

## ***Interactive comment on “Estimating daily full-coverage and high-accuracy 5-km ambient particulate matters across China: considering their precursors and chemical compositions” by Yuan Wang et al.***

**Anonymous Referee #2**

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### General comments

In this paper, Wang et al. proposed a framework to estimate the daily PM<sub>2.5</sub> and PM<sub>10</sub> concentration over China by combining multiple data sources with a Light Gradient Boosting Machine learning method. They included satellite product (TROPOMI) and modeled data assimilation dataset (GEOS-FP) as main predictors. Even though they showed some reasonable statistics from the validation process, the advantages of this method are not well justified. Also, some flaws are found in their validation process. As a result, I do not recommend this paper for publication in ACP.

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### Major comments:

1. The selection of predicting variables from TROPOMI is too arbitrary and lacks justification. In this paper, the authors only considered column NO<sub>x</sub> and SO<sub>2</sub> observations, which are the precursors of sulfate and nitrate. Both of them are large components of PM<sub>2.5</sub>. However, other components are also important. For example organic aerosols. Why the precursors of organic aerosols were not chosen as input predictors? Also, this idea of “PM is associated with ozone, so choose column ozone as one of the predictors” needs more justifications.
2. By using GEOS-FP, this method loses its flexibility to adjust its input conditions. GEOS-FP is one of the data assimilation products from GMAO. It is based largely on the model output of GEOS. The method in this paper is largely based on this dataset. According to their fig 6, 4 out of 6 top feature importance for their PM<sub>2.5</sub> prediction are from GEOS-FP. What this means is that their prediction is mostly controlled by a dataset that they could not make any modifications to. For example, the GEOS-FP system has been updated to version 5.25 in January 2020 (see this link [https://gmao.gsfc.nasa.gov/news/geos\\_system\\_news/2020/GEOS\\_FP\\_upgrade\\_5\\_25\\_1.ph](https://gmao.gsfc.nasa.gov/news/geos_system_news/2020/GEOS_FP_upgrade_5_25_1.ph)). Let alone some discontinuous issues posed by all the updates along with the release of each GEOS-FP products. On the other hand, the authors argue that previous studies using CTMs have the limitation of large uncertainty in the emission inventories of the CTMs. By saying that, GEOS-FP also has the same problem of large uncertainty in their emission inventories. By using CTMs instead of data assimilation datasets as prediction inputs, researchers can do their best to narrow the uncertainty in their model simulations and could also conduct sensitivity experiments. From this point of view, methods using CTMs or earth models (e.g. GEOS) would be better for this kind of prediction.
3. The validation process exists flaws. First, the authors defined their “AOD-based” control estimate, which is using VIIRS AOD to replace the TROPOMI and GEOS-FP in their estimating framework. However, this is not the case of previous AOD based

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studies, especially those most influential ones, for example, van Donkelaar et al. 2019. Previous AOD based studies usually combined with model simulations and ground-based measurements to best use the information of satellite AOD products. What has been done in the paper was that comparing an estimate from a missing data AOD product with an estimate from the combination of satellite observations and model output. So that it caused the issue of comparing apple with orange; moreover, the selling point of this paper is the daily PM<sub>2.5</sub> estimate. So validation regarded to a daily resolution would be the most convincing. Otherwise, why not just estimate the seasonal or annual PM concentrations, which are more useful in current epidemiological studies. The validation results from the time-based cross-validation method were the worst according to their table 1.

#### Specific comments

1. Line 99: is it necessary to mention that the area of this study was chosen because that China has the largest population in the world? Maybe rephrase this sentence or delete it.
2. Line 106: "monitor"? maybe use "estimate"
3. Line 120: any publications about the data validation, calibration, and uncertainty analysis of the Chinese PM<sub>2.5</sub>/PM<sub>10</sub> measurements from CNEMC?
4. Follow point 3, the authors could try to use this method on other regions where have an extensive ground-based measurements network, for example, North America, to test their validity.

#### Technical corrections

1. Section index is wrong. For example, two "3.2" sections exist in the main text.
2. Line 40: should be "van Donkelaar" instead of "Van Donkelaar"
3. Define "DUCMASS" in the main text.

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#### References

Van Donkelaar, A, Randall V. Martin, Chi Li, and Richard T. Burnett. "Regional Estimates of Chemical Composition of Fine Particulate Matter Using a Combined Geoscience-Statistical Method with Information from Satellites, Models, and Monitors." *Environmental Science & Technology* 2019. <https://doi.org/10.1021/acs.est.8b06392>.

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