We thank Anonymous Referee #3 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 and our replies in body text.

Referee #3 comments and suggestions:

While none of the parameterizations are new, the more physical approach is a useful addition to aerosol modelling tools, in particular for M7 and derivatives thereof. The article is in general well structured although sometimes it is a bit unclear when the authors describe the results from the old or the new version. A somewhat more problematic issue is that the authors often conclude that this is caused by emission changes and this is due to secondary aerosol formation. While it may be obvious for most cases I think the authors should do a sensitivity test that only show the impact of the emission changes. The assumptions on yields is another parameter that should be discussed although the emission test will probably work as a first approximation on understanding yields.

To analyse the effect of emissions on SOA production we have performed an additional sensitivity simulation similar to the OLDSOA simulation but using surrogate SOA emissions with the MEGAN v2 monoterpene emissions that are used for the NEWSOA simulation. The main results will be discussed in the paper.

Sporre et al. (2020) analyzed the behavior of the model with our NEWSOA scheme using a set of sensitivity experiments. By scaling the SVOC and ELVOC yields up and down by 50%, the resulting SOA mass was increased / decreased by almost 50% (Sporre et al., 2020, Fig. 6). Products from isoprene and monoterpenes contributed about 80% and 20% to the SOA mass (Sporre et al., 2020, Fig. 7). The response of the number concentration and number size distributions was rather non-linear since it very much depends on the competition between new particle formation and growth of existing particles. The balance between the two processes was changed by adapting the SVOC and ELVOC yields.

We will add the text above also to Sect. 2.3.2 the manuscript.

Sporre, M. K., Blichner, S. M., Schrödner, R., Karset, I. H. H., Berntsen, T. K., van Noije, T., Bergman, T., O'Donnell, D., and Makkonen, R.: Large difference in aerosol radiative effects from BVOC-SOA treatment in three Earth system models, Atmos. Chem. Phys., 20, 8953–8973, https://doi.org/10.5194/acp-20-8953-2020, 2020.

It may beyond the scope of the article and or due to missing capabilities of the model I am wondering if the authors also considered comparing the Ångstrøm component in particular in light of the focus of aerosol size changes from the new parameterisaton.

We find this an interesting idea, but it is beyond the scope of this paper.

Minor / technical issues.

The introduction splits the organic compounds between ELVOC, LVOC and SVOC, Later on it only use ELVOC and SVOC. My usual interpreation is that SVOC s reserved for thos compounds that can commonly re-evaporate. As I understand none of the VOCs described in this work is re- evaporating. I think this should be stated explicitly if the authors continue to use SVOC.

That is correct, we assume that all of the SVOC from the oxidation will reach equilibrium within the time step of the model. We will add the following sentence in Section 2.3.2:

“In this work we assume that SVOC does not re-evaporate.”

And in Section 2.3.3 describing the partitioning to particles:

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

Abstract line 11. “clearly lower as is” new versus old ?

This is indeed a bit unclear. Here we compare to the observations and we have rephrased the sentence as:

“However, the modelled concentrations of formed particles are clearly lower than in observations as is the subsequent growth to larger sizes.”

Line 32: Missing in first volatility word

Corrected.

Line 34. Numbers inside “vegetation”

Corrected

Line 39: Two “rathers” are rather too much. Can one of them e.g. the first be replaced by a number / fractional estimate.

Indeed, we have rephrased the sentence; it now reads:

“Emissions of BVOCs constitute roughly 90% of total VOC emissions, but due to complex chemistry the actual processes and the total amount of SOA formation is rather uncertain (Tsigaridis et al. 2014).”

Line 76: “between” → “divided between” ?

Corrected as suggested.

Line 93, but this has also consequences for the remaining manuscript. Does “trace gases “include OH? If not the description of chemistry must include information on the OH concentration used in the model.

If OH is part of the chemistry. Will the SOA chemistry have noticeable impact on the OH concentration?

Both simulations have the same emissions and chemistry of monoterpene and isoprene despite the fact that the surrogate SOA emissions 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 in OLDSOA while SOA production is provided separately. In NEWSOA using the chemical reactions describing the removal of isoprene and monoterpenes also the production of the SOA mass is calculated. The change in aerosol fields will cause some change in gas-phase concentrations of oxidants, but there is no direct influence in the reactions themselves. We have explored how big the impact is, and the change in annual global mean of OH concentrations in all model levels is well below 1%. At the surface there are regions with annual mean change of 1–2% while higher up it changes to 5% at maximum. In our view the change is relatively small.

Figure 1: No evaporation of SVOC?

As in Jokinen et al. (2015) we assumed that the particles are in equilibrium with the gas phase and the yields are calculated to take into account only the mass remaining in the particles.

Line 121. Are isoprene and monoterpene emitted at ground level or follow the height distribution of the oldSOA.

They are emitted at ground level. We have added the following on line 143 (after declaring the emission strength and reference to Fig. 2):

“It should be noted that the gases are emitted at ground level.”

Line 143: Unclear. "geographical distribution ..." As far as I understand the model reads regridded monthly mean emissions from a specific MEGAN set-up. TM5 does not do anything to the emissions themselves e.g. related to meteorological fields? so it is just how the emission look like from the model (except for the diurnal cycle)

That is correct. We have changed this to "; the geographical distribution of emissions is shown in Fig. 2a,c"

Line 156. Choice of yield looks a bit arbitrary. Tuning ? A sensitivity test might be interesting although information from "initial tests" may be useful. Are the production and results linear with yields?

Yes, SOA mass in close approximation scales as the yields. Sporre et al. (2020) analyzed the behavior of the model with our NEWSOA scheme using a set of sensitivity experiments. By scaling the SVOC and ELVOC yields up and down by 50%, the resulting SOA mass was increased / decreased by almost 50% (Sporre et al., 2020, Fig. 6).

Regarding the choice of yields, for monoterpene the yields are directly 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.

Sporre, M. K., Blichner, S. M., Schrödner, R., Karset, I. H. H., Berntsen, T. K., van Noije, T., Bergman, T., O'Donnell, D., and Makkonen, R.: Large difference in aerosol radiative effects from BVOC-SOA treatment in three Earth system models, Atmos. Chem. Phys., 20, 8953–8973, https://doi.org/10.5194/acp-20-8953-2020, 2020.

Line 164. Do you think these assumptions are important for the results.

We think that when NEWSOA is compared to OLDSOA, where SOA is put in one mode as mass only, the assumptions are important in understanding the differences.

Line 172: Sometimes the different properties of hygroscopicity is taken into account by a employing a bulk coefficient to the condensation term with an interval of [0 1]

This is not relevant here, or just too uncertain ?

We assume that the applied yields give the amount of SOA which will remain on the particles following Jokinen et al. (2015). Therefore, all of the SOA mass is distributed to the particles by organic mass or by surface area of the existing particles and condensation is not solved explicitly. So in our view this is not relevant here.

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.

Line 214. Refer back to section for readability

We do not think this will improve the readability since it refers to all of this section until this point.

Line 220. Any reference to the entire model system (van Noije et al ?) Given a reference which SOA parameterisation is used in that work

The EC-Earth3-AerChem version is documented in van Noije et al. (2021), and uses the new SOA and NPF parameterisation as explained there.

van Noije, T., Bergman, T., Le Sager, P., O'Donnell, D., Makkonen, R., Gonçalves-Ageitos, M., Döscher, R., Fladrich, U., von Hardenberg, J., Keskinen, J.-P., Korhonen, H., Laakso, A., Myriokefalitakis, S., Ollinaho, P., Pérez García-Pando, C., Reerink, T., Schrödner, R., Wyser, K., and Yang, S.: EC-Earth3-AerChem, a global climate model with interactive aerosols and atmospheric chemistry participating in CMIP6, Geosci. Model Dev. Discuss. [preprint], https://doi.org/10.5194/gmd-2020-413, in review, 2020.

Line 225. Is the resolution the same as in the referenced version of the model.

TM5-MP can be run in 1x1 degree and 3x2 degree resolutions. In EC-Earth the atmospheric component IFS is running on T255L91 grid and is coupled with TM5-MP running at 3x2 degree resolution. This is the same as in the model version documented in van Noije et al. (2021).

van Noije, T., Bergman, T., Le Sager, P., O'Donnell, D., Makkonen, R., Gonçalves-Ageitos, M., Döscher, R., Fladrich, U., von Hardenberg, J., Keskinen, J.-P., Korhonen, H., Laakso, A., Myriokefalitakis, S., Ollinaho, P., Pérez García-Pando, C.,

Reerink, T., Schrödner, R., Wyser, K., and Yang, S.: EC-Earth3-AerChem, a global climate model with interactive aerosols and atmospheric chemistry participating in CMIP6, Geosci. Model Dev. Discuss. [preprint], https://doi.org/10.5194/gmd-2020-413, in review, 2020.

Line 250. overpresented is likely not the right word

We have changed this to overrepresented.

Line 325: Very low dry deposition. This should be the same for sulphate and elemental carbon.?

The fraction of particle removal by dry deposition varies between 1.1% to 2.8 % for BC (2.2%), OA (1.7%), SOA (1.1%), and SO4 (2.8%). It is shown in Section 4.1.4 of Tsigaridis et al. (2014) that models that employ M7 show very low dry deposition rates for organic aerosol. Furthermore, the SOA dry deposition fraction varies between 1.1% in the NEWSOA and 1.9% in the OLDSOA simulation, which seems to suggest that production aloft in NEWSOA increases the importance of wet deposition. For other compounds there is almost no change.

Line 329 -335. Unclear description, can be made shorter. Can be describe it as a combination of increase (production aloft) and decrease (aitken vs accumulation) ?

We will rephrase this:

“In NEWSOA the SOA mass is distributed into the aerosol size distribution more evenly than in OLDSOA. This means that mass in accumulation mode is decreased while Aitken mode mass is increased which could suggest lower lifetime (see Schutgens and Stier (2014) for modewise lifetimes in M7). However, in NEWSOA the SOA production aloft is clearly higher meaning that the deposition takes more time leading to a longer lifetime. “

Schutgens, N. A. J. and Stier, P.: A pathway analysis of global aerosol processes, Atmospheric Chemistry and Physics, 14, 11 657–11 686, https://doi.org/10.5194/acp-14-11657-2014, https://www.atmos-chem-phys.net/14/11657/2014/, 2014.

