

Interactive comment on “Quantification of environmentally persistent free radicals and reactive oxygen species in atmospheric aerosol particles” by Andrea M. Arangio et al.

Anonymous Referee #4

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This paper describes measurements of the concentrations of reactive species embedded in atmospheric aerosol collected from the roof of the MPI. It is a followup to the earlier paper this year by Tong et al which compared field samples to lab samples collected for a shorter period of time and focused on OH generation. The measurement method consists of extracting soluble molecules from the particles and reacting them with a scavenger. This is essentially a physical chemistry paper and my comments are from that perspective. This paper will be publishable after some edits to respond to the following comments.

(1) I found the terminology used by the authors to be confusing in places. Through use of words such as “we have also characterized and quantified ROS including OH,

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Discussion paper



superoxide (O₂⁻) and carbon- and oxygen-centered organic radicals, which were released upon extraction of the particle samples in water.” the reader could conclude that the radicals are persistently present in the particle, rather than being formed by reaction of water with precursors during the extraction process and later scavenged. The multiple chemical steps involved in the experiment lend an ambiguity to how to relate the lab processes that are responsible for formation of detectable spins to atmospheric and physiological processes. As another example, on pages 4-5 the authors talk about spins per microgram but do not define this quantity. I presume they mean spins detected by extraction from a sample of this mass using the protocol described. The concentration of the scavenger is not given so it is not clear how closely this process is controlled or how repeatable it is. Clarification of the terminology and relation of the experimental conditions to those found in the lung and in clouds, for example, would be helpful.

(2) Only particles smaller than 1 micron contain extractable ROS material. Do the authors understand why this is? Since peroxides are photo labile I might have expected the opposite - the larger, more optically opaque particles would have more precursors than the smaller ones assuming the extraction processes work the same way for all particle sizes. Some discussion of the size effects would be useful.

(3) The reactive oxygen species released and scavenged during the analytical protocol are well known to have rich chemistry in water and very different reactivities compared to each other. Have the authors determined how efficiently are they being detected (absolute and relative values)?

(4) On page 9 line 269ff there is a section discussing implications for aerosol chemistry and lung chemistry. Since there are no data in this paper specifically looking at these implications but there are in Tong I recommend the reader referred back to the earlier paper instead.