Atmospheric Chemistry and Physics Discussions

Atmos. Chem. Phys. Discuss., referee comment RC2
https://doi.org/10.5194/acp-2021-979-RC2, 2022

Comment on acp-2021-979

Anonymous Referee #2

Referee comment on "Linking Switzerland's PM_{10} and $PM_{2.5}$ oxidative potential (OP) with emission sources" by Stuart K. Grange et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-979-RC2, 2022

Grange et al. investigated the oxidative potential (OP) of ambient PM_{10} and $PM_{2.5}$ at five sites located in different environments of Switzerland. OP was assessed using three different endpoints - ascorbic acid (AA) and dithiothreitol (DTT) consumption, and dichlorofluorescein (DCFH) assay. They explored the spatiotemporal variability of OP and compared the OP levels with those measured in France. The source analysis followed by the investigation of sensitive components were then conducted among measured PM species, mass, and OP, and the finding suggests a higher level of OPm associated with non-exhaust traffic emissions and wood burning.

Overall, this paper provides a thorough analysis of OP levels in Switzerland and its comparison with France. The selection of sites covered common types of geographical locations with high population, and the comparison of OP with those in France provided a wider spatial scope of the health effects of PM_{10} in Europe. The results of sources with high OPm potency identified in this paper were generally consistent with previous studies in different regions, and the results pointed towards the importance of controlling local traffic emissions and woodburning. However, the protocol of sensitive chemical identification seemed to be flawed. By filtering the species with multicollinearity, some important contributing species might be also filtered. The regressions with significant R^2 did not present the important contributors, which might indicate the lack of scientific implications of the results from this method. Furthermore, the species that were found to be highly correlated with OP in the multiple linear regression were not critical and could be easily replaced by other species, further pointed out the weakness of this method. Therefore, I would like to recommend that the paper should be majorly revised majorly for further consideration.

Specific comments are listed below.

Major comments:

- The materials and methods section written in the paper were too concise. The characteristics of five sampling sites, the list of measured chemical species on filters, selection criteria applied in random forest, and the factors used in PMF are not provided, and should be further described to provide a full understanding of all the protocols.
- The level of PM used in this study (25 μg/L, i.e. 0.025 μg/mL) is far lower than that applied in most other studies (10-50 μg/mL). I highly doubt that this level may generate valid OP results. Please check the unit of the concentrations.
- While explaining that AA is a major constituent of lung lining fluid, the paper used an AA-only model for monitoring the consumption of AA, but many studies used the endpoint of AA in surrogate lung lining fluid to better represent the biological environment in human. Please provide a justification of using the AA-only model.
- Figure 2 combining all three endpoints in the same box seems to be confusing. Since the comparison among these endpoints is not reasonable, I would suggest splitting them into different boxes as per different endpoints, for presenting the data.
- Grange et al. 2021 is not available online and it seems to contain a lot of information for the interpretation of this paper.
- In lines 218-221, the authors explained the trend of OPcoarse, but this term is not well defined or calculated anywhere in the entire manuscript. Therefore, I suggest to provide further description, trend and calculation of this term.
- The discussion of PMF results lacks depth. Even if most of the results might have been provided in Grange et al. (2021) which is not accessible now, the discussion for the MLR analysis between OP and PM sources should be enhanced. The contribution of sources towards different OP endpoints in OPv should be involved in this section.
- The method of random forest is very ambiguous: the selection of importance based on ranking should be provided in the paper. Also, in Figure 6, the justifying criteria "ranking highly" should be quantified.
- The results showing interchangeable species for the significant correlations between PM OP and concentration of components is concerning: the actual contributing species might be omitted during the selection and the final results could only find out the indicators towards important sources. Although some key contributing species like Cu and Mn were identified, they were eclipsed in the numerous correlation pairs of non-contributing sources and OP. This is further demonstrated by Figure 7: the pairs of species involving most significant correlations (Sb and galactosan) were not contributing to PM OP. Therefore, the causality is not indicated by this method. This should be included in the discussion of the limitations.
- Including PM mass in the regression might not be a good idea: PM mass might have a much higher weight than the chemical species included in the model. Therefore, the results might be biased, since OPv is determined by PM2.5 mass to some extent. Therefore, I would suggest removing PM mass for the MLR analysis between OP and species.

Minor comments:

- The introduction section should be supported by more literature. For example, in line 66-67, the statement of the different spatiotemporal trends of OP and PM mass could be supported by Yang et al. (2015) (DOI: 10.1016/j.atmosenv.2014.11.053), Liu et al. (2018) (DOI: 10.1016/j.envpol.2018.01.116) and Yu et al. (2021) (DOI: 10.5194/acp-21-16363-2021).
- Line 37: remove "say," .
- Please provide further details of DCFH assay, including the cells used for this assay and assay protocols.
- Line 148: Please provide the supporting citations for the statement "DCFH assay is sensitive to organic compounds".
- Line 158: Provide the full name of "SOURCES". Also, this sentence is highly confusing: is PMF informatively known as extended PMF, or is SOURCES program informatively known as extended PMF? Think restructuring this sentence.
- Line 220: Saying that OPcoarse is biological relevant is not rigorous since the biological relevance should not only consider the level of OP but also include the accessibility of these coarse particles in the respiratory tract. Suggest revising this statement.
- Line 237: The comparison did not involve OP^{DCFH} This information should be listed.
- Line 252: This sentence should be moved to the discussion of OP^{AA} in the previous paragraph.
- Line 386: Provide the comparison of numbers of pairwise combinations between PM_5 and PM_{10}.