Interactive comment on “Improvements of organic aerosol representations and their effects in large-scale atmospheric models” by H. Tost and K. J. Pringle

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Reply to reviewer 1 of our manuscript:
Improvements of organic aerosol representations and their effects in large-scale atmospheric models by H. Tost and K. J. Pringle

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.

We thank the reviewer for his positive acceptance and constructive proposals for improving our manuscript.

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.

We agree that we cannot make such a statement, since we do not evaluate our water uptake approach with measurement. The more “accurate description” we obtained by taking the additional information of the O:C ratio into account, which represents a more physically or chemically based approach which we esteemed “better”. This will be rephrased to make clear that the consistency is meant and not a comparison with reality.

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”.

As the other reviewer pointed out as well, this was exactly the point we wanted to make, but unfortunately we simply did not write it this way. This will be corrected.

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.

We agree, that the 2-species VBS by Shrivastava et al. (2011) does not require more compounds, the other VBS approach is computationally more expensive. However, Shrivastava et al. (2011) state that the 2-species VBS provided partly unsatisfactory results for simulating SOA (similar to the 9 species VBS) and it requires some more calculations than our simplified approach. Nevertheless, the partitioning cannot be represented by our scheme making it inferior to any 2D VBS approach.

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.

We will elaborate more on this in the revised manuscript. The concept is as follows: the online calculated OH concentration in each grid box is used to calculate an OH exposure time by using the length of the model time step. As we assume that the OH is not net consumed during the processes (or it is continuously recycled) a constant OH concentration is maintained per model time step and grid box. The individual tracers are equivalent to a minimum OH exposure time and hence act as a starting point on the function shown in Fig. 1. Then the oxidation state follows the fitted function for the corresponding OH exposure time as an increment. The last full threshold passed determines the new minimum oxidation state. However, a fraction is moved to the next
tracer (=oxidation step). This fraction is determined by an interpolation routine using again the OH exposure time increment and the relative differences to the lower and higher threshold value. Consequently, even the smallest OH exposure time can lead to a transfer into the next higher oxidation state, but the fraction of the total mass will be small as well. An oxidation past the upper threshold value is not done, hence the values for O:C ratios larger than 0.7 cannot be reproduced with this approach. However, since the linear relationship between O:C ratio and kappa is not based on measurements for higher O:C ratios, we simply state that in terms of hygroscopicity this is the maximum the organic compounds can reach.

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?

The terminology of a “best guess” resulted after analysing the results, e.g. comparing the kappa values for Amazonia from the model with results from AMAZE and the O:C ratio with the data from Ng et al. (2011). Since this emission assignment represented still a reasonable (based on our own expertise) distribution and it gave the best results and did not destroy the comparison with atmospheric burden of OC compared to observations from e.g. the IMPROVE network (as the “Insol” simulation did). The hydrophilic OC emissions originate from biomass burning, marine emissions, and biogenic SOA. OC from fossil fuel burning is assumed to be completely hydrophobic, and the global emission data set used only provided primary emissions from fossil fuel. The choice of the emission assignment mainly is based on a Gaussian distribution, such that the mean kappa value of the emissions is also close to 0.1 as the value assumed for the “No-Ageing” simulation.

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?

This sentence has to be rephrased, since it was misleading. The reviewer is right, that based on the OH exposure time a spectrum of oxidation states is achieved. The hydrophobic material is not allowed to age in the Ageing-BG scenario. Therefore, the ageing process can start only after a conversion into the hydrophilic mode by coagulation or coating. However, after those microphysical processes the aerosol particle are usually grown and are found in the accumulation mode, and hence the OCks has a relatively low importance. Additionally, the O:C ratio increases quickly for low OH exposure times and low O:C ratios whereas the slope of the function gradually decreases.

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.

Based on the suggestions of reviewer2 we will replace the figure by one using the correct seasons for the comparison and the results have slightly changed. Still the maximum values for the O:C ratios are not obtained due to the lack of values higher than 0.7. A season dependent deficiency is found, i.e. summer values are relatively well represented, whereas winter values are usually underestimated. This could be related to insufficient information at the emission stage of different types of emissions, e.g. OC from local heating, power plant energy production etc. The discussion of the Figure and the statement in the conclusion will be modified in the revised manuscript.

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?

Dust and BC are assumed to be completely hydrophobic, i.e. even after transfer into the hydrophilic modes, they do not contribute to water uptake. Even though we are aware of potential water uptake by calcium compounds (especially after coating with other acids) we do not consider these processes. Therefore, dust is a source of uncertainty, but we tried to minimise it in the most conservative way, i.e. by ignoring the water uptake on dust.

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).

The “No-Ageing” case resulted in OC kappa values lower than 0.1 for the aitken mode over some of the continents compared to the ageing simulations (e.g. greenish colours in South America and Central Africa in the left pattern of Fig. 5). Consequently, the water uptake is lower than in the case where a constant OC kappa value of 0.1 is assumed. This results from the emission assignment into the Aitken mode in this regions, which is not fully achieving the Gaussian distribution around a kappa of 0.1 as desired. The sentences has to be modified as the reviewer correctly pointed out.

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

We completely agree, and thank the reviewer for pointing this out.

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.
We also agree and will extend the caption to contain this information as the reviewer correctly guessed.

References: