Replies to the Anonymous Referee 2

We thank the referee for the valuable comments which helped us to improve the manuscript. Please find below our responses (in black) after the referee comments (in blue). Changes in the revised manuscript are written in *italics.*

General Comments This is a review of "On the formation of biogenic secondary organic aerosol in chemical transport models: an evaluation of the WRF-CHIMERE (v2020r2) model with a focus over the Finnish boreal forest" by Ciarelli et al., submitted to GMD. This paper investigates predictions from WRF-CHIMERE versus measurements at a boreal forest site (SMEAR II in Hyytiala Finland). The predictions compare well with most meteorological data (temperature, wind speed, RH, and wind direction) and gas phase species (monoterpenes, O3, NOx), but struggles with isoprene concentrations and precipitation events. The authors focus on the model's ability to predict biogenic secondary organic aerosol (BSOA) formation. To this end, the authors run a series of sensitivity simulations, altering the OH reaction rates and the isoprene emission rates. This manuscript could be improved by more clearly detailing which simulation is being discussed at any given time, and the goal and conclusions of running these different simulations (see below specific comment about this). This paper should be published after the below specific comments are addressed, and should be of interest to readers of GMD.

We thank the referee for her/his comment on our manuscript. Our specific replies follow below.

Technical comments Section 2.2:

Section 2.2: can you add an explanation of *why* these different simulations are performed? I'm confused on the role of these simulations and what is discussed where in the following sections. The next time these simulations are mentioned as defined here is not until section 4.3/ line 303.

We thank the reviewer for this comment. Aging of biogenic aerosols have been evaluate in previous modeling application at European scale (Bergström et al., 2012; Cholakian et al., 2017; Zhang et al., 2013). However, very few of these studies have investigated the different effects of using varying biogenic aging scheme in an environment that is largely affected by biogenic emissions and by combining parallel measurements of biogenic precursors, meteorological parameters, and aerosol size distribution. We therefore performed an evaluation of the difference aging schemes, and underlying biogenic emission inventories, as currently available in literature, and by using the latest measurement data. Specifically, our evaluation study is built upon the increasingly comprehensive data sets available at supersite measurements stations like, for example, the Station for Measuring Ecosystem–Atmosphere Relations (SMEAR-II) located in the Finnish Boreal Forest, which provides a platform to evaluate model results to a great level of details thanks to parallel state-of-the-art measurements of a vast array of atmospheric compounds. As model simulations are growing in complexity, we believe that model evaluation studies are vital to support the modeling communities in reducing the sources of uncertainties in current biogenic secondary organic aerosols schemes and in the development of new numerical approaches, hopefully resulting in a better predictions of future climate scenarios.

We add the following paragraph at page 6, line 158 of the revised manuscript to better clarify the motivation for such analysis.

Aging of biogenic aerosol have been tested in previous modeling application at European scale (Bergström et al., 2012; Cholakian et al., 2017; Zhang et al., 2013). However, very few of these studies have investigated the effects and impacts of using difference biogenic aging scheme in an environment that is largely affected by biogenic emissions and by combining parallel state-of-the-art measurements of a vast array of atmospheric

compounds. In this study we performed a comprehensive evaluation of the difference aging schemes as currently available from the literature.

Line 234: can you quantify "a slight underestimation" in the text (from Table 3)? Can you speculate why this is occurring? It looks like the model predicts a lower nighttime temperature on almost every night except a handful, and actually does best during the heat wave, while capturing the daytime highs?

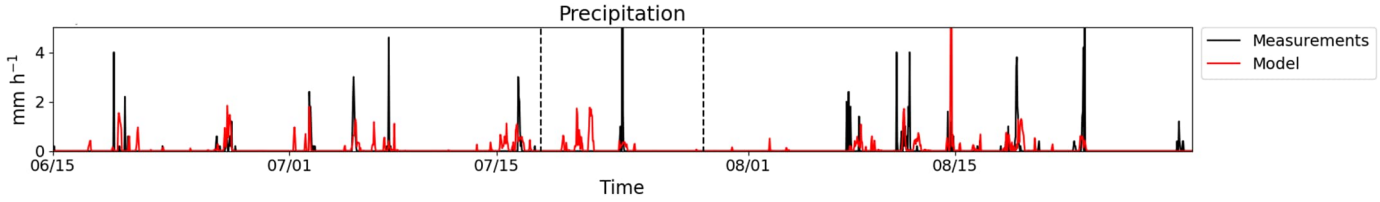
We thank the referee for this comment. Among the model resolution adopted here (about 10 km for the nested grid) which might influence the overall performance of the meteorological model, another important parameter that might affect model performance over large forest areas is the canopy effect. While the underlying emission model does account for the canopy effect, the current version of the CHIMERE model used in this study does not include any canopy effect. Recently, the canopy effect on vertical diffusion, wind speed, and shortwave radiation in the model was implemented and tested over the Landes pine forest in southwestern France (Cholakian et al., 2022) which we are planning to test also on the domain presented here.

