Response to Comments on the Manuscript (acp-2020-1004):

"Estimating daily full-coverage and high-accuracy 5-km ambient particulate matters across China: considering their precursors and chemical compositions"

Response to Comments of Fei Yao:

General comment:

This study presents an admirable, solid work of estimating PM concentrations using data sources other than conventional AOD products. The manuscript is clearly well-written and easy to follow. From my humble view, I am only concerned with the high-resolution use of the coarse-resolution GEOS-FP datasets (further explained below). If this can be properly justified, I believe it will be a very nice paper.

Response: We are particularly grateful for your approval of our research. An item-by-item response to the constructive comments follows. Thanks for your time.

Major comment:

Q1: The authors explained that it is reasonable to estimate PM concentrations using datasets of chemical precursors and species. But because the high-resolution TROPOMI only provides chemical precursors, the authors employed the coarse-resolution GEOS-FP for chemical species. However, using coarse-resolution datasets as MAJOR predictors (also confirmed by their relatively high ranks shown in Figure 6) for high-resolution PM mapping inevitably introduced uncertainties rather than more valuable information. This is simply because we typically do not have accurate, high-resolution emission inventories to drive a data assimilation system like GEOS-FP. The authors should justify this issue. Otherwise it would be possible to doubt the significance of this study. In other words, because GEOS-FP provides PM species data and thereby can provide total PM data at a coarse resolution via some sort of add-up, is it really

necessary to do a big load of modeling (correlating) work to derive PM concentrations with a plausible high resolution but associated inevitable uncertainties?

Response: Thank you for your significant comment. According to the previous works [1-2] of our team, the high-resolution geographical factors (e.g., land cover map) can help improve the spatial resolution of the estimated PM. In this study, several high-resolution geographical factors (i.e., land cover map, NDVI, road density, and population density) were exploited as the ancillary variates to maintain the spatial information. The space-based CV results show that the proposed framework performs well in the study area (e.g., R^2: 0.88 for $PM_{2.5}$ and 0.83 for PM_{10}), indicating that GEOS-FP data did not introduce large uncertainties. Meanwhile, the comparison for the spatial distribution (see an example in Figure 1) also signifies that the spatial resolution of the estimated PM is much higher than that of GEOS-FP data. Moreover, some relevant works that estimated ambient concentrations of air pollutants have employed coarse-resolution datasets as major predictors, such as [3-5]. In conclusion, the adoption of GEOS-FP datasets is justified in our study.

