

Authors have answered/addressed many of my comments; however, a few things should have been added to the revised MS for the better clarity for readers. I suggest addressing the following comments made on author's responses (**text in bold green**).

My comment: Lines 127-129: Insoluble particles can be a large source of uncertainty, as they are not uniformly mixed in the solution. They can interfere with spectrometric analysis via physical absorbance.

Reply: The extraction procedure in this study is based on Calas et al. (2018), also published by our group. This procedure has been tested on both soluble and insoluble compounds that are (as much as possible) within the range of atmospheric concentrations. To avoid the interferences in the wells by insoluble particles, we subtracted the intrinsic absorbance of all PM extractions before adding reactants. Also, the particles are extracted in the Gamble solution (an artificial lining fluid) where we add a surfactant: this was shown to maintain a good dispersion of particles, leading to homogeneous results (see Calas et al., 2018). This is summarized in Table S5 of Calas et al. (2018). All analysis was performed in triplicate, with a coefficient of variation (CV) $\leq 5\%$.

Information about avoiding interferences from insoluble particles is not given in the methodology section. Brief methodology shall be clearly given in this MS, and for details it is fine to give the reference. Please include.

My comment: Lines 134-135: This suggests the precision of the measurements. How do you ascertain the accuracy of the measurements for each assay?

Reply: In every experiment, a positive control 1,4 naphthoquinone and an ambient filter (PM sampled from the lab roof) were analysed to ensure accuracy of measurements. All analysis was also performed in triplicate, with a coefficient of variation (CV) $\leq 5\%$.

This answer is not clear. Was 1,4 naphthoquinone used for all the three assays? I don't think all the three assays respond to this chemical. Please explain. How the ambient PM filter was used for the accuracy of all the three assays? It can only be used for the consistency (precision), which is already shown by CV of each analysis. Authors should provide the output of this accuracy experiment for each assay in the main MS. This information will also be very useful for readers.

My comment: Lines 281-283: This is very important point of the paper but not clear at all. Mass-normalised assays obviously depend upon the PM composition and not the PM mass. Different assays respond to different species. The statement written in lines 281-283 is confusing. Please elaborate this sentence in detail.

Reply: The comparison of the two measures (OP_m and OP_v) allows us to see its dependency on mass concentration. An r -value of 0.76 between variable A and B represents a direct proportionality between two variables. Since, OP_v is calculated by multiplying OP_m by mass concentration, then the linear relationship between the two measures is actually the dependence of both measures to mass concentration—mainly driven by meteorological conditions especially in the Alpine valleys.

This discussion is related to Fig. S3 where OP_m vs OP_v are plotted for all the three assays. The slope of this plot would be the inverse of PM mass. The reason of this plot is still not clear. One should plot OP_v vs PM mass conc. The slope of this plot would be average OP_m . If the correlation between OP_v and PM mass is very strong, it would reflect that the intrinsic OP of PM is uniform over the study site. If there is a poor correlation, intrinsic OP of PM is expected to be variable due to various reasons. I suggest to plot the Fig.S3 again in the recommended form and discuss.

My comment: Fig. 5: BB is not contributing to OP-DTT as much as it contribute to OP-AA and OP-DCFH. This is unexpected as OP-DTT is most responsive to organics. Please explain.

Reply: Thank you for this comment. We acknowledge the fact that OP from DTT assay has been reported to be responsive/sensitive to organics. However, recent studies have reported that OP from DTT assay is not affected by some metals (specifically iron) like other assays, namely AA and GSH. Because of this, OP from DTT assay may not fully capture ROS generated through Fenton chemistry or even the synergistic effects with regards to $\bullet OH$ generation as reported by Xiong et al. (2017). Similarly, Yu et al. (2018) has reported that soluble manganese showed synergistic effects with quinones on OP from DTT assay, while soluble copper appears to have an antagonistic effect with quinones on the same assay. On the contrary, manganese showed an antagonistic relationship with quinones on $\bullet OH$ generation. Quinones and soluble iron or copper react synergistically to form $\bullet OH$.

Generally, there is an undeniable interplay between species that needs to be considered as well as the sensitivity of each assay to species. As much as each analysis attempts to fully characterize the chemistry of PM, there can still be many species that are unmeasured but, in fact, plays a role in ROS generation. Hence, reported associations could be due to similarity in variations with PM concentration rather than a significant causal relationship between assays and PM components. Due to the sensitivity of DTT assay to wider range of compounds, such as organics and metals, that are present in various sources, this lead to a more balanced distribution of OP sources (and so weighting the contribution of biomass burning with regards to other sources) than the other OP assays, such as AA and DCFH.

This discussion shall be appropriately included in the revised MS.

My comment: Why industrial (or other) sources are responding differently to OP at different sites? Explain.

Reply: In the companion paper (section 3.5.1), we have presented the metric PD-SID (Pearson distance and standardized identity distance) that measures (dis)similarities of chemical profiles by each source. There are some sources that have been identified as heterogenous sources, including the industrial source. This means that the tracers used to identify the industrial source can be different between the 3 sites in this study. It could also imply that there is a varying origin of this source across the Grenoble basin. Due to this difference, it is expected that the OP contribution of the industrial source can be different as well, after all it is considered a heterogenous source. A similar comment by Referee #1 has also been addressed in Line 51.

Use of multiple tracers for Industrial source is confusing because different tracers respond to OP assays differently. Authors can split this Industrial source in different subsets using their unique proxy. In present form, it is very confusing for the readers.

My comment: How the OP-DTT, OP-AA, and OP-DCFH of PM10 observed over the study regions compare with the other parts of the world? This should be included and discussed.

Reply: The authors deem that this is outside of the scope/goal of this paper. After all, this is not a review paper on OP studies. However, our group has a paper (currently under review process in ACPD) that tackles the synthesis of OP measurements over many sampling sites in France.

This is not a correct thinking. Authors have reported the values of three assays over three closely located sites. It will be meaningful to add a paragraph (with a Table) on how the measured OP values compare with some other sites of the world with similar (or different composition). Add some discussion on this comparison. It is obvious that this MS is not a review article. But for readers, it will be useful to see some discussion on 'comparison'.

My comment: Lines 49-51: Give a proper definition of OP.

Reply: Thank you for this comment. This sentence now reads as:

Action: The oxidative potential (OP) of PM, defined as the capability of PM to generate ROS, makes an interesting complementary to regulated metrics of ambient PM exposure (Bates et al., 2019; Daellenbach et al., 2020; Guo et al., 2020; Gurgueira et al., 2002; Park et al., 2018; Shiraiwa et al., 2017; Valavanidis et al., 2008).

It should be -

....., defined as the capability of PM to generate ROS/deplete anti-oxidants,