

Interactive comment on “Impact of aging mechanism on model simulated carbonaceous aerosols” by Y. Huang et al.

Anonymous Referee #2

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This paper presents a global modeling study on how aging due to ozone oxidation affects the lifetime, burdens, and concentrations of carbonaceous aerosols, with aging here defined as the conversion of carbonaceous from being hydrophobic to hydrophilic. This is a very important topic because accurately modeling this aging process is critical to predicting carbonaceous aerosol concentrations and determination of their health and climate impacts. While the topic is important and well within the scope of ACP, I have major concerns with this paper and do not recommend it for publication.

Major Comments

1. As noted by the other reviewer, this study considers only one of several aging mechanisms. This is a major weakness that renders the paper incomplete for

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publication in ACP. The only aging process considered in this study is the ozone oxidation of organic material coated on the carbonaceous aerosols. There are several issues with considering only this mechanism:

- (a) The impact of this specific aging mechanism has already been studied using a global model by Tsigaridis and Kanakidou (2003). In fact, this study used the exact formulation as Tsigaridis and Kanakidou (2003) (based on the chamber study of Pöschl et al., (2001)); therefore, the implementation of this mechanism and implications discussed in Section 3.2 are not new.
 - (b) Among all the processes by which carbonaceous aerosols are aged in the atmosphere, oxidation by ozone is not even the most important. The processes by which carbonaceous aerosols are aged are summarized nicely in section 5 of Kanakidou et al., (2005). At the minimum, this study needs to consider aging by condensation of H₂SO₄ and coagulation, which are more important in polluted regions.
 - (c) The aging process implemented in this study is based on ozone oxidation of benzo[a]pyrene coated on carbonaceous aerosols. Benzo[a]pyrene is not representative of organic material on carbonaceous aerosols. The implication of this should be discussed.
2. I also agree with the other reviewer that comparison to observations at two marine sites is absolutely inadequate. There is no reason provided why those two sites were chosen over the other sites. As the authors noted, the two sites evaluated are implicated by marine carbonaceous aerosol– so why not chose sites where marine carbonaceous aerosol is not a concern? The evaluation should not be limited to IMPROVE sites but should include other surface measurements outside of the US. Furthermore, since significant differences are found above the surface layer between the updated and the control runs, a comparison of modeled vs.

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observed vertical distribution is needed. There are data from field projects, e.g., ACE-Asia, that can be used for such comparison.

Other Comments

1. Line 2, Page 28999: "This *significantly* [emphasis mine] improves..." The use of "significantly" overestimates the improvement because the improvement is much smaller than the difference between the updated model results and observations.
2. Line 21, Page 28999: " τ in surface air ... (Table 1)." "Surface air" here is a typo. The values reported in Table 1 are global average lifetimes, including contributions from above the surface layer.
3. Page 28997, around equations (1) and (2): The text should mention that this is the same formulation implemented in Tsigaridis and Kanakidou (2003).

References

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