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Atmospheric Chemistry and Physics
Discussions
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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.

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§2.3: How long were the filters stored at -18âŮę before analysis? Did the author observed any reduction/increase of the DTT activity during this storage process?

Response: The sampling began in June, 2012 and continued till September, 2013. The analysis of the stored filters started in March, 2013 and completed in January, 2014. Thus, the filters were stored in freezer (-18 oC) on an average for about 6 -12 months. The decay of DTT activity during storage might be a valid concern, however, this has not yet been assessed through a systematic study. For example, a study where DTT activity of stored filters is measured at various time intervals (e.g. after 10, 20, 30, 50, 80. . ..days) is planned. Additionally, our lab is developing an online system for measuring the DTT activity of ambient PM in real-time, to help address the concerns related with chemical alteration of collected particles in the stored samples. Following reviewer's comment, we would explicitly note in the present manuscript that the samples have been stored for up to a year prior to analyses and therefore the DTT activity measured on these samples represents only the activity associated with stable PM compounds.

§2.6: The measured DTT activity in this study corresponds to a sub-fraction of the total PM2.5 (water extractable fraction). For consistency reasons, the expression of the DTT reactivity should be based on the amount of the water extracted mass and not based on the total PM2.5 mass. This last calculation could be done only if the insoluble mass fraction is negligible compared to the water soluble fraction. Is it the case in this study?

Response: What normalizing mass should be used to determine the intrinsic DTT activity is a complex question. As the reviewer notes, water-soluble PM mass seems more appropriate when focusing on water-soluble DTT activity. However, we couldn't measure this mass in the present study, given the particles were collected on quartz filters. But, it could be argued that even the water-soluble mass is not perfect since many water soluble components are not likely to be DTT active, such as inorganic species, yet they comprise a large fraction of the water-soluble mass. In a subsequent paper (Verma et al., 2014, submitted to ES&T) we report the intrinsic activity for specific organic sub-fractions, determined with an aerosol mass spectrometer, which provides more insights than normalizing by total mass or even water-soluble mass. We believe the main issue is that the normalizing parameter be explicitly stated, which should then remove all ambiguity. In this paper, we have normalized the DTT activity by total PM2.5 mass, which was determined from the parallel TEOM measurement.

§3.2, line 6-8: Regarding the daily variability of the DTTv activity, the authors attribute

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it to different and general factors. Do the authors have access to meteorological data collected during this sampling period and could they identify variables such as temperature, solar irradiation, wind, ozone level, which may contribute to such variability?

Response: In our companion paper (Fang et al., 2014), we showed that the volume-normalized DTT activity is well correlated with PM2.5 mass concentrations. Therefore, the factors (including the meteorological parameters in addition to the daily variations in emission sources) that affect PM2.5 concentrations at a site similarly affect the DTT activity. Thus, in the context of present study which focuses on the emission sources of DTT activity, assessing the role of meteorology is not important given the daily variability of DTT activity can be explained by the similar variability in PM2.5 mass concentrations.

§3.3.1: As EC and OC have been determined on the total particle (insoluble + soluble part), their correlation with the DTT activity should be done with the total collected particle only and not with the soluble sub-fraction as it is done in this study. It is possible that the insoluble part contribute to some extent to the DTT activity (see for example McWhinney et al., “Filtrable redox cycling activity: A comparison between diesel exhaust particles and secondary organic aerosol constituents”, Environ. Sci. Tecnol.,2013, 47, 3362-3369). Do the authors have some information about the contribution of the insoluble part to the DTT activity?

Response: The reviewer is correct that the insoluble fraction of ambient PM contributes significantly to the DTT activity, as shown in McWhinney et al., 2013 and also our previous paper (Verma et al., 2012; about 20-50 %), however, the present manuscript is focused only on the water-soluble fraction of DTT activity. The correlation of EC and OC with the water-soluble DTT activity as shown in this study is not to imply that these components are participating in the DTT reaction, but to indicate their sources (e.g. as source tracers - vehicular emissions for EC, and secondary sources and biomass burning for OC), which also probably emit the water-soluble species contributing to the measured DTT activity. Thus, EC and OC should be considered as the surrogate for

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those unknown DTT-active water-soluble PM species.

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