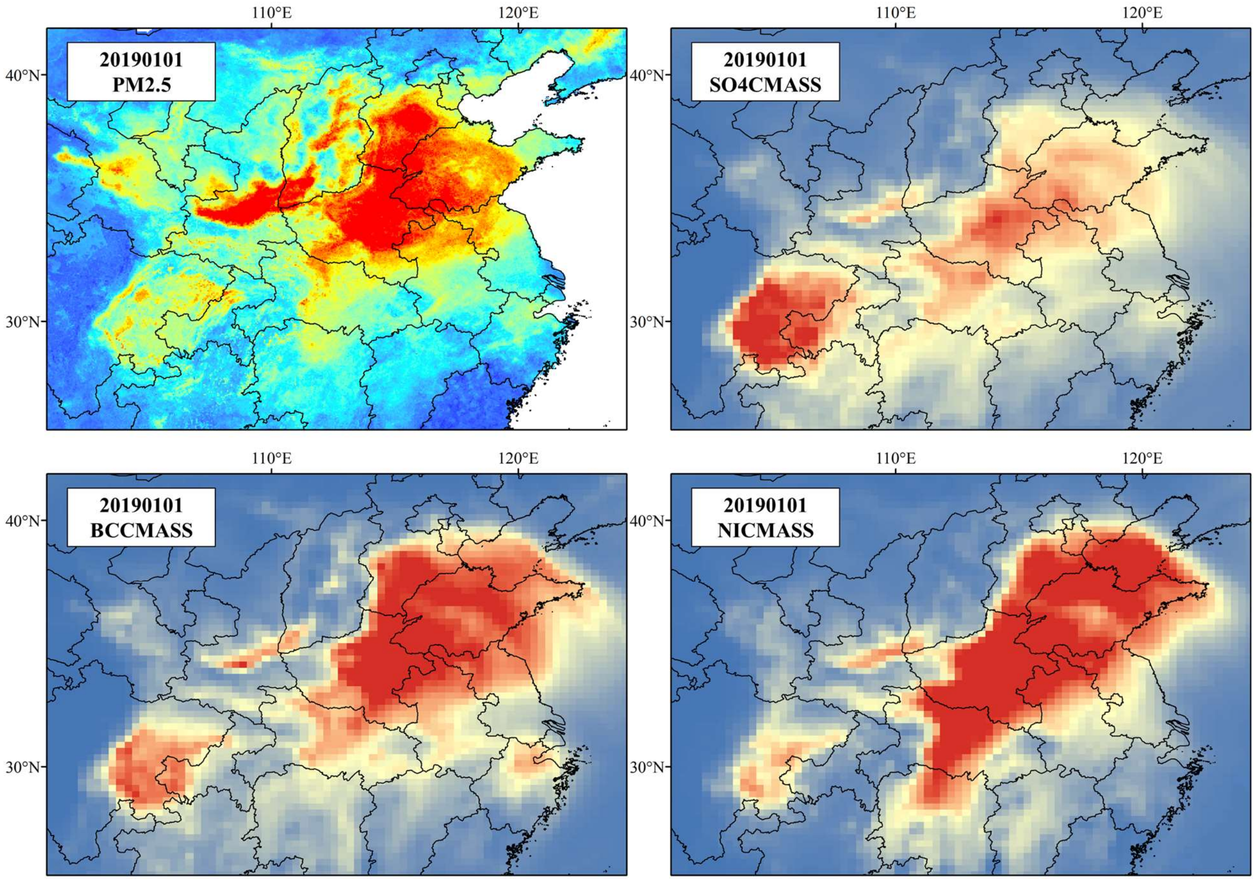

Figure 1: Daily (20190101) spatial distribution of the estimated $PM_{2.5}$ and GEOS-FP data (SO4CMASS, BCCMASS, and NICMASS)

References:

[1] Yang, Q., Yuan, Q., Li, T., & Yue, L. (2020). Mapping PM2. 5 concentration at high resolution using a cascade random forest based downscaling model: Evaluation and application. Journal of Cleaner Production, 277, 123887.
[2] Yang, Q., Yuan, Q., Yue, L., Li, T., Shen, H., & Zhang, L. (2020). Mapping PM2. 5 concentration at a sub-km level resolution: A dual-scale retrieval approach. ISPRS Journal of Photogrammetry and Remote Sensing, 165, 140-151.
[3] Zhan, Y., Luo, Y., Deng, X., Zhang, K., Zhang, M., Grieneisen, M. L., & Di, B. (2018). Satellite-based estimates of daily NO2 exposure in China using hybrid random forest and spatiotemporal kriging model. Environmental science & technology, 52(7), 4180-4189.
[4] Liu, D., Di, B., Luo, Y., Deng, X., Zhang, H., Yang, F., ... & Yu, Z. (2019). Estimating ground-level CO concentrations across China based on the national monitoring network and MOPITT: potentially overlooked CO hotspots in the Tibetan Plateau. Atmospheric Chemistry and Physics, 19(19), 12413-12430.
[5] Chen, Z. Y., Zhang, R., Zhang, T. H., Ou, C. Q., & Guo, Y. (2019). A kriging-calibrated machine learning method for estimating daily ground-level NO2 in mainland China. Science of The Total Environment, 690, 556-564.

Minor comments:

Q2: The GEOS-FP provides more than described variables for use. Why do you choose the column mass density variable rather than others, say surface concentration variables?

Response: Thank you for your comment. The estimation accuracy for the proposed framework using column mass density variables is slightly better than that using surface concentration variables. Hence, column mass density variables are adopted in our study.

Q3: Line 279-280, because you are modeling on the same dependent variable (i.e. PM concentrations), RMSE is comparable though you can choose not to describe for conciseness.

Response: Thank you for your comment. In our study, the numbers of matched samples are provided in Figure S9. Since missing values exist in the VIIRS DB AOD product, the data distributions of the estimated results through the proposed framework and AOD-based are different. For instance, the estimated results through the AOD-based is probably available for tens of days in a year, which cannot represent the annual condition. As for a matched grid, if the estimated results of $PM_{2.5}$ through the AOD-based is mainly valid in DJF/JJA, their RMSE could be relatively large/small. As a result, the comparison for RMSE is likely inappropriate and was not discussed in the manuscript.

Q4: Line 314, larger => smaller?

Response: Thank you for your comment. This statement has been revised in the manuscript.