

In this work, Chrit et al. used the air quality model Polyphemus to describe the organic aerosol formation and properties (notably oxidation state) at a measurement site in Corsica during the winter campaign of 2014. The OA concentrations are well simulated by the model, however, their oxidation state is systematically underestimated compared to observations. They also stress the importance of an accurate characterization of emissions since they found that the volatility distribution at emissions is the prime factor that control the simulated OA concentration levels. Overall, the manuscript is well written and scientifically sound. I recommend this study for publication after taking the following comments into account.

Specific comments:

1. Page 1, lines 15-16: What is the difference between oxidation and oxygenation state of OA? If there is no difference please remove the term “oxygenation state” from this sentence

There is a difference between the oxidation and oxygenation represented by OM:OC and O:C ratios respectively. According to Gilardoni et al. 2009 and Kroll et al. 2011, the organic mass to organic carbon ratio (OM:OC) is an index of the contribution of hetero-atoms (O, H, S, N, ...) to the organic mass: chemically processed and aged particles are expected to have higher OM/OC ratio compared to freshly emitted and unprocessed aerosols. However, the oxygen to carbon ratio (O:C) indicates only the contribution of oxygen to organic molecules and the ability of carbon atoms to form bonds with oxygen. This difference is now detailed in the introduction (see reply to comment 6).

2. Page 1, lines 16-17: Why do you assume that only the multigenerational ageing of the residential heating OA can improve substantially the results? Is this the dominant sector in the area? What about the multigenerational ageing of OA from other sources?

We thought that multigenerational ageing applied to I/S-VOCs from the residential heating sector may be more efficient at producing oxidized SOA, because the primary aerosols are more oxidized than those of other sectors. However, as stressed by the reviewer, it is more accurate to stress that multigenerational ageing of all sectors does not improve the results, as shown by the simulations performed. Therefore, The sentence “The observed organic oxidation and oxygenation states are strongly under-estimated in all simulations, even when a recently developed parameterization for modeling the ageing of I/S-VOC from residential heating is used.” is replaced by “The observed organic oxidation and oxygenation states are strongly under-estimated in all simulations, even when multigenerational ageing of I/S-VOCs from all sectors is modeled.”

3. Page 2, line 7: Please replace OA with POA.
“OA” is replaced by “POA” in the revised paper.

4. Page 2, line 3: You can also add the work of Jathar et al. (2014) and Tsimpidi et al. (2017).
These works are added to this sentence in the revised paper.

5. Page 2, line 30: You can also add the work of van der Gon et al. (2015)
This work is added to the revised paper.

6. Page 4, line 4: What is the difference between highly oxidized and highly oxygenated OA?

As explained in the reply to comment 1, oxidized and oxygenated are different. The following sentences are added at the beginning of the paragraph 17, p4: “The oxidation state is represented by the organic mass to organic carbon ratio (OM:OC). According to Gilardoni et al. 2009 and Kroll et al. 2011, OM:OC is an index of the contribution of hetero-atoms (O, H, S, N, ...) to the organic mass: chemically processed and aged particles are expected to have higher OM/OC ratio compared to freshly emitted and unprocessed aerosols. The oxygenation state is represented by the oxygen to carbon ratio (O:C). It indicates the contribution of oxygen to organic molecules and the ability of carbon atoms to form bonds with oxygen.”

7. Page 4, line 8: These studies are not recent. Please add more recent studies, e.g. (Aiken et al., 2008; Tost and Pringle, 2012; Canagaratna et al., 2015; Tsimpidi et al., 2018)
These more recent studies are added to the revised paper.

8. Page 5, section 2.1: OA formation from alkanes, olefins, S/I-VOC from open biomass burning, and marine OA is missing from the model setup. Can you please add a comment on their potential importance in the examined area?

