Improvements of organic aerosol representations and their effects in large-scale atmospheric models

The authors present a methodology for describing the chemical oxidation state in terms of O:C ratio of organic aerosol (OA) based on OH exposure times for computational efficiency and use in large scale modeling. Using 5 sensitivity scenarios, they evaluate the effects of including description of OA aging using the EMAC climate model. This is an interesting work and merits publication in ACP. But I have several comments which need to be addressed before the manuscript is accepted.

1. Abstract: The authors claim that including ageing of OA results in more accurate description of water uptake by organic aerosol. But in section 3.3 they do not validate their organic aerosol water uptake with measurements. I suggest the authors rephrase their sentence.
2. Page 10334: Last sentence: Rephrase “reasonable contribution of organic material to mixed aerosol particle”. I think the authors mean “representing the spectrum of OA oxidation states”.
3. Section 2.1.1: The OC tracer is split into 6 species representing different oxidation states in terms of O:C ratio. The last sentence of page 10338 says this approach requires fewer tracers compared to Shrivastava et al. (2011) in the WRF-Chem model. But this does not seem right. The 2-species VBS developed by Shrivastava et al. (2011) requires 2 tracers for a given non-volatile SOA source and size bin. These represent the carbon and oxygen parts of that species. The primary emissions are represented by 2 non-volatile tracers and 2 tracers for gas phase. In summary both primary and secondary OA could be represented by 6 tracers for any given source and size bin. This is equal to the number of 6 OC tracers used by the authors. But the 9-species VBS in Shrivastava et al. (2011) is more computationally expensive. This point should be clarified.
4. The authors also need to better explain how their scheme divides the OC tracer into different species based on OH exposure time. How do they use the time varying concentration of OH of respective grid box to divide the OC tracer? Including an equation depicting this approach would help. Also, in case of insufficient OH concentrations or too short an exposure time, how do they decide what fraction of mass of the tracer is transferred to the next higher oxidation state? Since this methodology is central to this paper, the authors should clarify it better in the text.
5. Page 10341: How do the authors decide “Best Guess” for emission attribution? What sources constitute the hydrophilic organic carbon emissions? The biomass burning and marine emissions have a different emissions attribution, so it’s necessary to mention other hydrophilic SOA sources (including biogenic SOA). Are fossil sources of SOA considered hydrophobic? Are there any references for their choice of specific emission fractions into WSOC categories?
6. Page 10342: last sentence. It’s not clear what is meant by “unaged OC is hardly converted any more to hydrophilic OC with a low O:C ratio, but only into categories of higher O:C ratio”.

Shouldn't this process depend on OH exposure time resulting in a spectrum of O:C ratios for hydrophobic material similar to hydrophilic material?

7. Page 10346: Line 5: "Even though the model shows a similar tendency in increasing values as the observations, the magnitude cannot be reproduced." This is not clear. Modeled values in Figure 4 are often tightly clustered between O:C ratio of 0.4 and 0.5, and do not seem to have any spatial patterns similar to measurements.
8. Page 10353: Line 5: The authors mention that in regions of mixed pollution inorganic aerosol components also contribute significantly to aerosol water uptake. What about contribution of dust to aerosol water uptake? Does dust represent a source of uncertainty in their calculations of aerosol water?
9. Page 10354: Line 5: The no-aging aerosol case shows less water only in accumulation and coarse modes, but not in aitken mode in Figure 2. Why? Also, the authors need to revise their sentence which currently states that no-aging aerosol shows less water in all three modes (not true for Aitken mode).

Other comments on Figures:

Figure 1: The units of OH exposure time on X axis is not right. It should be (molecule cm^3 s)

Figure 2 captions: The authors should define what is meant by k, i, s, a and c. These may be confusing to readers if not defined clearly. For example: k, a and c denoted Aitken, accumulation and coarse modes, i and s represent hydrophobic and hydrophilic aerosol components.