

We thank Anonymous Referee #1 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 #1 comments:

There are a number of figures and tables in the supplement which are never referred to in the paper. In addition, Fig 13 in the paper does not show the figure being described in the text.

We will add references to supplementary figures and tables. The wrong Fig. 13 will be exchanged.

In the following I detail changes/explanations that are needed:

Please explain how you calculate OH and O₃. Are these read-in?

No, the model has interactive chemistry. In the relevant parts of the atmosphere, OH and O₃ are calculated online. We have added the following to the Section 2.3.2:

“The O₃ and OH oxidant fields are calculated online in the chemistry code.”

Line 34: odd addition of numbers in “vegetation”

Corrected.

Line 46: please add references to models producing up to 121 Tg/yr SOA

We have added the following to the text:

“IMAGES (Müller et al. 2009; Stavrou et al 2009; Ceulemans et al. 2012) and IMPACT (Lin et al. 2012) models in Tsigaridis et al. (2014)”

Line 47: While VBS schemes are “more intricate” they are essentially parameterizations. You need to state this.

Changed “schemes” into “parameterisations” in the text.

Line 52: “subsequently” is a strange word here. “In this theory”?

Changed as suggested.

Line 60: remove first “of”

Done.

Line 69: add Wang and Penner, 2009:

Wang, M. and J. E. Penner, 2009: Aerosol indirect forcing in a global model with particle nucleation, *Atmos. Chem. Phys.*, 9, 239-260, www.atmos-chem-phys.net/9/239/2009/.

After all, this paper was published almost a full year before Merikanto et al., 2009

Thank you for the suggestion. We have added the citation.

Lines 101-103: You do not follow the mixing of soluble and insoluble particles? This seems highly unrealistic, since, for example SO₄ would almost certainly condense onto insoluble particles. Are simulating sulfate condensation only on the soluble modes? If on both types of particles, how do you separate into the modes after condensation, since sulfate and dust forming on an Accumulation particle, could, after separation, form 2 Aitken mode particles or 1 Aitken mode and 1 accumulation mode, depending on the size of the original particle after condensation.

We use the standard M7 formulation (Vignati et al. 2004), where sulfuric acid condenses onto soluble and insoluble particles. Ageing of insoluble particles takes place by both condensation of sulfuric acid and coagulation. The fate of particles formed by coagulation depends on the modes of the coagulating particles, as described by simple rules. Particles in the insoluble accumulation mode can only coagulate with soluble Aitken-mode particles, resulting in a transfer to the soluble accumulation mode. The sulfate condensation onto insoluble modes will lead to transfer of particles into soluble modes. The fraction of particles that will be transferred is calculated as the number of particles for which a layer of sulfate molecules with a thickness of one molecule can be created.

Vignati, E., Wilson, J., and Stier, P.: M7: An efficient size-resolved aerosol microphysics module for large-scale aerosol transport models, *J. Geophys. Res.*, 109, D22 202, 10.1029/2003JD004485, 2004.

Line 110: delete “more” in “A more detailed description”, since no description is given.

Done.

Figure 1: Is there some reason that the reaction of isoprene and monoterpenes with NO₃ is omitted?

We started out to reproduce the method used by Jokinen et al. (2015), which only includes yields for reactions with OH and O₃. We had a discussion to include oxidation by NO₃ but due to time constraints of CMIP6 we were unable to implement this reaction. The chemistry does include NO₃ which will allow this reaction to be added in the future.

From the figure the dotted line for the insoluble mode is only green for the Aitken model. So, it seems ELVOC and SVOC only condense on the insoluble Aitken mode. Why is this?

That is because in M7 the insoluble accumulation and coarse modes only include a dust tracer.

Line 139-140: does the diurnal emission variation for monoterpene at least depend on the local temperature variation? Surely the model has this variable available. Please explain why it should not take this into account, if not and give the max/min dependence of the emission.

As explained in the text, we normalize the daily mean emission to produce the emitted mass from an inventory. In order to use temperature we would need to simulate a full day first and then normalize the emission accordingly, which is not feasible to do.

We have changed

”These variables are not available in the model and therefore” to:

“Since we rely on emission inventories the daily emissions need to be normalized which makes the use of drought or temperature not feasible and therefore...”

141: This reads like the monthly mean isoprene emission is 572.3 Tg/yr, but I know that is the annual emissions rate. I think you mean you employ a monthly varying emission with an annual mean of...