OA from alkanes and olefins are not accounted for in the model setup because their emissions are very low and highly uncertain (Roest and Shade, 2017, Hajbabaei et al. 2012). Long-chain alkanes may also be included in I/S-VOCs. I/S-VOCs from open biomass burning and fires are not accounted for in the model setup because we are dealing with the OA formation during winter, and their contribution may be low. Marine OA contribute up to only 2% of OA according to Chrit et al. 2017 in summer. Its contribution during winter time may therefore be negligible.

9. Page 5, line 13: Please change „inorganics and inorganics“ with „inorganics and organics“. “inorganics and inorganics” are replaced by “inorganics and organics” in the revised paper.

10. Page 5, line 14: Please add a reference for the algorithm.
The reference for this algorithm is added in the revised paper “... moving diameter algorithm (Jacobson, 1997)”.

11. Page 5, line 15: According to the presented results (e.g., Fig. 3), the simulation lasts until 2nd of April. “01 April” in this sentence is replaced by “02 April” in the revised paper.

12. Page 5, line 16: It would be convenient if you can state the spatial resolution used here.

The spatial resolution and the vertical resolution used here are added to the revised paper: “...in Chrit et al. (2017). The spatial resolutions used for the European and Mediterranean domains are 0.5°x0.5° and 0.125°x0.125° along longitude and latitude. 14 vertical levels are used in this study for both domains from the ground to 12 km. The heights of the cell interfaces are 0, 30, 60, 100, 150, 200, 300, 500, 750, 1000, 1500, 2400, 3500, 6000 and 12 000 m. Boundary conditions...”.

13. Page 5, lines 23-24: Please remove the sentence: “Other sea-salt.... not modelled”
This sentence is removed from the revised paper.

14. Page 5 line 34: How much is this constant factor (RRH)? And how much is the constant factor R stated later in the text?

Both R and RRH are equal to 1.5 in the reference simulation, and they vary in the sensitivity simulations, as summarized in Table 3. For clarity, P6, l6, the following sentence “Different approaches will also be used to represent the ageing of I/S-VOC, as described in section 3.” Is replaced by “Different estimations of R and R_RH will be used, as well as different approaches to represent the ageing of I/S-VOC (section 3).”

15. Page 6, section 2.2: PMF analysis results would be very useful for comparison with your model results. Are they available at the Ersa site? If so, please add this comparison.
Unfortunately, PMF analysis results are not available at Ersa.

16. Page 6, line 10: Are these coordinated the center of the model cell? Does the dimension of the model cell include the coordinates of the station mentioned above?

Yes, these are the coordinates of the center of the model cell. The dimension of the model cell does not include the coordinates of the station. Therefore, we compare the measured data at Ersa with the simulated ones at the center of the cell the closest to the station.

17. Page 6, line 10: Please define the abbreviation ACSM
ACSM abbreviation is defined in the introduction (page 2 line 23).

18. Page 6, line 12: Did you compare the measurements with the model results with size sections from 0.056 to 1.0 (as they appear in the previous page)? Please clarify.
The measurements are compared to model results between 0.01 and 1µm. This sentence is added to section 2.2.

19. Page 6, line 14: Please correct “he” with “the”
“he” is replaced by “the” in the revised paper.

20. Page 7, section 3: What about traditional VOCs? Are they subject to photochemical ageing?

The traditional VOCs considered are toluene and xylene. These VOCs are subject to photochemical ageing based on chamber measurements. They are modelled with a one-step oxidation scheme. Their contribution to OA is low as specified in the introduction p3. In the introduction p3, the sentence “In winter, when anthropogenic emissions impact the most air quality, anthropogenic emissions such as toluene and xylenes may also form SOA, although they may be less efficient than I/S-VOC (Couvidat et al., 2013a)” is replaced by “In winter, when anthropogenic emissions impact the most air quality, anthropogenic emissions such as toluene and xylenes may also form SOA, although they may be much less efficient than I/S-VOC (Couvidat et al., 2013a, Sartelet et al. 2018)”

21. Page 8, section 3.3: How do you treat OA from sources other than residential heating in this case? Do they follow the oxidation scheme described in section 3.2? In that case, can you justify why you use a different oxidation scheme especially for residential heating and not for other sources?

