Atmospheric Chemistry and Physics Discussions

Atmos. Chem. Phys. Discuss., referee comment RC2
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Comment on acp-2021-77

Anonymous Referee #2

Referee comment on "Source apportionment of atmospheric PM_{10} oxidative potential: synthesis of 15 year-round urban datasets in France" by Samuël Weber et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-77-RC2, 2021

In this manuscript, the authors presented source apportionment results of OP-DTT and OP-AA measured on PM_{10} filter samples collected at 14 different locations in France using PMF and multiple linear regression models. The authors mainly focus on discussing the intrinsic OP, the variability of different sources, and the daily mean and median contribution of sources to OP. The limitation of the study is well discussed. This study has a unique dataset. However, in my opinion, the authors did not fully utilize their dataset. One of the uniqueness of this study is that the dataset spans 15 years and covers a wide range of environments. Seasonal variations of OP-DTT and OP-AA are discussed but the authors may consider looking into other aspects that are more interesting, for example, the spatial homogeneity or the historical changes of OP. How do the OP sources and source contributions change over the 15-year period? Are there differences in the historical trends of OP-DTT and OP-AA and how does it compare to PM mass? What chemical components are the most important drivers to the changes of OP and PM mass? It is well known that biomass burning, traffic emissions, secondary processing are important OP sources. Presenting something other than sources would enhance the scientific significance of this manuscript. Below are my comments:

Major comments:

- Please provide detailed protocols for both the DTT and AA assays. Multiple versions of DTT assays are currently used in the community. It is important to show which DTT protocol was used in this work. Perhaps the most important is the initial concentration of DTT as studies have shown that the initial DTT concentrations can have a large impact on the DTT consumption rates e.g. (Lin and Yu 2019).
- Line 110, particles removed from the filters were added to 96-well plates for DTT analyses, and the authors claimed that this included “soluble and insoluble” particles. What is the extraction efficiency? Could there be particles that are not extractable and attached to the filters. Other studies that measured total DTT run the extract along with the filter in DTT solutions. E.g. (Gao, Fang et al. 2017). A note should be added to emphasize the differences in the protocol and state that the DTT activities may not be “total”.
- In Figure 3, OP-AA from road traffic, biomass burning, dust and OP-DTT from biomass

burning and dust sources are bi-model distributions. Why? It would be interesting to look into details in chemical components to figure out the observed distributions.

- Section 3.3.6-3.3.11, I appreciate the authors' efforts in discussing the variability of the intrinsic OP. However, it is not clear to me what do variabilities of different OP sources really bring about. It seems more interesting to compare the intrinsic OP or the contribution of different sources to OP with those from other studies or those from other regions of the world. These subsections lack in-depth discussion on each source. For example, for road traffic, transition metals (non-exhaust) and quinones from PAHs or soot (exhaust) can contribute to OP-DTT and OP-AA. How does your source profile from road traffic differ from others? What are the linkages of traffic-related chemical components to measured OP? What new insights does this work bring? One interesting questions is whether you can differentiate the contribution of exhaust and non-exhaust emissions to OP.
- It would be useful to present how are metals (especially Fe and Cu) apportioned into each factor. Atmospheric metals can be found in biomass burning, road traffic, dust, or sulfate particles by acid processing. Discussions on how metals are distributed in these factors may help to interpret the contribution of different sources to OP.
- It is not clear that what are the new findings in this manuscript compared to previous work from the same group, e.g. (Weber, Uzu et al. 2018).

Other comments:

- Line 257, "organic specie" should be "organic species"
- Line 258, define "HULIS"
- Use the same font type throughout, for example, line 464, numbers seem to be a different font than other contents

References:

Gao, D., T. Fang, V. Verma, L. Zeng and R. Weber (2017). "A method for measuring total aerosol oxidative potential (OP) with the dithiothreitol (DTT) assay and comparisons between an urban and roadside site of water-soluble and total OP." Atmos. Meas. Tech. Discuss. **2017**: 1-25.

Lin, M. and J. Z. Yu (2019). "Dithiothreitol (DTT) concentration effect and its implications on the applicability of DTT assay to evaluate the oxidative potential of atmospheric aerosol samples." Environmental Pollution **251**: 938-944.

Weber, S., G. Uzu, A. Calas, F. Chevrier, J. L. Besombes, A. Charron, D. Salameh, I. JeÅ¾ek, G. MoÄ□nik and J. L. Jaffrezo (2018). "An apportionment method for the oxidative potential of atmospheric particulate matter sources: application to a one-year study in Chamonix, France." Atmos. Chem. Phys. **18**(13): 9617-9629.