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Comment on acp-2021-683

Anonymous Referee #4

Referee comment on "Estimation of secondary $PM_{2.5}$ in China and the United States using a multi-tracer approach" by Haoran Zhang et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-683-RC3, 2022

The manuscript presents a method for estimating the relative contributions of primary and secondary PM by proxy. The observed input parameters are CO, PM10 and PM2.5, however, the metohod also relies on estimated emissions of OA, EC, OC, fine dust, PM2.5, sulfate and nitrate, from emission inventories. The authors develop a proxy for secondary particulate matter on the basis of the observed parameters and estimated emissions.

The motivation is presented as the need for a low cost, operational method for monitoring the contributions of secondary aerosols to the total PM2.5 levels.

The method appears to have some use for informing operational air quality management or for informing policy, but the scientific value of the method is not convincingly presented. It relies on assumptions and inventories that are not universal, and the manuscript does not present a convicning argument for its use, other than that it is cheaper than source apportionment methods based on chemical speciation. But it does not present comparative estimates of primary-secondary contributions with those methods.

It is questionable if this method has any value. It requires a big body of inputs, as other chemical transport models, but also relies heavily on assumptions and coefficients that are externally adjusted, even tuned to fit the model.

The manuscript describes comparisons between estimated and observed primary particulate matter. Categorisation of measured historical data into secondary and primary aerosols for comparison with the MTEA seems ot be based on chemical compositions, but this process is not clearly described and the criteria are vague.

There has been no attempt to verify the MTEA estimates for ppm by comparing with published estimates based on receptoro modelling, CTMs or AMS studies. There are many studies in the literature that have produced estimates that can be easily compared with the outcomes of the MTEA approach, but that has not been done.

It is true as the authors state that those other methods are labour-intensive and expensive, but they are also scientifically tried and tested and therefore more convincing, so it would make sense to develop the perfomrance of the MTEA against such methods more than has been done in this manuscript.

Also, the manuscript states that the numerical calculations were done on a supercomuting system. It can be argued that if the approach requires a supercomuting facility then it is no less costly or inaccessible than the existing source apportionment methods, but the cost has been shifted from scientific equipment to IT services.

The manuscript does touch on a discussion that has scientific interest, and that is contained in the sections 4.1 and 4.2 on spatial and temporal variation. The discussion on spatial variation has some merit. There is potentially a better motivation for developing the MTEA approach in order to inform a discussionon the spatial and temporal variation where only proxy parameters are available, by leveraging national monitoring networks to learn more about geographical distribution of secpondary aerosols and feed into a discussion on variations in atmospheric processes.