

***Interactive comment on* “Oxidative potential of ambient water-soluble PM_{2.5} measured by Dithiothreitol (DTT) and Ascorbic Acid (AA) assays in the southeastern United States: contrasts in sources and health associations” by T. Fang et al.**

Anonymous Referee #3

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Review of “Oxidative Potential of Ambient Water-Soluble PM_{2.5} Measured by Dithiothreitol (DTT) and Ascorbic Acid (AA) Assays in the Southeastern United States: Contrasts in Sources and Health Associations” T. Fang, V. Verma, J. T. Bates, J. Abrams, M. Klein, M. J. Strickland, S. E. Sarnat, H. H. Chang, J. A. Mulholland, P. E. Tolbert, A. G. Russell, and R. J. Weber

Summary This manuscript uses a number of important methods to bridge the continuing gap between detailed composition information and the association between PM and negative health effects. The DTT and AA assays were run on PM extracted from a

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number of locations across two year in the southeast United States. Correlations were made between the DTT and AA assays and PMF and CMB modeling of sources and with ED admissions from long term epidemiological data. On the whole I feel the paper has a lot of strong characteristics, particularly the new assay methodologies and their connection with source modeling. I think the paper overall is fairly strong and should be accepted with minor revisions.

Major Comments One concern is that there is a lot of detail and information in the methods section, but that the results and discussion section feels light on new results. As noted by another reviewer, a large amount of the DTT data has been shown before in prior publications. Though comparisons with prior data are good, it feels a little like a second bit at the apple for the DTT data. A revised manuscript that shows more data from the AA results would be compelling, though perhaps due to the strong Cu response there was concern that the AA results were not strong enough on their own? I rarely suggest making a paper longer, but at 18 pages with only 4 of results and discussion that could really be expanded and strengthened.

The extensive literature citations are commendable and the introduction very nicely sets up the need for this work. Along the lines of the previous comment, at times I felt like the new findings from this study got lost in the sea of references to this group of authors previous papers, particularly those from the last 2 years and Balachandran et al. 2012. A revised manuscript that more clearly delineates the findings of this study from prior work will help readers understand the findings of this work.

Though the data is admittedly abundant and at times dense, the results section feels like a list of numbers in a few paragraphs. Rewording to bring the science out from behind these numbers would be helpful.

In Figure 3 the amount of AA response from brake/tire wear being 44% is notable. I worry that since as the authors note that these are mechanically generated particles that they are likely solid and smaller than the 0.45 micron filter used in the methods.

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The still solid metals might only be dissolved when nitric acid is introduced prior to the XRF analysis. If so this would seem to suggest that this is not truly a water soluble, but rather an insoluble source that becomes soluble during analysis. Overall the possibility of solid particles less than the 0.45 μm filter should be addressed in more detail. If this concern is plausible then some of the phrasing in the manuscript (and potentially even the title) should be qualified since much of the AA response might be from insoluble material.

AA is used as a surrogate for a more complex lung fluid and found to only really correlate with Cu. Are the authors concerned that perhaps this more simplistic model fluid is not capturing sensitivity to other species that perhaps a more complex simulated fluid might? A little bit of further discussion on whether AA is really a great choice would be helpful, otherwise it might not be sensitive enough to mimic the responses in the lung that are observed from other types of studies.

Minor Comments/Concerns

LWCC is defined on both 30617 and 30619

700 nm was chosen as the background, but some things in the atmosphere can absorb at that wavelength. Washenfelder et al. 2015 GRL showed episodic variation of absorption for the 4 wavelength of aerosol absorption they monitored (highest was 530 nm). Have the authors done checks with full absorption spectra to determine if 700 nm is truly a safe choice for background? This would be particularly important when biomass is playing such an important role in both absorption and DTT assay activity.

As a curiosity is there a reason that XRF was used and not ICP-MS?

This might just be a lack of knowledge, but ammonium bisulfate is a class listed for one of the models. How is that determined and separated from ammonium sulfate? Is that from ISOROPPIA or some other thermodynamic model?

Section 2.3.3 Though referencing the other work is important, I am a bit suspicious of

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using PM mass concentrations estimated from the sum of chemical component from Hi-Vol samples. More detail on how this has been justified (even one sentence), beyond just referencing citations would appease that concern.

Are the %'s (filter sampling, etc.) chosen for propagating uncertainty on page 30632 from a reference or chosen arbitrarily? I was not clear from the text where they came from.

Page 30630: “the model did not observed” should be “the model did not observe”

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 30609, 2015.

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