Line 346. Ocean production: Any Ricco NPF above the boundary layer (outflow from continents?)

There is some production over oceans but it is very low. This is mainly because the model does not include any monoterpene or isoprene emissions from the oceans and all of the NPF over oceans depends on the continental outflow of these gases. Marine monoterpene and isoprene emissions are low and uncertain as noted in the manuscript.

Line 354: No observation of NPF over ocean?

We did not use any observations over oceans. We have made it clearer by adding "modelled" before NPF rate.

Line 359 Solitary "fig" word

Removed.

Line 461 Increas → e

Corrected.

Section 3.4.2 Is the reduction in PM2.5 mainly due to reduction of SOA mass or are there other non- linear effects?

Table 3 seems to be a bit unclear, but it lists only the OA concentrations as stated in the caption, we will clarify the table in the revised manuscript. Similarly, in the main text we only discuss organic mass in PM2.5 not the total PM2.5. Nevertheless, aerosol dynamics are inherently non-linear, and here the scheme in OLDSOA is unphysical since it distributes all of the produced SOA mass into Aitken mode in the model. This will cause the Aitken mode to grow rapidly into the accumulation mode with longer lifetimes, while in NEWSOA the mass is more evenly distributed with less mass in the accumulation mode. This is more prominent for the IMPROVE stations, but it will also affect the organic mass in PM2.5 in Europe.

Line 501-506: Very unclear.

We will rephrase this as follows (note that due to another reviewer's suggestion the seasonal cycle will be shown separately for both hemispheres):

"Figure 14 shows the comparison of AOD measured by AERONET and modelled AOD in NEWSOA (a), and OLDSOA (b). Fig. 14c shows the hemispheric seasonal cycles of AOD in AERONET observations, in NEWSOA and OLDSOA. Additionally, Fig. 14d presents a map showing the station locations and their regional groupings for the statistics in Table 4.

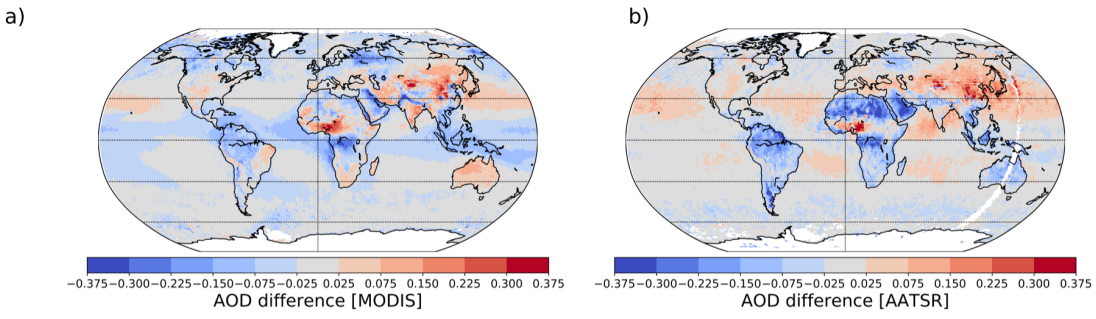
In general the simulations show similar behaviour where they overestimate the low AOD and underestimate the high AOD observations. However, in NEWSOA the absolute bias across all stations improves in NEWSOA to 0.184 with a low bias of 0.003 from 0.176 in OLDSOA. Furthermore, the correlation coefficient (R; see last row of Table 4 for the values across stations) increases by 0.009. Visually both simulations show a very similar deviation from the observations."

Line 514-520: Are the results from satellite and Aeronet consitent. I do not expectant large analysis here but nice to point out.?

The AERONET map in Fig. 15b and satellite map figures — the manuscript included a wrong Fig. 13, the correct one is included below and will be changed in the paper — and the correct satellite comparison maps show similar changes. In the Amazon both satellites show a decrease in AOD as do the AERONET sites. Europe, the US, and Southeast Asia show similar behaviour for both satellite instruments and AERONET. For Australia, the AERONET comparison shows a similar behavior as the MODIS instrument. Summarizing, the AERONET comparison is consistent in the direction of the change with satellite instruments in most regions.

We will add a discussion along these lines to the paper:

"A comparison of biases in AERONET (Fig. 15b) and satellite retrievals (Fig. 13) —note that the color scales do not match — shows qualitative agreements between the two sets of observations in most locations and times . Over the Amazon and Southeast Asia both satellites show a decrease in AOD as do the AERONET sites. Over Europe and the US both satellite instruments and AERONET show small or no change. It is noteworthy that in Australia where satellite retrievals do not agree, the AERONET comparison shows a similar behavior with the MODIS instrument."



Line 524. The magnitude of AOD ? → The increase of AOD varies by an order of magnitude of 0.003 to 0.03

Changed as suggested.