Reviewer #3

0. *This is a very nice and detailed work to constrain BC emissions in southern Jiangsu. The approach and uncertainty analysis may be applied to other regions. The paper is well written in general, suitable for ACP. Below are a few suggestions to further improve the paper.*

Response and revisions:

We appreciate the reviewer's positive remarks on the importance of the work. Please see the details in the following response and revision list to reviewer's comment.

1. *It would be nice to discuss in the conclusion section the potential of applying the method to other regions.*

Response and revisions:

We thank the reviewer's comment. The method could be applied to constrain the BC emissions for other regions effectively if there are sufficient observation data with satisfying spatiotemporal coverage. We added the statement **in lines 796-799 in the revised manuscript**.

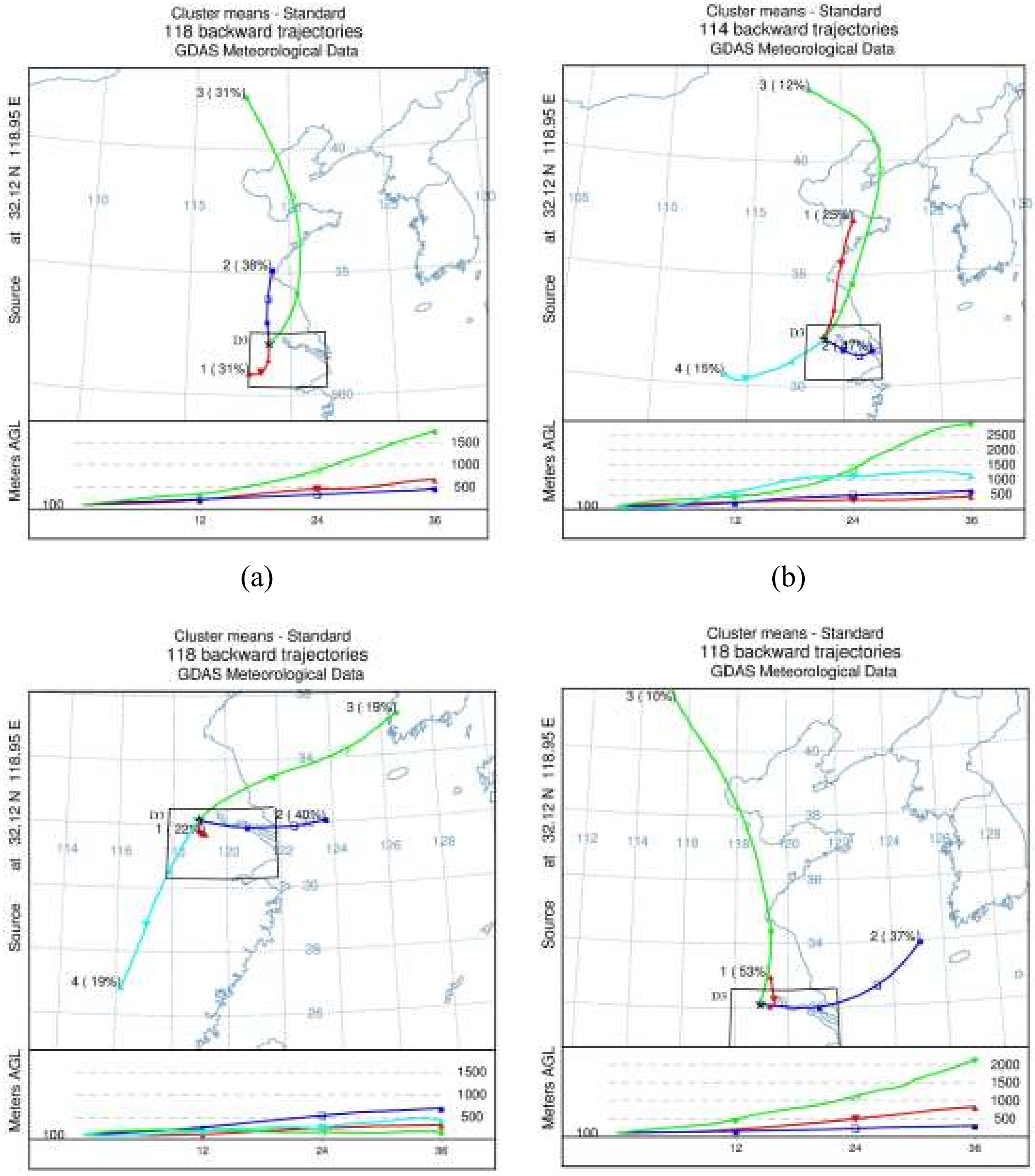
2. *The regression model needs to be further clarified. Are the scaling factors (betta) for each month, day, or hour? Why is there not a term in Eq. 1 for the background (e.g., lateral boundary condition) reflecting the effect of horizontal transport from regions other than southern Jiangsu? Table S3 and Fig. S7 show that the sum of southern Jiangsu contributions is much smaller than 100%, implying a large contribution from regions other than southern Jiangsu.*

