We thank Anonymous Referee #4 for helpful suggestions and comments, which helped us improve the manuscript. Our point by point answers to the comments are presented below. Referee comments are in bold-italic and our replies in body text.

Referee #4 comments and suggestions:

General points:
Throughout, the reader would benefit from some more clarity or consistency as to how the two schemes are referred. The terms "NEWSOA" and "OLDSOA" could be introduced earlier to help the reader keep track of which parts of the model description relate to which results. For example, referring to the "TM5 aerosol distribution" (as in the Fig 9 caption); it is not clear whether this is TM5 with the SOA developments included or not.

We will add a sentence stating the simulation names already in Section 2.3 when describing the schemes. Furthermore, we will correct the Fig. 9 caption and Sect. 3.3.1 to show that the simulation is NEWSOA.

Similarly, it would be clearer throughout to refer to the model performance being improved or degraded relative to observations rather than increased or decreased, as there are a lot of numerical values that increase and decrease.

We will check the text and change to improve/degrade when appropriate.

Minor / specific points:
Line 10-11: Reword to clarify which value (modelled or observed) is lower than which.

We have clarified this by adding "modelled".

Line 17-18: Reword to clarify what the retrievals or observations are of (AOD?)

"of AOD" added

Line 34: correct "vegetation"

Corrected

Line 150: this section could be clarified to confirm whether the split of the 15% and 5% yields

into ELVOCs and SVOCs (I.e., 1% ELVOC and 14% SVOC for the monoterpene + OH reaction) is also based on Jokinen et al., (2015) - if not how were those values determined? If the total values and the split values are from Jokinen et al., (2015) then this citation should be added to the Table 1 caption.

For monoterpene the yields are from Jokinen et al. (2015) but for isoprene we have modified the total yield from 5% to 1% because SOA production from isoprene in our initial tests was very high. Furthermore, as explained in the text Kroll et al. (2005) report mass yields of 0.9%-3.3% which align with our choice of 1% molar yield. We added in the caption a citation to Jokinen et al. (2015) and note to check the text for explanation on isoprene yields.

Jokinen, T., Berndt, T., Makkonen, R., Kerminen, V.-M., Junninen, H., Paasonen, P., Stratmann, F., Herrmann, H., Guenther, A. B., Worsnop, D. R., Kulmala, M., Ehn, M., and Sipilä, M.: Production of extremely low volatile organic compounds from biogenic emissions: Measured yields and atmospheric implications, Proceedings of the National Academy of Sciences, 112, 7123–7128, https://doi.org/10.1073/pnas.1423977112, http://www.pnas.org/content/112/23/7123.abstract, 2015.

Kroll, J. H., Ng, N. L., Murphy, S. M., Flagan, R. C., and Seinfeld, J. H.: Secondary organic aerosol formation from isoprene photooxida- tion under high-NOx conditions, Geophysical Research Letters, 32, https://doi.org/10.1029/2005GL023637, https://agupubs.onlinelibrary. wiley.com/doi/abs/10.1029/2005GL023637, 2005.

Line 164: can you clarify in this section whether the equilibrium partitioning approach is reversible or irreversible?

The equilibrium model is irreversible, because we assume yields and therefore the produced SOA mass to represent the amount of SOA remaining in the particles in equilibrium state. Furthermore, we assume that equilibrium is reached within the model time step. To clarify this we have added the following to the text:

"The equilibrium model is assumed to be irreversible, since the yields are determined in the equilibrium state."

Line 175-176: this implies that ELVOCs cannot condense onto particles in the nucleation mode but could you add a note to confirm? If so, does that leave a gap - i.e., ELVOCs can grow particles up to 5 nm diameter, but not beyond?

This was incorrectly written. We have changed the indices so that i refers to ELVOC condensation and j to SVOC condensation. So i stands for all soluble modes and

insoluble Aitken mode as has already been explained in the text earlier. And j stands for Aitken, accumulation and coarse soluble modes as well as insoluble Aitken mode.

Line 191-192: In Riccobono et al., (2014), BioOxOrg represented the product of oxidation of monoterpenes by OH specifically so this description could be clarified slightly to explain that you are including a wider set of products (by using ELVOCs).

