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Comment on acp-2021-839

Anonymous Referee #2

Referee comment on "Nine-year trends of PM_{10} sources and oxidative potential in a rural background site in France" by Lucille Joanna Borlaza et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-839-RC3, 2021

The authors present 9 years trends of PM_{10} sources and oxidative potential in a rural background site in France. Their analyses include PMF source apportionment and estimation of OP contributions via MLR and trends analysis. Major results show that traffic-related concentrations have decreased throughout the last decade and that Long-Range Transport processes contribute to a large fraction of the total PM_{10} mass, while OP is mostly dominated by traffic, mineral dust and biomass burning. Overall, the article is well-written, the analysis presented is fairly simple but clean, the methodology is (for the most part) sound, and the interpretation and discussion of the results with previous literature is solid. However, I do have some concerns related to the novelty and the scientific questions addressed in this work, as I detail below. Some more specific and technical comments follow after.

General comments to the authors

1) Many of the results related to the PM_{10} analysis are neither surprising nor original, i.e. traffic-related concentrations are decreasing (even in rural areas), SIA are large contributors to the total mass of PM_{10} in rural areas. What did we learn from your analysis that has general implications for atmospheric science, and was previously not known? Citing from the scope of ACP, *"The journal scope is focused on studies with general implications for atmospheric science rather than investigations that are primarily of local or technical interest.".* In other words, I feel that a clear scientific hypothesis/research question is lacking in this manuscript, and perhaps the authors should elaborate more on what they are trying to uncover. The process of a scientific manuscript in a high-quality scientific journal (as ACP) should start with a clear question/hypothesis that authors should try to answer given some new observational data/new modeling experiments/new theoretical insights. Reading the manuscript, I felt that the focus of the paper is on the dataset itself, rather than using the dataset as a tool to answer a scientific question.

2) As an example of this, the results from Pandolfi et al. (2016) also suggest that traffic concentrations in a rural background site (in Spain) have decreased in the last few years and secondary inorganics and organics are large contributors to PM_{10} in rural sites. For the

PM_{10} results, what did we learn from your analysis that adds significant new knowledge with respect to Pandolfi et al. (2016) work (to cite one, but there are other similar works available in the literature)? This should exceed the simple time span differences considered in their work.

3) With that said, I do acknowledge that there is some merit in analyzing OP contributions and show the differences with PM_{10} contributions (Sections 3.6-3.8), as OP is emerging as a promising endpoint-related metric to measure health impacts. However, in the introduction, you state that '*The characterization of PM sources and OP in a rural site will enable us to see the large-scale effects of mitigation policies that target reduction of PM mass concentrations. This will also provide knowledge of efficiency of current air quality guidelines in terms of other emerging health-based metrics of PM exposure.'* (Lines 51-54). A similar statement is repeated at the end of the introduction (lines 58-60). What knowledge of efficiency of current air quality guidelines for OP did you find? This is not clearly reflected neither in the results nor in the conclusions, as the trends analysis for OP is not really revealing much given the relatively short (4 years) period available. Once again, I believe you should try to highlight the value of your analysis as it relates to a specific hypothesis/questions, rather than propose very general statements that confuse the reader.

Specific comments

1) In section 2.3, the difference between the two assays can be elaborated further. Are the large differences presented in Figure 9b and Figure 9c expected? Are the contributions to total OP completely different because the two assays are meant to look at different oxidative processes in the lungs? This can be discussed in more details either in the methodology (highlighting more clearly why the two assays are used) or in the results, when commenting on the differences between Figures 9b and 9c.

2) I have some concerns about the MLR method presented in Section 2.5. One of the key assumption of linear regression is that the residuals ε are iid, but you're using the MLR to model timeseries data, where the assumption is evidently violated (by definition the OP data are not independent, as there is a clear temporal dependence). Perhaps you should consider adding a temporal component to your model (e.g. ARMA) that takes care of the temporal dependence to avoid misinterpreting the results on the β coefficients. Adding a temporal component is somewhat equivalent to detrend the data and remove seasonality, as you seem to be doing for PM10 analyses but not for the MLR part.

3) In the same section, I am not sure if I am interpreting Equation 1 correctly. Why is there a subscript to your and β matrices? In previous sections, referred to the total number of observations whereas the meaning of here is unclear. Shouldn't your model simply be:

$$OP = G\beta + \varepsilon$$

With my comment above, the model should be extended to something like:

$$OP(t) = \alpha OP(t-1) + G\beta + \varepsilon$$

To take care of the temporal dependence. Obviously the choice of AR(1) is very simple but different AR or ARMA models can be investigated. Also if G is a matrix, what does it mean the subscript ? I believe you should double check your notation to be consistent. If you decide to use matrix notation, you should be consistent throughout.

4) At the end of the introduction (line 59), you mention *long-term trends of emission sources*. Please be sure to use the right words here, I do not believe you are discussing emission trends at all, but only the decomposition of PM10 *concentrations* in different PMF factors.

5) I wonder if an effort could be made to actually show these emission data (aggregated at some level, for instance for the traffic category in a certain region), and see if there is some sort of correlation with the concentrations of the traffic factor that you showed from your analysis). A recurring theme of your manuscript is about analyzing the effect of recent changes in the source emissions (e.g. line 38-39), but I actually have not seen emission data at all. Can you make an effort to better substantiate that the reduced concentrations from the traffic sector (Figure 6) are actually related to emissions rather than, say, changing meteorology?

6) In Figure 2, it might be helpful to add error bars showing one or two standard deviations of your annual data. This would help interpreting how 'significant' (at least on a visual level) the differences between different years are.

7) Section 2.4.4 is quite unclear (or at least it's unclear until reading the results related to that section). I believe rephrasing the last couple of lines (166-170) and better defining what is implied by 'homogeneous' and 'heterogeneous' will help the reader in the interpretation of the associated results.

Technical comments

In the abstract the acronym OPE is not defined. Consider using a more general phrase (e.g. rural site).

Line 43: In-depth

Line 93: Define MSA

Line 226: Remove 100 out of 100

Line 475: Fix reference to Figure

References

Pandolfi, M., A. Alastuey, N. Pérez, C. Reche, I. Castro, V. Shatalov, and X. Querol, 2016: Trends analysis of PM source contributions and chemical tracers in NE Spain during 2004–2014: a multi-exponential approach. *Atmospheric Chemistry and Physics*, **16,** 11787-11805.