Response and revisions:

We thank the reviewer's comment. The scaling factors were obtained for each month and used to constrain the monthly emissions in southern Jiangsu. We clarified it **in lines 235-237 in the revised manuscript**.

Regarding the background reflecting the regional transport, c_{power}, $c_{industry}$, $c_{residential}$ and $c_{transportation}$ in the multiple regression model were simulated by brute-force method in CTM in which emissions from corresponding sector in the third domain were zeroed out. Therefore the contributions of emissions outside southern Jiangsu in the third domain were considered in the model. Moreover, ε reflected the effect of background conditions (e.g., emissions in the first and second domain in CTM and emissions not included in the a priori inventory like those from natural sources). We clarified it **in lines 222-227 and 237-239 in the revised manuscript**. For example, the ε was estimated at 0.96 μg/m^3 in the multiple regression model for April in JS-posterior. By zeroing out the emissions from the third domain in CTM, the monthly contribution from boundary conditions were calculated at 0.76 and 0.77 μg/m^3 at NJU and PAES, respectively. In spite of the modest bias between ε and the estimated contribution of boundary conditions, including ε would reduce the uncertainty of the multiple regression model.

We added the contributions from four sectors in the third domain at the two sites **in Table S5 in the revised supplement**. The total contributions were larger than 50% for all the months and sites except for January. We assumed that the smaller contributions in January resulted partly from the longer lifetime of BC in winter due to less wet deposition. We also identified the transport pathways of air masses sampled at NJU for the four months through cluster analysis of back trajectories with Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT, version 4) model as illustrated in Figures R1. Compared to other months, fewer air masses passed through the third modeling domain in January due to the prevailing northerly wind, implying more contribution from regional transport to the air quality at the site in January. Similar results were found for other region. Jia et al. (2008) estimated that regional transport on average contributed nearly 50% of PM (up to 70% in southerly regions) in winter in three sites in Beijing. Sun et al. (2014) considered the accumulation of local BC emissions and estimated a contribution of 53% from regional transport to BC in Beijing. Given the smaller contribution of emissions within the third domain in January, we acknowledged that the multiple regression model was less effective on identifying the sources of BC in winter by constraining the emissions in southern Jiangsu city cluster alone. We added the discussion **in lines 360-370 in the revised manuscript** and included Figure R1 as **Figure S6 in the revised supplement**.



(a) (b)

(c) (d)

Figure R1. The transport pathways of air masses sampled at NJU based on cluster analysis of back trajectories in HYSPLIT model in January (a), April (b), July (c) and October (d).

3. *The idea of testing the spatial representativeness of measurements is very nice. Given the spatial representativeness difference between the two sites, is it possible to use Case 3 as your best case? Alternatively, it would be nice to improve the regression model by taking into account the transport path, e.g., by basing on WRF modeled winds to design a model that considers the trajectory of air movement. The much higher bias in JS-posterior than JS-prior in Case 1, which is a concern, is related to this spatial representativeness issue.*

Response and revisions:

We thank the reviewer's comment. Among all the cases discussed in the paper, the best CTM performance was obtained in Case 3 in which observations at both sites were used with their difference in spatial representativeness considered in the constraining method. We also appreciate the reviewer's suggestion, which could potentially improve the analysis of spatial representativeness and could be applied with more observation data available in the future. The larger NMEs in July and October at NJU in JS–posterior than JS-prior were related to the spatial representativeness issue, which was discussed **in lines 511-516 in the revised manuscript**.

