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

Anonymous Referee #2

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The manuscript presents a modeling study which calculates the O:C ratio of organic aerosols based on the aging time against OH oxidation in a CPU-efficient way, suitable for large-scale modeling. The authors study the importance of different chemical aging parameterizations and compare them with the default no-aging version of their model. They also construct a “best guess” (BG) scenario for emissions distribution based on their oxidation state and use that as a reference. The paper is generally well written and I suggest publication to ACP after addressing the comments/concerns outlined below.

In general, I do not understand why the authors decided that the hydrophobic aerosols
cannot oxidize. They are less oxygenated, thus they should have higher vapor pressure and even probably some unsaturated bonds, making them susceptible to volatilization and oxidation. I see no reason why their BG simulation is not the “Insol” one.

The discussion is overly qualitative. When comparing different simulations the authors should try to make it more quantitative, when possible.

There are two “kinds” of agings in this study: one is the chemical one, and one is the microphysical one. Not everywhere it is clear which process is implied. Care must be taken throughout the manuscript, and especially in section 2.2, to clarify this every time aging is mentioned.

Abstract, lines 20-22: I do not understand this sentence at all.

Page 10333, lines 9 and 22: It is not true that most models have only one tracer. The simplest models have at least two tracers, in order to parameterize fresh and aged organic aerosols.

Page 10334, lines 9-12: This assumption, although clearly stated throughout the manuscript when needed, might not necessarily be true for all cases around the globe. In fact, the authors do mention it later (end of page 10351). A statement is needed that quantifies how much this assumption will affect the results, and, more importantly, whether it will modify the conclusions. If needed, an additional simulation can be helpful.

Page 10334, last line: I do not understand what this means.

Page 10337, top: how can hydrophobic material go in the cloud? It is only partly hydrophobic? Also, what is the bulk sea salt?

Page 10338, line 5: OH is not abundant, it is only extremely reactive, which is why it is so important.

Page 10339, line 3: The approach described here is very old and is being abandoned
fast by global climate models. It should not be mentioned here as the current standard, but rather as an obsolete parameterization that should be eliminated.

Section 2.1.2 needs great expansion; it is extremely short for the importance it has and the discussion that follows. The reader needs to know more about the processes described here. In addition, although it is obvious that it will make the model more computationally expensive, it is not clear why it is suitable for “process research rather than long-term climate studies”. A reference or some explanation is needed here. The last sentence of the section is also confusing: is this process included or not in the current study? Aqueous SO2 oxidation is very important, is it included in the simulations or not?

Page 10341, lines 11-13: these add to 50%, where is the rest half?

Page 10341, lines 15-16: the marine organics are first mentioned here. The reader knows nothing about them, how they are being produced, what they are, what are their properties, their mixing state with other aerosols, etc. They are being used later in the discussion, thus a description of them is needed somewhere in the model description sections.

Page 10341, point 5: This aging happens via which mechanism? Same as the hydrophilic one? At what rate?

Page 10345, line 15: no need to believe that the other model is correct. “accurate” should be used with caution, maybe “similarly” is a better word.

Page 10345, lines 20-22: This should also be mentioned in the figure legend.

Page 10346, lines 5-6: I do not see a similar tendency in the model results, they are rather flat, compared to the measurements that show up to a factor of 2 differences.

Page 10347, lines 1-6: how can you perform statistics with just a single year of simulations? The 12 data points of the monthly means are not good numbers to be used to compare 2 annual means, due to the strong seasonality. The correct way to do it is to
have 5 or 10 years of simulations and compare the 5 or 10 annual mean numbers of a simulation with the corresponding data of another simulation.

Page 10347, line 8: I wouldn’t use “storm tracks”, since this implies that storms are related with (or, they are responsible for) the low values. In case they are, this has to be shown.

Page 10348, second half: It would be interesting to calculate the mean time that takes for the changes to not be important at all. This will show how far downwind of sources the emissions assumption is unimportant.

Page 10352. Line 7: “less efficient” compared to what?

Section 3.3: How about aerosol size? What fraction of water per size is due to organics? How about the fraction at CCN sizes?

Page 10360, lines 15-20: Does this change improve results when compared with measurements, or not?

Page 10360, lines 24-25: If you are referring to figure 4, I disagree. There is no reasonable agreement.

Figure 4: This is a very interesting figure and well explained in the text. I suggest trying to find the temporal information from Ng et al. and plot it with the appropriate model data, not the annual means. I have the feeling that this might improve the comparison with measurements.

Technical corrections

Although the English of the manuscript are generally good, there are several sentences that appear to be confusing, mostly due to displacement (or lack of use of) the comma. I suggest using it properly in order to clarify the meaning.

Page 10334, line 4: Donahue et al. (2006) does not mention 2D VBS.
Page 10339, line 2: “reproduced” should be “taken into account”.
Page 10340, line 22: “identical all” should be “identical in all”.
Page 10340, lines 24-26: the sentence is confusing, please rephrase.
Page 10341, line 22: “conversion the” should be “conversion of the”.
Page 10342, line 11: “effect” should be “affect”.
Page 10343, line 15: “not a show” should be “not show a”.
Page 10358, line 19: “campaign mean” means which period of the year(s)?
Page 10358, line 24: “analysis” should be “analyzes”.
Figure 2: what are the symbols “k”, “s”, etc.?