Agreed. The following sentence was added: „Whereas BioOxOrg represents the products from oxidation of monoterpenes by OH, our ELVOC includes a wider set of oxidation products."

Line 218+: Add a note here to clarify whether / if all the other emission sources are identical between your simulations (i.e., the only difference is OLDSOA -> NEWSOA).

Only the SOA differs between the simulations: in OLDSOA it is read in from a file and in NEWSOA it is calculated online. Both simulations have the same emissions and chemistry of monoterpene and isoprene despite the fact that the surrogate SOA emissions in OLDSOA are consistent with monoterpenes from MEGANv1. Chemical reactions with these VOCs and OH and O_3 are already present in OLDSOA but describe only the removal of VOCs while SOA production is provided as an external file. In NEWSOA using the chemical reactions describing the removal of isoprene and monoterpenes also the production of the SOA mass is calculated.

We will add a note that the emissions are the same in both simulations. As requested by another reviewer we will also add a table showing the emissions and corresponding references.

Line 250: should "overpresented" be "overrepresented"?

Corrected

Line 358: stray "fig" here

Corrected

Line 360+: it would be useful to restate in this section (and the Figure 8 caption) where these stations are, even just by reference to Table S3.

We have added a reference to the table in the first sentence:
"Figure 8 shows the annual means from the NEWSOA and OLDSOA simulations compared to the observed particle number concentrations at the stations listed in Table S3."

Line 361: correct "increas"

Corrected

Line 368-369: can you specify at which two stations the particle number concentration decreased in NEWSOA relative to OLDSOA? This may aid with the subsequent explanation.

Good suggestion, and we found out that these are two stations in Antarctica "South Pole" and "Neumayer". Our explanation for the reasons will be revised as follows:

"In general the updated SOA and new NPF parameterisation caused an increase in particle number concentrations, but at two Antarctic stations the concentrations decreased. Main reason is the change in NPF parameterisation where in OLDSOA the NPF production is calculated directly by BHN nucleation and in NEWSOA the early growth to 5 nm diameter particles is parameterised (see Sect. 2.3.5.), which are therefore less numerous."

Line 479: I couldn't find a Fig. S13

We made an error in producing the uploaded files and this figure was actually inserted in place of Fig. 13. We will add this figure in the supplement and put the correct Fig. 13 in the paper as noted in the next point.

Line 490+: this description, in terms of the differences between the MODIS and AATSR comparisons, doesn't seem to align with Figure 13 – can you check that this is the correct Figure? (this may relate to below comment about Figure 13 caption/legend)

Indeed when finalizing the upload version the figures in the supplement and manuscript were not correct. We have now put here the proper figure (see below) and the figure that was incorrectly placed has been moved to the supplement.

a)

b)

AOD difference [MODIS]

AOD difference [AATSR]
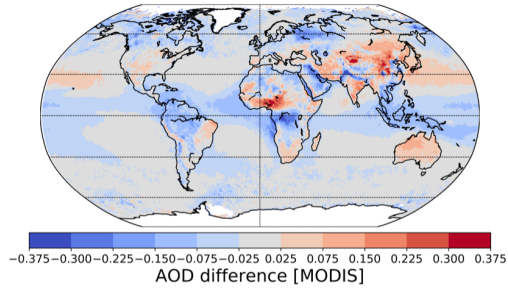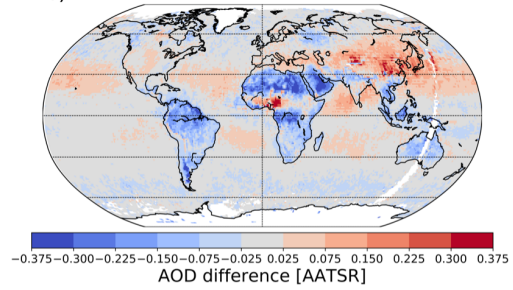
Line 507+: would it be more informative to show the seasonal cycle for each hemisphere separately?

This is a good point. We will plot the seasonal cycle for each hemisphere and revise the relevant discussion in the text.

Page 41, Figure 13: the caption suggests that this is the difference between NEWSOA and two different observations rather than NEWSOA – OLDSOA as the legend indicates, can you correct the caption / legend?

As stated above.