

This article reported three methods of analyzing aerosol oxidative potential (OP) based on DTT assay, focusing on different sample extraction strategies to distinguish soluble components from the total aerosol particulate matter (PM). I think this is a good work with practical meaning to assist the understanding of oxidative aerosol's transport in atmosphere. I have one major suggestion as below.

Figure 1 delivered a clear demonstration of the three sample extraction and analysis methods. However, I think a better job can be done to explain what component(s) from the total PM species is measured by each method throughout the text. Basically, two factors, including extraction solvent (water versus methanol) and filter (i.e., with or without filtering) differentiate the three methods. Which component in the aerosol is screened by each factor should be clearly specified to elucidate what is the actual difference across the three methods. From my understanding, the use of water or methanol discriminates the water-soluble (herein "soluble" matter includes dissolved molecules/ions and "dispersed" solids (or small particles)) and methanol-soluble component, whereas filtering or not filtering a sample discriminates dissolved molecules/ions and dispersed solids in that sample. Overall, extraction combined with these two factors actually categorizes the total PM into six groups: water-insoluble species (those which cannot be extracted into water), water-soluble molecules/ions, water-soluble solids (dispersible small particles), methanol-insoluble species (those which cannot be extracted into methanol), methanol-soluble molecules/ions, and methanol-soluble solids. The authors should clarify which groups are measured in each of the three methods, either with a diagram or plain text.

Elucidating the above may rationalize a few ambiguous places better in the articles. What follows are some examples.

L59-60,76-77, there seem to be some conflicts between these two references: the first place says up to 99% of the DEP CANNOT be extracted by water or methanol while the second one says the measurement is based on water extraction. "after which the filter was removed from the analysis" is very confusing. What are the authors trying to say here?

L100-101: what is "10%" here?

L300-323: The justification of choosing method 3 over the other two is poor. It maybe improved if the authors can specify each species category corresponding to each separation technique as I suggested. I have specific questions as below:

L305-309, it seems method 1 is more consistent with SCAPE study, than method 3; then why is method 3 selected over method 1?

L309, what is indicated by "very little correlation between $OP^{WI-DTT-3}$ and OP^{WS-DTT} ?"

L315, I am uncomfortable with "overall" here, because the only justification of method 3 over method 1 is that it measures higher total-DTT (which is actually

quite common sense). The whole paragraph seems quite over-informative and little relevant to the conclusion.

L319-323, the same problem is with the comparison of method 2 and method 3. There is not actual justification for why method 3 is chosen over method 2. Information is very redundant and little relevant to the conclusion.

L330-339, it's somewhat ironic here. I was told that method 1 is better than method 2 and 3 in terms of seeking correlation between OP-DTT and PM compositions but then suddenly method 3 was selected to be used... Why?

L348: I recommend clarifying how 35-42% is determined and specify the error range as well.

Overall, I strongly suggest the authors give their recommendations on which method should be used in what scenarios or for what purposes.

Other minor suggestions:

L133-134: provide possible explanations on what gives the response in blank samples.

L275-278: Maybe provide a figure in SI to illustrate the validation with five ambient samples as well?

Table 1: Does "N" filters correspond to different samples? (I assume CV is determined with three replicates on each filter (each sample)?) and what is the range standing for.

Table 2: I think a figure is better to let readers see the correlations of different variables, although I know there are many comparisons here. Maybe the authors can think if the illustration here can be improved.