**Interactive comment on** “Reactive oxygen species associated with water-soluble PM2.5 in the southeastern United States: spatiotemporal trends and source apportionment” **by V. Verma et al.**

Anonymous Referee #4

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Although it is not yet known which chemical species drive the correlation observed between particulate (PM) levels and negative health outcomes, there are a number of PM assays that are thought to relate in some way to these effects. The DTT assay is one, where the assay measures the ease by which PM can transfer electrons from electron-rich substrates to oxygen under physiologic conditions. While this assay has been used in controlled laboratory and short-campaign style investigations, this is the first study which reports the DTT response across a coupled spatially and temporally resolved study. In particular, measurements were conducted over several seasons in
both urban and rural locations close to Atlanta. It is found through both PMF and CMB approaches that a variety of sources contribute to the response and that there is a relatively uniform response across the study dimensions of time and space. This is significant because it indicates that this intrinsic property of the particles is relatively uniform, at least for the extent of conditions sampled. As well, this sub field has been focussed on either a metals-based or organic (usually quinone) based interpretation of significance. I like this paper because it highlights that it is both metals and organic components that are important, depending on the source of the PM.

I recommend publication of the paper. It is well written, it is a significant step forward in terms of scope of DTT field measurements, and it is very much strengthened by the extent of (a lot of) simultaneous PM chemical measurements performed.

A slight weakness in the paper is that the PMF analysis is not described in detail, in particular to the choice of the number of factors chosen. Can the different factors chosen be shown to be independent of each (perhaps by including R2 values for their relative correlations)? I also wonder how the uncertainties of each chemical species included in the PMF are handled, and whether there is any need to down-weight the importance of one input variable or another, if their uncertainties are unusually low compared to others?

Also, although this is perhaps in the references, I would like more information on how the DTT analysis was actually done, e.g. temperature, buffer solutions, how DTT loss is analyzed, extraction of species from the filters, sample handling, etc. There are many ways to do this assay and so it is important to add these details.

Interactive comment on Atmos. Chem. Phys. Discuss., 14, 19625, 2014.