

Atmos. Chem. Phys. Discuss., referee comment RC1
<https://doi.org/10.5194/acp-2021-57-RC1>, 2021
© Author(s) 2021. This work is distributed under
the Creative Commons Attribution 4.0 License.

Comment on acp-2021-57

Anonymous Referee #1

Referee comment on "Disparities in particulate matter (PM₁₀) origins and oxidative potential at a city scale (Grenoble, France) – Part 2: Sources of PM₁₀ oxidative potential using multiple linear regression analysis and the predictive applicability of multilayer perceptron neural network analysis" by Lucille Joanna S. Borlaza et al., Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2021-57-RC1>, 2021

The manuscript reports the second part of the results of the chemical analyses of a yearly set of PM₁₀ samples collected at three urban sites in Grenoble (FR). The assessment of the main source contributions performed by positive matrix factorization (PMF) is discussed in the first paper (Borlaza et al., 2020) while here the focus is on oxidative potential (OP) and its relation to the PMF factors identified in the first study. This is carried out using simple multiple linear regression (MLR) analysis as well as by an artificial neural network (ANN) approach: the multilayer perceptron analysis (MLP). This is probably among the first applications of machine learning techniques to the investigation of the chemical characteristics of PM determining its OP. The MLP analysis can account for possible non-linear behaviours of OP due to synergic or antagonistic effects between reactive PM chemical compounds, hence providing a more realistic representation of the way OP is determined by aerosol compounds present in mixtures. I list my major comments below:

The MLR method has a clear advantage on MLP: it enables to assess OP source contributions explicitly (Fig. 5). Therefore, it is mainly by means of MLR that this study addresses the main policy-relevant questions on the sources of aerosol toxicity in this environment. Ideally, the individual sources would carry constant specific intrinsic OP and the different OP_v levels between sites would be explained entirely by the spatio-temporal variability of the sources. This is only partly achieved, because undefined "site-specific features" (line 358) remain. Most notably, the specific intrinsic OP of the industrial factor is inconsistent between the PU site and the UB+UH sites for two out of three OP assays. It is therefore unclear on what basis, OP source contributions can be generalized and averaged between the three sites (Fig. 6) to provide a ranking of them. The impression is that the PMF analysis was unable to capture the full sources of OP variability in this environment.

The MLP analysis represents the most innovative aspect of the methodology. However, the results show that the performance of MLP is not really superior to that of MLR in reproducing the observed OP, except for the AA assay at the UH site. The true highlight about MLP stands in its ability to detect non-linear behaviours between chemical

compounds. However, such behaviours are not characterized explicitly by the ANN analysis, they remain "hidden" and can only be diagnosed (Fig. 8). The way this is carried in this study (Section 2.4.3.3) is not completely convincing. According to this method, the OP for a given source is estimated as the difference between the modelled OP and the modelled OP obtained on a dummy dataset where the PMF factor for that same source is omitted. However, the PMF factors are not orthogonal, they often exhibit a certain degree of covariance. Even if we remove the nitrate-rich factor, for instance, some features of its time series are still present in the trend of the biomass burning factor. The variability of a given source cannot fully be omitted in the dummy dataset. As a consequence, the source-contribution of OP calculated as a residual (equation 3) risks to be underestimated.

Specific comments:

Line 29 (Abstract): "underlining the importance of PM redox activity over mass concentration". This statement is unclear: is this a claim for PM redox activity being a superior metric respect to PM mass concentrations? I do not think the Authors would dare to say that. I suggest to rephrase into something like: "underlining the importance of PM redox activity for the identification of potential sources of PM toxicity".

Lines 38-39: "explore both the small- and large-scale variabilities of PM pollution accounting for local variations in different urban environments". Please, rephrase more clearly. What are the actual spatial scales at issue? What are the "different urban environments"? cities with different characteristics or different economic districts within a single city?

Lines 49 – 50: The definition of oxidative potential (OP) introduced by the Authors suggests that OP can really traces the ability of aerosol particles to induce oxidative stress in biological systems. However, the actual link between the OP determined by acellular assays and the ROS assays employing in vitro system is still currently matter of debate between scientists (e.g., J Øvrevik, International journal of molecular sciences 20 (19), 4772). The Authors are encouraged to provide a concise treatment of this fundamental issue.

Lines 63 – 64 ("because numerous factors could affect OP"): make such factors explicit.

Line 79 ("fine-scale spatiotemporal characteristics"): again, what are the scales of importance for the present study? If the city scale is the target, 24h-integrated samples collected at three sampling points is not properly "fine scale". Clearly, there should be more emphasis on the chemical resolution. Please, explain.

Line 88 ("catch the non-linear pattern of OP"): why non-linear behaviours of OP in this specific environment are taken for granted?

Line 133. The term "exposure" can be misleading in this context. Actually, the volume-normalized OP activity can be related to exposure only upon an assessment of outdoor exposure itself, which certainly is season-dependent. I would more safely define OPv as the OP carried by the aerosol expressed in OP units per cubic meter of air.

Figure 7: are source-contributions to OPv calculated using the MLR method? Please specify in the caption.