found in July and October at NJU when applying JS-posterior resulted mainly from the limitation of the constraining method. We used observations at two sites to constrain emissions from southern Jiangsu as a whole. Therefore, overestimation and underestimation in concentrations at different sites could not be corrected simultaneously without considering the spatial representation of observation sites, as discussed **in lines 511-516 in the revised manuscript**.

The underestimation in BC concentrations for July and October with JS-prior could be partly due to the lack of biomass open burning emissions. However, such influence was expected to be insignificant (please see our response to Q2), and the impact of the a priori emission input was found limited on the top-down estimation, as discussed **in Section 4.2 in the revised manuscript**.

7. *Tables: There are already many tables in the paper (and maybe not everyone is absolutely necessary). But a table that summarizes the different cases may be helpful for readers to keep track.*

Response and revisions:

We thank and follow the reviewer's comment to make the tables concise. We integrated the original Table 8 to a new Table 3 in the revised manuscript to summarize the modeling performance for different cases. For the original Table 9, moreover, the scaling factors and statistical indicators from the multiple regression model in Case 7 were integrated to Table 5, and the emissions by sector and the relative deviations to JS-posterior in Case 7 were integrated to Table 6. We also moved the original Table 5 that summarizes the emissions from Nanjing and other cities in southern Jiangsu in different cases to Table S9 in the revised supplement.

8. *Table 4 and related discussion on case 3: would the authors expect somewhat different driving conditions and emission factors for automobiles in urban and* *suburban settings? If so, is it still a valid assumption to assume the same scaling factor between NJU and PAES for transportation?*

Response and revisions:

We thank the reviewer's comment. In Case 3, we assumed a same scaling factor for transportation for different cities in southern Jiangsu to avoid the collinearity in the multiple regression model. As the observation data at NJU and PAES were applied to constrain emissions from Suzhou-Wuxi-Changzhou-Zhenjiang city cluster and Nanjing, respectively, the assumption of a same scaling factor at NJU and PAES did not mainly indicate the similar driving conditions or emission factors for automobiles in suburban and urban. Instead, it mainly indicated that the relative changes in emissions from transportation were similar across the cities in southern Jiangsu from 2012 to 2015. As we stated **in lines 591-593 in the revised manuscript**, such assumption is expected to be reasonable, because of the same progress of emission standard implementation (National Standard Stage IV) in southern Jiangsu and the frequent circulation of vehicles among the cities.

References

Wang, X., Wang, Y., Hao, J., Kondo, Y., Irwin, M., Munger, J. W., and Zhao, Y.: Top-down estimate of China's black carbon emissions using surface observations: Sensitivity to observation representativeness and transport model error, Journal of Geophysical Research: Atmospheres, 118, 5781-5795, 10.1002/jgrd.50397, 2013.

Yang, Y., and Zhao, Y.: Quantification and evaluation of atmospheric pollutant emissions from open biomass burning with multiple methods: a case study for the Yngtze River Delta region, China, Atmospheric Chemistry and Physics, 19, 327-348, 10.5194/acp-19-327-2019, 2019.