The paper from Arangio et al. measures the concentration of environmentally persistent free radicals (EPFR) and reactive oxygen species (ROS) in size segregated ambient aerosols. EPFR were measured directly by EPR spectrometer, while the ROS were measured by extracting the particles in water and then EPR analysis. As per the reviewer's knowledge, this is first comprehensive measurement of EPFR and ROS in size-segregated aerosols. ROS are an important species in ambient aerosols and could be biologically relevant. In addition to these novel measurements, authors also throw lights on the possible mechanisms of ROS generation through redox cycling between organic compounds and transition metals. An improved understanding of these mechanisms is important to comprehend the aging process of atmospheric aerosols. The paper is well written and easily comprehensible. Therefore, I recommend the publication of this manuscript. However, I have few comments which the authors should consider to make their work better:

Page 2, Line 42: Are there literature evidences that organic radicals also mediate in the oxidative stress? If yes, then authors should include them.

Page 3, Line 87: Why these two samples were collected for a longer duration? Are the authors not concerned about the loss of semivolatiles during that long sampling duration?

Page 4, Line 128: Authors should somewhere explain these units of spins μg^{-1}, probably in the method section.

Page 4, Line 129-131: Can authors elaborate on their sentence that EPFR distribution is similar to soot? Do you mean that there is commonality in the sources of two?

Page 5, Line 143: Why the samples collected on these two days are significant and discussed separately?

Page 5, Line 143-150: I am not sure why the authors have discussed the sampling duration separately. The EPFR concentration expressed in units of spin/μg should not be affected by the sampling duration.

Page 6, Line 176: Is it 41 % or 40 %?

Page 6, Line 201: What are the units here for ROS, is it spins/μg?

Page 7, Line 203-215: I think the authors are completely confused here. DTT assay doesn't measure the ROS in the particle, rather the capability of particles to generate ROS in surrogate biological environment. I am not clear what the authors want to deduce in this discussion and what is the significance of this number of (2-7) x 10^{14} ug^{-1} of DTT molecules? It is important to note that DTT activity is a completely arbitrary unit and depends on the initial DTT concentration used in the assay.

Page 8, Line 237: Can the authors add references showing HULIS is known to contain substantial amount of quinones?

Page 10, Line 308-310: I don't think that this study shows that ROS can be generated in lung fluid. I think again the authors are confusing between ROS activity (capability of particles to generate ROS) vs. ROS on the particles (measured in this study).