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Comment on acp-2021-683

Anonymous Referee #3

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-RC1, 2021

- This study developed a new method to determine the portion of primary and secondary PM2.5 using some basic measurements and inventory. They evaluated this new approach through the comparison with lots of observations in China and US. In addition, they analyzed the temporal and spatial variation as well as correlation between O3 and PM2.5 using the results from their new method. Although their evaluation looks very well, I think their results were not enough convincing because of unclear statement of their method and defect of this method. I would suggest major revision before reconsideration. My detail comments are following.
- Eq(1) and Eq(2): These equations are the core of their method. They regarded CO as one tracer to represent the combustion process and assumed the combustion emission sources are same for CO, OC and EC. This assumption is mostly correct, but the emission factor/emission ratio of CO, OC and EC from different combustion sources are different. I think it is unconvincing to use one single coefficient without the influence of diversity of sources to standard for all conditions. I may misunderstand something, please discuss this uncertainty or make this clear.
- Eq(2): why did you name b as emission of fine dust? To my knowledge, MEIC does not include the emission of dust even urban dust.
- I did not understand how you did the sensitivity experiment to examine the uncertainty in the inventories. Page 16, you said you changed the emission coefficient with 10%. If so, how can you keep a+b=100%? According to my understanding on this new method, the results should have large dependence on the inventory of PM2.5, OC, EC even the factor you used to decide OA, SO4 and NO3. I would strongly suggest setting up more comprehensive and scientific sensitivity experiments to discuss the dependence on the inventory.
- Figure 4, as I saw, the largest concentration is < 60 μg/m3. Why not short the range of axis to spread those dots?
- P8L7: Why did you remove the heavy pollution cases here as well as in Section 4? As you stated at P10L25, you would like to avoid the influence of extreme high primary emission cases. However, mostly heavy pollution cases are caused by unfavored meteorological condition but not caused by sudden high primary emission (except the biomass burning cases). I would be curious that how your method applied to analyze the heavy pollution cases. In general, it is more important to understand the contribution of secondary particles to heavy pollution cases than the general conditions.
- P10L30: Could you explain what is regional background cities you defined here? Usually, cities are not background.
- Section 4.2.1: I think the seasonal variation of PPM and SPM is largely depend on the

seasonal variation of emissions you applied.

- Section 4.2.2: Did you use the emission inventory for specific year here? China conducted a large reduction on PM2.5 emission since 2014. If you did not use the specific inventory, the estimated trend of PPM and SPM would not make sense, even though they agreed with observations. In addition, could you show the correlation coefficient between the observation and estimation here?
- Section 4.3: The same issue as above. Did you update the inventory to the lockdown condition? If yes, please state the inventory you used here and the decrease in the emission of PM2.5, CO, OC, EC.
- Section 4.4: How did you decide the diurnal variation of emission? Was your result sensitive to the diurnal pattern? Because the diurnal pattern of O3 concentration is almost constant.
- Section 4.4: Why did you exclude the wet deposition case here but include in other sections? I would suggest adding the application condition for your method somewhere.
- The general method to calculate the portion of secondary PM2.5 is chemical transport model using bottom-up inventory. It's better to examine the difference in the result between your method and CTM with same inventory.