

This manuscript showed the chemical composition and oxidative potentials (OP) of fine particulate matter (PM<sub>2.5</sub>) in Atlanta at a year-long time scale. Moreover, the authors investigated the correlation of probe-based aerosol OP with abundance of different PM constituents. They found that dithiothreitol- and ascorbic acid-based OP exhibited moderate correlation with the abundance of water-soluble transition metals (Fe and Cu) and organic compounds (WSOC and brown carbon), whereas the glutathione (GSH)-based OP showed strong correlation with the water-soluble Cu. Finally, the authors developed a multivariate linear regression model to evaluate the plausible contributions of metals, organic compounds, metal–organic and metal–metal interactions to aerosol OP. Overall the topic is interesting. The manuscript cannot be published in its current form, but it may be publishable in *Atmos. Chem. Phys.* if the following comments can be thoroughly responded in the revised paper,

1. What are the atmospheric implications of aerosol OP, which may merit the current work to be publishable in *ACP* rather than an aerosol or air pollution health related journal? Some relevant discussions may be needed in the section 1 or 4.
2. The authors mainly described the correlations of different aerosol constituents with the OP reflected by different types of acellular assays. However, the manuscript lacks discussions and insight into the underlying chemical mechanisms of the interactions among different probes and PM constituents in water or the synthetic respiratory tract lining fluid.
3. In the manuscript especially the Figure 2, the authors only showed the OP values in the unit of nmol/min/m<sup>3</sup>, which strongly associates with PM<sub>2.5</sub> concentrations. In contrast, the OP values in the unit of nmol/min/μg may exhibit stronger correlation with PM<sub>2.5</sub> composition. Therefore, the authors should present and discuss the OP values in the unit of nmol/min/μg as well as their dependence on different types of OP assays.
4. The Figures 1 and 3 are related to the chemical composition of PM<sub>2.5</sub>, and the Figures 2 and 4 are for aerosol OP. Thus, it may be more suitable to present the current Figure 3 as Figure 2, and the current Figure 2 as Figure 3.
5. What is the association of water insoluble organic matter and metals in PM<sub>2.5</sub> (in Figure 1) with probe-based aerosol OP?
6. L158-172: whether the efficiency of NADPH and GR to reduce GS-TNB to GSH can be interfered by the co-existence of ascorbic acid? Similarly, to which extent the co-variation of ascorbic acid and GSH concentrations will influence the OP<sup>AA</sup> and OP<sup>GSH</sup>?
7. L243-244: The sentence of ‘However, they could be considered as indicators of other compounds simultaneously produced by the same source’ is a vague statement, which needs further clarification.

8. L249: What does the 'PM species' exactly refer to?
9. L274 (3.3 Temporal variation): to discuss the seasonal distribution of OP clearly, the averaged PM<sub>2.5</sub> OP of different seasons should be presented in Figures 2, 3 or SI, similar like the seasonal distribution of different PM components in Figure 1.
10. L281: Except for aerosol composition, the concentration of PM<sub>2.5</sub> and size distribution of redox active PM<sub>2.5</sub> constituents may also influence the seasonal distribution of OP (Lyu et al., Environ. Sci. Technol. 2018, 52, 6592-6600). Thus, the temporal variation of PM<sub>2.5</sub> OP should be discussed deeply.
11. L305-313: It has been found that secondary organic aerosols-bound water-soluble substances such as organic peroxide, highly oxygenated molecules, and semiquinone radicals etc. are redox active in producing reactive oxygen species through reactions with water, antioxidants, or lung cells (Khachatryan et al. Environ. Sci. Technol. 2011, 45, 19, 8559-8566; Tong et al., Environ. Sci. Technol. 2018, 52, 11642-11651; Tong et al., Environ. Sci. Technol. 2019, 53, 12506-12518; Zhou et al., Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2019-190>; Chowdhury et al., Environ. Sci. Technol. Lett. 2018, 5, 424-430; Chowdhury et al., Environ. Sci. Technol. 2019, DOI: 10.1021/acs.est.9b04449), thus the contribution or connection of these and other relevant WSOC substances to the OP of PM<sub>2.5</sub> should be discussed properly.
12. The y-axis title of the upper left panel (for BrC) in Fig. 3 should be corrected.