We will rephrase this as follows:

“In this work we employ monthly varying isoprene and monoterpene emissions (with annual emissions of 572.3 Tg yr⁻¹ and 95.5 Tg yr⁻¹, respectively) from an inventory derived from MEGANv2.1...”

175-176: Why wouldn't SVOC partition onto Accumulation and coarse modes if OA is present? (for example, by ELVOC condensation on these modes)?

This was not clearly written. We have rephrased the text as follows:

“soluble Aitken, accumulation and coarse modes and insoluble Aitken mode.”

215: I gather “all other biogenic emissions” really just refers to isoprene? Why not say this?

TM5 is a chemistry transport model and it also includes other biogenic VOCs in addition to isoprene and monoterpenes although these do not directly affect the aerosol phase.

218-225: I think a paper should give sufficient information so that the reader does not need to read other papers in order to understand. Why not add a table with all annual emissions and references? After all, the emissions are one basic element affecting the evaluation

We will add a table listing the annual emissions for aerosol and relevant precursors with their references.

230: I don't think you mean SOA “on” the surface. Perhaps “in the boundary layer”? or in the first level of the model.

Here we refer to the observations that we are using. We will rephrase to “in the boundary layer”.

246: The factor of 1.6 should be referenced, unless there is no reference. If basically unknown, then say so.

This is documented in the paper by van Noije et al. (2021). We have included a reference to this paper.

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.

247: do not use “on the surface”

We will rephrase it to “in the boundary layer”.

250: change “overpresented” to “overrepresented”

Done.

313-314: here you first mention that there is no SOA formation from isoprene in OLDSOA. This should be stated earlier when talking about the sources from MEGAN (new and old).

It is mentioned already on lines 144 and 214 that in OLDSOA SOA formation is based on monoterpene emissions from MEGANv1. We have added a sentence on both locations to point out that isoprene does not contribute to the formation of SOA.:

“Isoprene does not contribute to the SOA formation in the old scheme.”

326-327: can you speculate why the wet deposition fraction is higher in your model? Perhaps the vertical distribution places SOA higher in the atmosphere than in the other models?

The fraction of wet deposition of SOA does change from 98.1% in OLDSOA to 98.9% in NEWSOA, but it is high in both cases. So it would seem that the wet deposition is somewhat increased due to additional production aloft. However, high wet deposition is also found in other models using the M7 aerosol module (Tsigaridis et al. 2014), which would indicate that it is an M7 module feature and not so much related to SOA, although the new formulation increases it further. Furthermore, many of the AeroCom models simulate SOA as emitted particles similarly to OLDSOA, which indicates that the difference from OLDSOA to AEROCOM models does not result from the vertical distribution.

Figure 7: Are (c) and (d) concentrations in the surface layer?

Yes, we will add this information in the caption accordingly.

341-342: change to “using the additional particle formation” - I had to look back to see if you also include the Vehkamki scheme for sulfate in NEWSOA

Done.

352-357: please explain why southern ocean nucleation mode particle numbers are higher in OLDSOA than NEWSOA. After all, both these schemes include binary homogeneous nucleation

The two simulations handle the newly produced particles differently; in NEWSOA nucleation mode particles growth to 5nm is parameterised based on sulfuric acid and/or

ELVOC while in OLDSOA no growth is assumed. We have added an explanation into the text:

“Furthermore, OLDSOA shows higher concentrations (and NPF) over Antarctica. The reason for this is the different handling of nucleation mode particles in the two simulations; in NEWSOA the initial particle growth to 5 nm in diameter is parameterized while in OLDSOA no growth of particles is assumed (see Sect. 2.3.5 for details).”

361: missing e in “increase”

Corrected.

365: “of of”

Corrected.

369-370: I do not understand why particle formation in NEWSOA isn't always larger than in OLDSOA, since the former includes RiCCO AND binary sulfate formation

In NEWSOA the new particle formation is calculated at 5 nm while in OLDSOA the new particle formation from Vehkamäki et al. (2002) is used as is. That parameterisation can produce a high number of particles with diameters around 1 nm.

371-373: what is meant by “compensated”? this explanation needs some expansion – it is hard to follow.

Due to the suggestion from another reviewer we have looked into it more closely and both stations with higher concentrations in OLDSOA are located in Antarctica. We will rephrase the last paragraph 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, which are therefore less numerous.”

384: I think you do not mean observed N2O, since this is a gas. Also, it appears CCN particles do not reach 5000/cm3: the scale only goes to 3000/cm3, but Vavill goes to 5000/cm3. BTW if you are referring to the top 2 lines of Fig 9b, there is no scale, so one cannot tell if the observation goes to 5000/cm3.