4. *A clearer discussion of temporal resolution in bottom up inventories and how this resolution affects the top-down constraint will be very helpful.*

Response and revisions:

We thank the reviewer's comment. We derived the hourly bottom-up emission inventory for CTM. The monthly distributions of emissions from power plants and industry plants in JS-prior were dependent on those of electricity generation and typical industrial production, respectively. Such information was investigated by Zhou et al. (2017) according to the official statistics of the country (http://data.stats.gov.cn/). Meanwhile, the real-time monitoring on urban traffic in Nanjing was applied to allocate the temporal distribution of emissions from on-road vehicles in the whole regions in JS-prior. The weekly and hourly distributions of different sources in YRD (Li et al., 2011) were adopted to further allocate emissions in JS-prior. For MEIC-prior, we obtained the monthly emissions directly and applied the same weekly and hourly distributions as JS-prior. We described this **in lines 207-215 in the revised manuscript**. The temporal distributions based on local statistical data were expected to be more reliable in CTM than other information. Regarding the effect of the monthly variation on the constraint method, we compared top-down estimate derived from JS-prior and MEIC-prior in April, respectively, **in Section 4.2 in the revised manuscript**. Similar emission estimation, spatial distribution and modeling performance were found for the two a posteriori emissions, even clear difference existed in the two a priori inventories. The result thus implied the insignificant effect of monthly variation of emissions on the top-down constraint. We discussed this **in lines 667-671 in the revised manuscript**. We did not constrain the hourly emissions in this study and the hourly distribution was thus unchanged in the top-down estimate.

5. *Comparison with near-surface measurements is sensitive to WRF/CMAQ modeled vertical processes, including the number of vertical layers within the PBL, the thickness of the first layer, and the model error in vertical mixing representation. WRF/CMAQ may have some issues with PBL mixing (Liu et al., 2018). Please specify these model setups. Please discuss the potential effect of model vertical resolution/mixing/transport errors on the BC constraint.*

Response and revisions:

We thank the reviewer's comment. The PBL module adopted in WRF 3.4 was ACM2, and the information was added **in line 285 in the revised manuscript**. There were 27 vertical layers in the model, with the heights of 54, 132, 234, 362, 523, 729, 974, 1417, 1887, 2385, 2914, 3900, 4890, 5886, 6885, 7885, 8891, 9907, 10946, 12000, 13070, 14158, 15278, 16441, 17662, 18966 and 20405 m, respectively. The simulated monthly average PBL heights along with the range of hourly simulations at NJU and PAES in four months were shown in Table R1. Therefore, there were average 5 vertical layers within the PBL. We found the similar result of the low simulated PBL height in WRF/CMAQ model as Liu et al. (2018) and the overestimation of BC concentration at PAES even after top-down constraint may result from it. We added the analysis **in lines 503-507 in the revised manuscript** and included Table R1 as **Table S7 in the revised supplement**.

The effect of vertical distribution on BC emission constraining was evaluated for Asia by Zhang et al. (2015). They repeated the top-down inversions using the OMI retrieval absorption aerosol optical depth (AAOD) based on the CALIOP and GOCART aerosol layer height and found the difference in the optimized BC emissions were less than 30% in April and 10% in October compared to the optimized emissions using the initial GEOS-Chem model. The difference was within the acceptable range compared with up to 500% enhancements in April and 10-50% in October with the top-down constraining. When applying ground observations in this study rather than column concentration in AAOD, the effect of vertical distribution could be smaller.

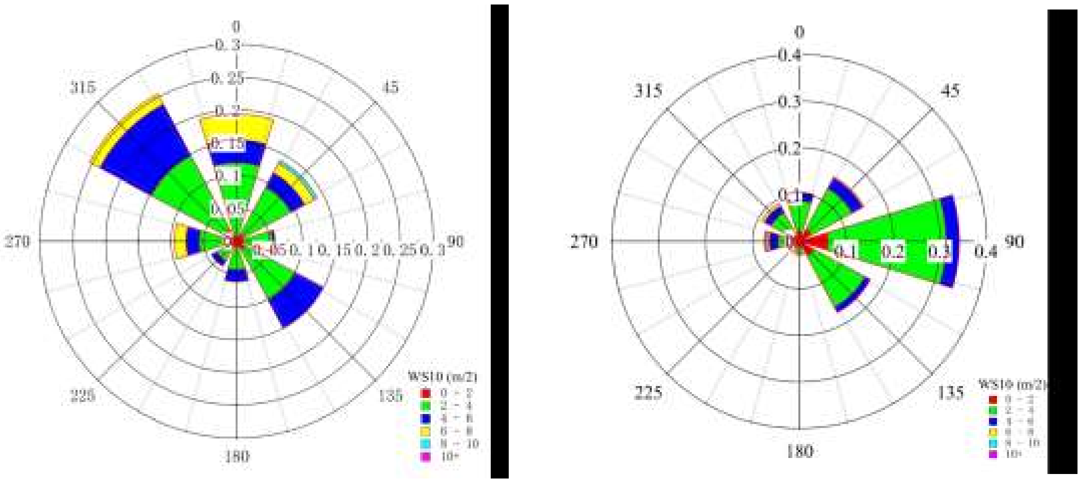
Table R1. The simulated monthly average PBL heights and the range of hourly simulations at NJU and PAES in four months.

