

Interactive comment on “Development of a chromatographic method to study oxidative potential of airborne particulate matter” by Pourya Shahpoury et al.

Anonymous Referee #2

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Shahpoury et al. present a chromatographic method to determine the oxidative potential (OP) of airborne particulate matter. The instrument operates “offline” on PM samples that have been collected, transported back to the lab, and subsequently extracted into a simulated lung lining fluid. Overall, I find the work interesting and likely to be of interest to readers of AMT but substantial revisions and additional work are needed. Specifically, the method was developed and validated with PM standard that is nearly 40 years old; no validation with real-world (contemporary urban PM) was conducted. Further, the introduction contains many speculative statements that are incorrectly presented as facts, which I found especially disconcerting. The manuscript has both grammar and stylistic problems. For example, verb tense is inconsistent through-

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out the introduction and several noun objects are singular when they should be plural. I recommend rejection with an invitation to resubmit once the manuscript is improved.

Specific Comments:

1. ORP stands for "oxygen reduction potential" in the broader electrochemistry field and should not be used here as an acronym for "oxidation/reduction potential".
2. Figures 2-6: Kinetic data should be presented on a molar basis (or mass), not a percentage basis because (1) others cannot compare their results directly to yours and (2) some of the method variability may be masked by this approach since outcome measures become normalized to input concentrations.
3. Abstract, line 10: I take issue with this statement. The OP of PM is not a direct measure of inhalation toxicity. It is a measure of the redox activity of the particles in solution. While some researchers have correlated OP with select measures of cellular toxicity, it would be better to say that OP is often used as a "surrogate" to estimate one form of PM toxicity. Lead is a highly toxic metal, but PM made from 100% lead oxide is not likely to have any demonstrative OP associated with it. So if PbO has no OP in solution, does that mean that it's non-toxic??
4. Abstract, line 12: I take issue with this statement. Again, while inflammation of the epithelial tissue is often problematic from a health standpoint, it is not always true that oxidative stress and inflammation are the CAUSE of chronic disease. Inflammation has been associated with chronic diseases (like asthma) but epithelial inflammation is not the cause of all forms of asthma, it is a symptom of an underlying immune disorder.
5. Abstract, line 13: What does “rapid” mean? Be more precise/explicit. Since PM samples must be collected, transported, and extracted, I would not categorize this method as “rapid”. The term ‘rapid’ as used in this context seems a little misleading. The method may be faster than previous reports on methods that examine PM oxidative potential, but the method isn’t real-time and also requires physical collection of a PM

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sample (which can take several days to complete). Further, the authors never define why this method is "rapid" or how rapid it is....

6. The manuscript defines 15 acronyms in the Introduction section alone. The acronyms themselves are OK but with 15 explicitly defined it becomes nearly impossible to track them down later on. The manuscript needs a list of nomenclature.

7. Intro, page 2, line 3: The term "fine particulate matter" should not be abbreviated as PM but as PM_{2.5} since the fine refers explicitly to the fine mode.

8. Intro, page 12, line 10: I take issue with the supposition that "inorganic salts" and "crustal dusts" are non-toxic. Crustal dust can contain heavy metals and microorganisms, in addition to elements like crystalline silica - all of which are potentially toxic or inflammogenic. Lead can also exist as an inorganic salt.

9. Intro, page 2, lines 12-14: This statement is outright wrong and the reference is incorrectly used as support. The Ayers article makes no such claim. Indeed, the second to last sentence of the concluding section of the Ayers paper states "If, however, it can be demonstrated that one or more oxidative stress potential tests are better predictors of adverse health outcomes than more traditional mass metrics, there may be a case to base air quality policy upon such tests." While I agree that OP has been shown to correlate well with certain measures of toxicity in vitro, there are only a handful of studies that have demonstrated this behavior in vivo.

10. Intro, page 2, line 14: This statement is incorrect. The OP is more often defined as the ability of the compounds present in solution to act as oxidizing agents.

11. Intro, page 3, line 24. Change "this" to "these". The word data is plural for datum.

12. While the ORP experiments are interesting, they are ancillary to (and largely decoupled from) the main focus of the manuscript. The ORP work should be more thoroughly vetted/explored or removed and submitted as a separate work for publication once properly done.

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13. Methods, page 4, line 25: I would hesitate to call SRM 1649b "typical urban PM" today. First, this sample was collected in Washington DC in 1982, so it perhaps represents urban PM from 37 years ago. Second, the NIST sample was a 12-month collection period and was not stored at -80 so all of the semi-volatile material (which likely contributes to PM OP) has likely evaporated. Third, this sample was collected from a large baghouse that was treated with copious amounts of fungicide (run the sample through a GC/MS in scan mode and look for the dichlorophene peak). I recommend additional validation with contemporary PM samples.

14. Figure 2 is not needed. This information can easily be included in one of the results tables or simply in the text alone.

15. Results, page 8, and elsewhere: Several of the findings reported in this work have been reported previously, such as the non-linear effects of PM concentration on thiol oxidation (Charrier and Anastasio, 2016) but those previous reports are not discussed or acknowledged here. A more thorough review (and inclusion) of the recent literature on PM OP is needed.

16. Conclusions, page 10: The authors suggest that AA is a better candidate for determining the OP of ambient PM but fail to recognize that many of the epidemiological studies, to date, have not reported associations between AA-determined OP and various adverse health effects (whereas GSH assays and DTT assays have shown associations with disease). The authors should conduct a more thorough review of the health effects literature before making such a claim.

17. Figure 3 - Why does the ORP of the reference SELF change over time?

18. Figures - Incubation time and PM concentration should be included in the Figure label where relevant.

19. I also think Figure 4B can be deleted since it was shown in Figure 3 that product formation is biased low (from a stoichiometry standpoint) because of likely complex

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formation between products and other solution compounds/proteins.

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