This was incorrectly written as we were using both N₂₀ and CN (condensation nuclei) interchangeably. However, it is not N₂₀ but N₂₀ and refers to particles with diameters larger than 20 nm which is noted as CN in the figure. We will change the notation in the figure.

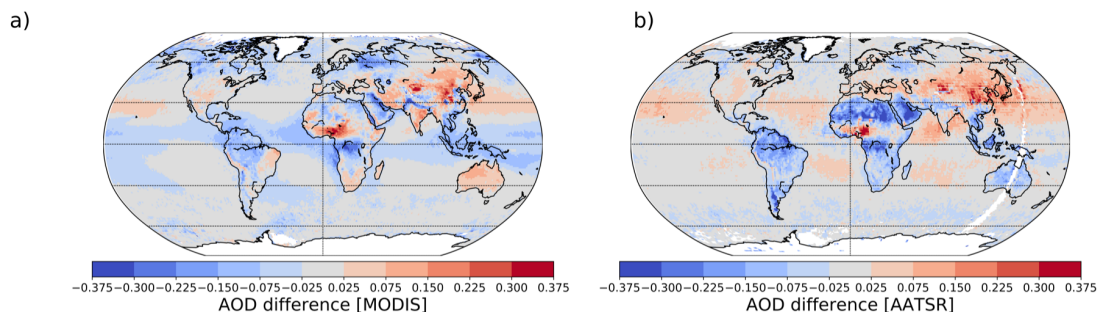
And for the particle concentrations, in the text we refer to CN, which reaches almost 5000 cm⁻³ at Birkenes, Norway (middle panel of Fig 9b, left (red) axis). The figures include both CN and CCN using different color lines, here CN comprises the particles larger than 20 nm (or 3 nm in Vavihill) in diameter and CCN are the particles larger than 70 nm in diameter. In the line plots of the figure the right hand side axis refers to CCN and left hand axis to CN. This is noted by the matching colors on “CN” and “CCN” in the axis label. We will change these to N₂₀ and N₁₀₀ for clarity. As requested we will add a colorbar for the upper panels. We will also revise the main text for more clarity and add text in the caption to describe the figure properly.

Figure 11: in (e) and (f), I assume the 2 NMB's and RMSE's are for PM_{2.5} and PM₁₀, but they are not labeled or in the caption. Also, what are the 2 numbers after R: in the upper 2 panels?

We have removed these since they are not referred to in the text.

Figure 13: the label on the graph states that both a and b are the difference between NEWSOA and OLDSOA, whereas the caption states the correct difference.

This was an error in the production stage: somehow another figure was exchanged with the actual paper figure. We will insert the correct figure here (shown below).



472: Where is OLDSOA compared with MODIS and AATSR? The figure caption of figure 13 states that this figure is only NEWSOA. Also, it appears that both panels in fig 13 have most of the tropics and subtropics overestimated, so how do you estimate an underestimated AOD? Much of the paragraph after this sentence in line 472 cannot be understood based on fig. 13.

See answer to comment above.

So, when you finally reference the supplemental figures, I see that the supplement Fig S10 and S11 compares OLDSOA and NEWSOA with MODIS and AATSR. I find it strange that this is perhaps the first supplemental figure (or table) that is meant to be referenced. Why have figures and tables in a supplement if they are not referenced?

BTW, I would at least include an annual average difference of OLDSOA and NEWSOA with MODIS and AATSR in the main text. As it stands, the main result of the data shown in the current Fig 13 was already discussed.

As stated earlier we will insert the correct figure in the main text and move the current figure to the supplement as noted earlier. The annual means have been noted in the text on lines 473 (MODIS) and 491 (AATSR).

552: add a comma after “events”

Done.

553: explain what the “numerical issues” are. This is the first mention of this fact. Also, I would add “perhaps” since you do not actually analyze the reason for low CCN sized particles (by comparison with observations, for example)

It is mentioned already in Section 3.3.1 on particle concentrations at selected sites on line 402, but to be clear we have added there some explicit issues:

“(e.g. errors from operator splitting, unrealistic size distributions due to mode merging)”

567: The change in AOD is ultimately from the change in particle number, but must actually be due to the change in size distribution, I think. This sentence should be expanded to remind the reader of the processes leading to the change in AOD

Thank you for a good suggestion. We will rephrase with the following:

“Therefore, the change in AOD results mainly from the different shape of the size distribution. Because the condensation of SOA in NEWSOA is physically described the particle concentrations in the optically important size range are higher than in OLDSOA.”