Month	Site	Monthly average PBL (m)	Hourly average PBL (m)
January	NJU	370.25	27.59-1443.64
	PAES	384.56	27.20-1460.07
April	NJU	432.73	28.61-2157.87
	PAES	441.72	28.61-2157.87
July	NJU	381.14	30.70-1617.69
	PAES	431.02	30.02-1975.01
October	NJU	462.57	29.70-2065.97
	PAES	488.30	29.78-2073.46

6. *Table S2 shows that the prevailing winds in all three meteorological sites are southerly or southeasterly. I thought there would be northerly in the cold months (January and October). Please double check.*

Response and revisions:

We thank the reviewer’s comment. We checked the simulated and observed wind directions again and found the same result. The NMEs of wind directions were found below 40% at three meteorological stations in January and October, reflecting the robustness of the WRF modeling. In January, the average simulations and observations **in Table S4 in the revised supplement** did not mean that the prevailing winds were southerly. The values were the mean of the northerly wind directions ranging from 0-45° or 315-360°. Taking the wind directions at Hongqiao in January and October as examples, the prevailing winds were northerly and easterly in winter and autumn, respectively, as shown in Figures R2.



(a) (b)

Figure R2. Wind speeds and directions at Hongqiao in January (a) and October (b).

7. *Some paragraphs are too long and should be splitted, for example, L71-111, L352-388.*

Response and revisions:

We thank the reviewer’s comment. As suggested by the reviewer, we split

L71-111 in the initial manuscript into two parts, one was about the large uncertainties in bottom-up emission inventories, and the other was the challenge existing in updating BC inventories continuously, **in lines 74-114 in the revised manuscript**. We split L352-388 in the original manuscript and reorganized the paragraphs. One was about the relative change between JS-prior and JS-posterior, and the other was the detailed description about scaling factors for different sectors, **in lines 387-424 in the revised manuscript**.

8. *Abstract – please specify that monthly, sector-level and city-level emissions are optimized.*

Response and revisions:

We thank the reviewer's comment. We followed the suggestion and specified the optimized monthly, sector-level and city-level emissions in **in line 24 in the revised manuscript**.

9. *L22 – "observations," should be "observations" (no comma).*

Response and revisions:

We thank the reviewer's reminder and deleted the comma **in line 23 in the revised manuscript**.

10. *Abstract – please specify that WRF/CMAQ is used.*

Response and revisions:

We thank the reviewer's reminder and specified the WRF/CMAQ model **in lines 21-22 in the revised manuscript**.

11. *L214 – is there is term for background (due to horizontal transport)?*

Response and revisions:

We thank the reviewer's comment. ε reflected the effect of emissions from background conditions, which was added **in lines 237-239 in the revised manuscript**. (please also see our response to Q2).

12. *L218 – "domain-wide" – here you optimize the southern Jiangsu emissions, not the domain-wide emissions. Also, as suggested above, an improved regression model may be used to better account for spatial representativeness of measurements.*

Response and revisions:

We thank the reviewer's comment and revised the words for β_1-β_4 **in lines 235-237 in the revised manuscript**. We appreciate the reviewer's suggestion to improve the multiple regression model and it could be applied with more observation data available in the future to better consider spatial representativeness.

13. *L256 – "coordinated" should be "coordinate"*

Response and revisions:

We thank the reviewer's reminder and corrected the word **in line 281 in the revised manuscript**.

14. *L274-288 – please specify the temporal resolution of bottom up emissions.*

Response and revisions:

We thank the reviewer's comment. We specified the temporal distributions of two bottom-up emission inventories used in CTM **in lines 207-215 in the revised manuscript**. The monthly distributions of emissions from power plants and industry plants in JS-prior were dependent on those of electricity generation and typical industrial production, respectively. Such information was investigated by Zhou et al. (2017) according to the official statistics of the country (http://data.stats.gov.cn/). The real-time monitoring on urban traffic in Nanjing was applied to allocate the temporal distribution of emissions from on-road vehicles in the whole regions in JS-prior. The weekly and hourly distributions of different sources in the Yangtze River Delta (Li et al., 2011) were directly adopted to further allocate the emissions in JS-prior. For

MEIC-prior, we obtained the monthly emissions and applied the same weekly and hourly distributions as JS-prior. The temporal allocations based on local statistical data were expected to be more reliable in CTM.