We reported the value from Table 3 at page 9 line 253 of the revised manuscript as below:

The model was able to reproduce such a temporal trend with a slight underestimation (-0.7°) occurring mainly during the nighttime periods (Figure 4).

Line 236: it looks like the model misses all or almost all of the rain events, even the relatively large one during the heat wave, do you know why? Are they short-lived, or low total volume (i.e. do they have to last a specific amount of time or have a minimum volume to be captured)?

We thank the reviewer for her/his comments. We have additionally checked the rain intercomparison analysis. For the current analysis we were using modeled rain data as available from CHIMERE output in kg m^{-2} h^{-1}, whereas the proper comparison be in mm h^{-1} (which is written in the deposition files of CHIMERE). We have now reperformed the analysis, which revealed that the signal of the small rain event is actually captured by the model (Figure 4 below, scale has been adapted to facilitate the comprehension of the panel). However, the largest events are still missed (Figure 1, below), suggesting that the model might have difficulties to accurately reproduced short-lived events probably induced by local weather systems and the orographic processes specific of the site. We updated Figure 4 in the revised manuscript accordingly.



Line 242: an r value is provided for the wind speed, is it possible to also provide this for wind direction (on line 240 probably, or table 3)?

We agree with the reviewer, and we have now included the r value for the wind direction (r = 0.5) at page 10, line 258 of the revised manuscript as below:

The analysis of the wind direction fields indicated that they were satisfactorily reproduced by the model, with the southern westerly (SW) sector being the most predominant wind direction during the summer period (r = 0.5).

Line 250: what is causing the relatively high isoprene emissions in the "localized" area?

We believed this is likely caused by the underlying emissions factor associated with the vegetation data used with the MEGAN model, and more detailed analysis are planned to probe into those highly localized emissions in those areas (i.e., by using domain-specific land use).

Figure 7: recommend making the percentages larger and bold, the text is small relative to the size of the wedges and hard to read.

We modified Figure 7 as suggested by the referee in the revised manuscript.

Line 270: can you add a statistic to the text to quantify how much isoprene is "largely overestimated"? Either one of the values from Table 4 or something like number of days overestimated, average % overestimation, etc?

We agree with the reviewer, and we have quantified the overestimate in isoprene air concentration based on the Timeseries presented in Figure 8 of the manuscript. The ratio between the modeled and observed isoprene concentration varies from 4 to 8, with few isolated peaks exceeding a factor of 10. We have included this information at page 11, line 290 of the revised manuscript as below:

The ratio between the modeled and observed isoprene air concentration varies from 4 to 8, with few isolated peaks exceeding a factor of 10.

Figure 10: I don't think this figure adds much, suggest removing/moving to SI or combining with Figure 9

The referee is right. However, in the revised manuscript we have revised Figure 10 to also include the additional model evaluation for OC measurements as available from the EBAS datasets (as requested by referee nr. 1). For this reason, we prefer to keep the Figure 10.

Line 296: this is the first time ASOA is mentioned in the body of the manuscript (not just the introduction), so suggest defining it again here

We have re-defined the definition of ASOA (anthropogenic secondary organic aerosol) also at this occurrence in the revised manuscript.

Figure 11: why is there a hot spot in POA over Turku but not ASOA?

