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Comment on acp-2021-77

Anonymous Referee #1

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-RC1, 2021

This manuscript is focused on the source apportionment of the oxidative potential (OP) of atmospheric PM_{10} from a long-term large-scale sampling campaign in France. More than 1700 samples from 14 sites and 15 years sampling period were analyzed for OP in two endpoints (OP^{AA} and OP^{DTT}) and various chemical species. The authors implemented a positive matrix factorization (PMF) coupled with multiple linear regression (MLR) protocol for identifying the sources from individual sites contributed to measured OP. The importance of different sources was discussed based on both intrinsic OP (OPm) and their contributions to the total OPv. The authors also attempted to explain the seasonality of certain sources in the context of OP. Overall, the manuscript presented very interesting and significant results of PM_{10} OP in West Europe area, and strongly supported the health effect study and legislation of air quality control in France. However, it lacks a discussion on OP's spatial distribution and the linkages of chemical composition to OP. Following are my specific comments.

Specific comments:

- Generally, fine particles (PM_5) have been linked to the adverse effect caused to human respiratory and cardiovascular system, since these particles have highest penetration efficiency in lower respiratory tract. However, the PM samples collected in this manuscript have involved larger sized particles which have shown lower oxidative potential (Hu et al., 2008;Ntziachristos et al., 2007) and somewhat lesser relevance to human health. Thus, a justification should be provided for using PM_{10} in this health-related study, i.e. source apportionment of PM OP.
- Lines 85 – 89: Please provide more details on the geographical information of all sites, e.g. distance to local highways, industries and/or coast, wind direction and relative location in the valley/peak of Alpine area etc. This can help to directly identify some potential sources.
- Line 119: Why 1,4-naphthoquinone was chosen as the positive control for AA? Does it have a consistent and high AA activity?
- The apportionment of SOA factors in this paper is ambiguous. The authors did not explain the contributions of OC in three highlighted SOA-related factors, i.e. nitrate-rich SIA, sulfate-rich SIA and MSA-rich. We suggest a better explanation should be provided

on their formation and the differences of their contributions to OP and mass of PM_5.

- The subsections in Section 3.3 are confusing. I suggest combining Sections 3.3.2 – 3.3.5 to a single subsection "Intrinsic OP of main PMF sources", and create a new subsection for Sections 3.3.6 – 3.3.12 (like Section 3.4), "Profiles of OPm sources".
- Lines 367 – 370: The authors tend to tone down the intrinsic OP of MSA-rich factor. In fact, I found very high $OP^{DTT}m$ for this factor at GRE-cb, NGT and STG-cle with reasonable CV (<0.6), while $OP^{AA}m$ at these sites were near 0. I would suggest the authors to further explore the redox-active components and explore the reasons for different activities between two endpoints for this factor at these sites.
- Lines 391 – 395: Please specify the factors with low variability of OPm and explain why the other factors show large variability.
- Lines 410 – 415: the authors should highlight the high contribution of sulfate-rich SIA factor to OP^{DTT} In comparison to its marginal contribution to $OP^{AA}v$, it is also important to find out the ROS-active components in this factor for the difference between two OP endpoints.
- Lines 421 – 425: The explanations on the mean and median values of contribution of sources are unclear. The authors should explain the meaning of these two values and justify why using them under different circumstances.
- Lines 473 – 476: I suggest the authors further investigate the sensitive chemicals (like transition metals) to both OP endpoints at all the sites with the industrial factor, in order to explain the huge difference of OPm found among all six sites.
- Line 507: A considerable contribution from sulfate-rich factor has been noted in Figure 4 – 6. Hence, the author made a wrong statement here saying both SIA sources contribute barely to OP which should be corrected.
- Line 512: The paper showed many sources with different spatial variability in intrinsic OP, which is not discussed. Therefore, caution should be exercised when making statements like "relatively stable intrinsic OP at a large geographical scale". The authors should discuss it in two types of sources – sources with low variability and stable intrinsic OP (e.g. road traffic), and sources with high variability and varied intrinsic OP (e.g. biomass burning).

Technical corrections:

- Line 14: to prioritized – insert "be" between these two words.
- Line 76: "multilinear" should be changed to "multiple linear" or "multivariate linear".
- Line 115: the abbreviation in the parenthesis should be "RTLF".
- Line 174, 237, 257: "specie" should be changed to "species".
- Line 266, 459: The word "inversion" in the title is confusing. I suggest replacing "OP's inversion" with "intrinsic OP".
- Line 431: I suggest moving "in Figure 5" to the end of the sentence or include it in parenthesis.
- Legend of Figures 5 and 6: the error bars __ the 95% confidence level – insert "represent" at the underline.
- Legend of Figure A2: "Searman" should be changed to "Spearman".

References

Hu, S., Polidori, A., Arhami, M., Shafer, M., Schauer, J., Cho, A., and Sioutas, C.: Redox activity and chemical speciation of size fractioned PM in the communities of the Los Angeles-Long Beach harbor, Atmospheric Chemistry and Physics, 8, 6439-6451, 10.5194/acp-8-6439-2008, 2008.

Ntziachristos, L., Froines, J. R., Cho, A. K., and Sioutas, C.: Relationship between redox activity and chemical speciation of size-fractionated particulate matter, Particle and fibre toxicology, 4, 5, 2007.