Please see section 3.5 where different sensitivity studies are performed using different oxidation scheme for I/SVOCs from residential heating but also for I/S-VOCs from other sources. The residential heating sector was studied separately because it makes a large part of I/S-VOCs emissions, but also because its emissions are more oxidized and oxygenated than the ones from other sources like traffic.

22. Page 8, line 11: The carbon number should decrease and oxygen number increases, please correct.

The sentence “... secondary surrogates increases and decreases respectively...” is replaced by “...secondary surrogates decreases and increases respectively ...” in the revised paper.

23. Page 8, Section 3.4: Can you provide the actual emission rates (e.g., in $Tn\ yr^{-1}$) of your OA precursor emissions (i.e., VOCs, NTVOCs, I/S – VOCs from different types of sources)?

A table of emission rates of OA precursors averaged over the Mediterranean domain and over the simulation period is added to section 3.4 of the revised paper.

OA precursor	Emission rate ($\mu g.m^{-2}.s^{-1}$)
VOCs from biogenic and anthropogenic sources	0.0314
NTVOCs	0.0062
I/S-VOCs from residential heating	0.0013
I/S-VOCs from other sources	0.0030

24. Page 9, Tables 1 and 2: Please improve the quality of the tables. For example, you should include names of surrogate species that you assign these numbers, names of sensitivity tests, and what these numbers express (i.e., emission factors, O:C, OM/OC should not be stated only in the caption but also inside the tables).

The name of primary surrogates are added between brackets to the table 1 according to their volatility coefficient and the definition of the numbers is also added to both tables 1 and 2 of the revised paper. Furthermore, a Table is added to section 3.5 to summarize the sensitivity tests.

Sensitivity study of:	Simulations to be compared
the impact of the volatility distribution of emissions	S1, S2
the impact of the ageing scheme	S3, S2
the impact of NTVOCs	S4, S2
the impact of the I/S-VOCs/POA ratio	S5, S2 and S6,S4

25. Page 10, line 11: This is not very clear. You apply a factor of 4 in the initial emission inventory, and then, on top of that you apply a factor of 4.75 to account for the NTVOOC (which are not part of your S/I-VOC). Can you please clarify and justify your hypothesis of such high additional emissions?

Yes, this is what we did, following the papers published by Ciarelli et al. (2016, 2017). In fact, it is a sensitivity test to investigate how the concentrations compare to measurements using these published emission factors.

26. Page 10, line 17: Please add in the sentence the average OM concentrations over these cities. Likewise, provide average concentrations for other mentioned areas (e.g., Ersal) later throughout the text.

The average OM concentration simulated using S4 over the mentioned cities is added to the revised paper.

27. Page 10, line 19: Why do you focus only in these two simulations?

We focus on these two simulations because they are the ones that simulate better the OM₁ concentrations.

28. Page 10, line 19: Do you mean in both simulations (instead of “in all simulations”)?

No, we mean all simulations, even though we do not show their composition.

29. Page 11, Figure 2: Please increase the font size of the fractions. Also, the fractions over the dark blue are not clear.

This figure is more readable in the revised paper.

30. Page 12, line 1: OA is already defined.

Organic aerosol is removed from this sentence in the revised paper.

31. Is this an assumption or did you actually check that you have a false rain episode in your model?

It is an assumption because we do not have rain observations at Ersa.

32. Page 15, line 5: OA is already defined.

Organic aerosol is removed from this sentence in the revised paper.

33. Page 15, Section 6: Can you comment on the importance of marine OA in your domain? Is this type of OA identified by measurements at Ersa site?

Chrit et al. 2017 examined the influence of marine OA during summer and found that the contribution of marine OA to OA concentrations over Ersa is lower than 2%. The contribution during the winter would be even lower.

34. Appendix A, table A1: The definitions are not clearly readable.

This table is more clearly readable in the revised paper.

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