We thank the reviewer for this question. Turku is located on a coast side inside the domain. These sites are likely more challenging to resolve, at the current resolution, compared to other regions giving that the model cell grid needs to be resolved for both the water bodies and the physical terrain. Even though the underlying emissions inventories might lack several anthropogenic precursors which might not be fully resolve at the current resolution, the local meteorological condition can also highly influence the accumulation, production, and removal processes of secondary species. Indeed, higher ASOA concentration are visible over the area of Helsinki, and additional analysis would be needed to understand the differences in the formation of anthropogenic secondary organics aerosol (ASOA) at these two sites (at least from a modeling perspective).

Lines 294-305 & Figure 11: the discussion of ASOA feels misplaced since the discussion is focused on BVOC and BSOA up until here. Suggest adding some details to the methods section, or removing the discussion of ASOA.

We agree with the reviewer The treatment of ASOA in model is now discussed in detail in the revised manuscript in the methods section (as also asked by reviewer 1). The title of Section 2.2 has been changed as below:

2.2 OA scheme

Figure 12: similar to figure 7, suggest making text on the wedges larger

We modified Figure 12 as suggested by the referee in the revised manuscript.

Section 4.4: I think this section would follow more logically if it was before current section 4.3?

We agree with the reviewer, and we have now inverted the order of section 4.3 and 4.4 in the revised manuscript.

Figure 14: I think this figure also might be removed or put in the SI.

We agree with the reviewer, and we removed Figure 14 from the manuscript.

Line 360-361: can you quantify the diurnal O3 agreement and overestimation of NOx in the text (from table S4)?

We added the values from Table S4 at page 14, line 413 of the revised manuscript as below:

As reported in the Figure 15, the model is capable to reproduce the diurnal variation and absolute values (ppb) of O_3 very well (mean bias of -0.1 ppb and 0.3 ppb for O_3 and NO_x, respectively, Table 4).

Line 387: I assume "a.s.l." means "above sea level"? Suggest defining, and I'm not familiar enough to know if it's typically capitalized?

We agree with the reviewer and the have defined the acronym in the revisited manuscript.

Grammatical comments

The manuscript is well written, although several minor grammatical errors exist throughout. While they do not impede the reader's understanding, the entire manuscript should be checked over before publication. Specific instances listed below, although please note I didn't not write them all down.

Line 23: "heat waves episodes"—waves should be singular

Corrected.

Line 73: "ration" should be "ratio"

Corrected.

Line 88&99: "where" should be "were"

Corrected.

Line 103 &140: "oxidization" should be "oxidation"

Corrected.

Line 194: "measurers" should be "measures"

Corrected.

Line 256: "there" should be "they"

Corrected.

Line 253: "measurement" should be plural

Corrected.

Line 257: “relatively” should be “relative”

Corrected.

Line 261: “differently” should be “different”

Corrected.

Line 262: “document” should be “documented”

Corrected.

Line 279: “instrumentation” should be “instrument” or could be removed entirely\

We removed the work.

Line 295: “it is noticed” should be “is noticed

Corrected.

Line 296: should “San Petersburg” be “Saint Petersburg”?

Corrected.

Line 314: “identify” should be “identified”

Corrected.

Line 320: “underestimate in the accumulation” should be “underestimate the accumulation”

Corrected.

Line 345: “increased” should be “increase”

Corrected.

Line 348: “over few regions” should be “over a few regions”

Corrected.

Line 349: “in the order” should be “on the order”

Corrected.

Line 350: “reacts” should be “react

Corrected.

Line 354: “to have also important effect” should be “to also have important effects”

Corrected.

Line 376: “detailed” should be “details”

Corrected.

Line 379: “simulated period” should be “simulation period”

Corrected.

Line 379: “since, the latest, yields the” is worded awkwardly and parenthesis are misplaced. Maybe something like “since it yields the…”?

Corrected.

Line 386: “respect to” should be “with respect to”

Corrected.

Line 398: “slight” should be “slightly”

Corrected.

Line 392: I think “statistically-significant” should be “statistically significantly”?

Corrected.

References

Bergström, R., Denier van der Gon, H.A.C., Prévôt, A.S.H., Yttri, K.E., Simpson, D., 2012. Modelling of organic aerosols over Europe (2002–2007) using a volatility basis set (VBS) framework: application of different assumptions regarding the formation of secondary organic aerosol. Atmos. Chem. Phys. 12, 8499–8527. https://doi.org/10.5194/acp-12-8499-2012

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