15. *L283-285 – do you remove emissions in the whole domain, or just southern Jiangsu cities?*

Response and revisions:

We thank the reviewer's comment. We removed emissions in the whole third domain, and it was specified **in lines 308-310 in the revised manuscript**.

16. *L288 – "Scenarios B and S" should be "Scenarios B and S1-S4"*

Response and revisions:

We thank the reviewer's reminder and revised it **in line 313 in the revised manuscript**.

17. *L324 – "double" should be "twice"*

Response and revisions:

We thank the reviewer's comment and corrected the word **in line 349 in the revised manuscript**.

18. *L340 – "VIF smaller than 10" – the VIF values in the table are much smaller than 10.*

Response and revisions:

We thank the reviewer's reminder and revised it **in lines 374-376 in the revised manuscript**.

19. *L386-388 – this sentence is not clear*

Response and revisions:

We thank the reviewer's comment. Based on the bottom-up approach, Huang et al. (in preparation) incorporated detailed information and changes of individual sources, and estimated BC emissions for Nanjing from 2012 to 2015. The emissions in 2015 were estimated to decrease by 60% compared to those in 2012, and this relative change was close to that for the southern Jiangsu (a 50% reduction from JS-prior to JS-posterior) found in this study. The top-down method could thus capture the changes in emissions due to improved control measures. We revised the sentence **in lines 395-398 in the revised manuscript**.

20. *L418-442 – A figure would be much better than a table for this type of analysis.*

Response and revisions:

We thank the reviewer's comment. **Figures 3 and 4 in the revised manuscript** illustrated the simulated BC concentrations based on JS-prior and observations in four months at NJU and PAES, respectively. The analysis mentioned by the reviewer was reflected in those figures.

21. *L426 – what do you mean by "commonly"? The wording may be improved.*

Response and revisions:

We thank the reviewer's reminder and replaced the word commonly with generally **in line 472 in the revised manuscript**.

22. *L443-446 – The increased bias from JS-prior to JS-posterior at NJU should be discussed in more detail.*

Response and revisions:

We thank the reviewer's comment. The increased bias from JS-prior to JS-posterior in July and October at NJU and the detailed analysis was mentioned **in lines 508-516 in the revised manuscript**. It resulted mainly from the limitation of current multiple regression model that overestimation and underestimation in concentrations at different sites could hardly be corrected simultaneously without further improvement in spatial distribution of emissions.

23. *L464 – some cases are for other months.*

Response and revisions:

We thank the reviewer's comment. The sensitivities to observation and bottom-up emission input were evaluated in April (Cases 2-5). We evaluated the near linearity between emissions and concentrations in July and October as the two months were identified as the months with the most and least impact from precipitation suggested by simulated wet deposition to emission ratio. The impacts of simulated wet deposition and satellite-derived accumulated precipitation on top-down estimate were evaluated in July (Case 6-7). We had specified it **in lines 518-525 in the revised manuscript**.

24. *L551 – "initial" should be "a priori". Please revise throughout the text.*

Response and revisions:

We thank the reviewer's reminder and revised it throughout the text.

25. *L573-604 – the paragraph contains multiple messages, and is better to be splitted.*

Response and revisions:

We thank the reviewer's comment. As suggested, the smaller difference in BC emissions and simulated concentrations between JS-posterior and MEIC-posterior were split **in lines 639-666 in the revised manuscript**. The effect of the a priori bottom-up emission inventories on top-down estimate was summarized in another paragraph **in lines 667-671 in the revised manuscript**.

26. *Figs. S8-11 – the dates of precipitation are also not very well simulated.*

Response and revisions:

We thank the reviewer's comment and delete the evaluation of simulated precipitation dates **in lines 703-704 in the revised manuscript**. Considering the large discrepancy between simulated and observed precipitation, we conducted Case 7 to screen satellite-derived precipitation and compared the top-down estimates in two cases.

27. *L701 – "insignificant" should be "modest"*

Response and revisions:

We thank the reviewer's reminder and revised it **in line 763 in the revised manuscript**.

28. *L715-717 – the increased bias at NJU should be mentioned.*

Response and revisions:

We thank the reviewer's comment and mentioned the increased bias **in lines 779-780 in the revised manuscript**.

29. *L735-737 – it would be extremely difficult to use satellite AOD to constrain BC emissions.*

Response and revisions:

We thank the reviewer's comment and deleted the texts in the revised manuscript.

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