Responses to the comments of anonymous referee #2

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

GENERAL

Understanding sources of uncertainties in ozone and SOA simulations are important steps for improving air pollution modelings. This study compares two different BVOC schemes and the consequent impacts on ozone and SOA in Europe. The differences between PSI and MEGAN schemes are discussed. The authors found that PSI scheme predicts more monoterpenes while the MEGAN scheme predicts more isoprene. As a result, the CTM based on PSI yields more SOA than the results based on MEGAN scheme. The topic of this study well fits the scope of ACP journal, however, some essential limits may largely weaken the scientific merits of the work.

First, the differences in BVOC are most likely attributed to those in land cover instead of schemes. In general, PSI uses an earlier version of MEGAN parameterization for isoprene and a current MEGAN parameterization for monoterpene. They should have similar responses to environmental factors such as light and temperature. The main reason why PSI and MEGAN schemes show such a large difference in BVOC emissions is that they use different land cover. The authors clarified that PSI is based on tree species while MEGAN is based on PFTs. What if the MEGAN scheme uses the same land cover as PSI, but with PFTs aggregated from tree species? The land cover should be uniform before the comparison.

We agree that different land cover data is one of the major factors leading to differences between the two model outputs. However, the key point is not the land cover data itself, but the corresponding emission factors (PFT-specific for MEGAN, and species-specific for the PSI model). The GlobCover inventory (for the PSI model) is based on the MERIS satellite data obtained by the European Space Agency, while the CLM4-PFTs are derived from MODIS satellite data. In spite of different processing methods for the final data products, we do not expect a large difference in the coverage of broadleaved and needle-leaved forests.

Second, no BVOC observations are used to constrain simulations. Though the authors use the measurements of ozone and OA to validate model results, these are not the direct observations of BVOC. One can simulate right air pollution with wrong reasons (e.g., poor model performance, incorrect meteorology and so on). The only way to check the validity of BVOC schemes is to compare simulations with direct measurements of isoprene and/or monoterpene, which I believe there are many over Europe. Without BVOC constraints, the current study is more like a sensitivity test of ozone and SOA in CTM to any perturbations in BVOC emissions.

For the evaluation of the model we prefer to do this on more stable species like the secondary products such as ozone and SOA. The BVOC concentrations are strongly influenced by local mixing processes and chemistry due to the high reactivity of these molecules. The model output is unlikely representative for species with strong spatial gradients. Also, such measurements are very sparse. In spite of these considerations we agree with the referee that after all it is important to give some idea about the BVOC model performance. We added a figure about the comparison of modelled and measured isoprene in the revised manuscript (see new Fig. 4). The only measurements of some monoterpene species during the simulated period were in Finland. We compared them with our modelled total monoterpene concentrations. We also compared our results with some measurements reported in the literature for other years. We inserted the following text in Section 3.1 (P10, L3-L24) of the revised manuscript:

„ BVOC measurements are rare and the concentrations are associated with very high spatial gradients (especially vertical) due to high reactivity and local mixing processes that are unlikely captured by the model in the respective grid cell. Nevertheless but with these caveats in mind we compared a few measurements available for isoprene with our model results to get an idea about the range of differences.

Compared to monoterpenes, there were more isoprene measurements at various European sites in 2011 (see Fig. 4). Clearly, the MEGAN-isoprene data are much higher than the measurements at all 12 sites while the PSI- isoprene results are closer to the measurements.

Unlike the single compound of isoprene, monoterpenes consist of several species and therefore it is even more difficult to perform comparisons with measurements, which are rare and have large uncertainties. Only a limited number of MT measurements were reported in Europe (only in Finland) during our simulation period (Hakola et al., 2012; Hellen et al., 2012). Hakola et al. (2012) reported average MT concentrations of about 508 ppt (with a range between about 150 and 800 ppt) in August 2011 at the SMEAR II station at Hyytiälä. MEGAN-MT for the same period was 117 ppt while PSI-MT was around 2 ppb (for the same site, Rinne et al. (2005) reported MT concentrations of between 200-500 ppt during daytime and more than 1 ppb at nighttime in summer 2004). On the other hand, the measured MT concentrations at a nearby urban background station SMEARIII in Helsinki were lower, with around 117 ppt in summer (Hellen et al., 2012). Both models predicted higher concentrations for that site (MEGAN-MT 303 ppt, PSI-MT 1 ppb). In order to get an idea about the model performance in other regions, we compared our results also with MT concentrations measured at Hohenpeissenberg (southern Germany) in June 2006 (Oderbolz et al., 2013). Both model results (PSI-MT: 75 ppt, MEGAN-MT: 130 ppt) in that region were similar to measurements (~100 ppt). Although this comparison of measurements and model results for different years under different meteorological conditions has a very high uncertainty, it might help to understand the range of differences between the model results and the measurements. In general, all these comparisons suggest that MT concentrations might be underestimated using MEGAN emissions while PSI emissions might be too high over Scandinavia. On the other hand, both models seem to predict MT emissions relatively well in central Europe."

SPECIFIC

Page 1, line 32: "improving substantially the model performance", How do you know it improves the model for correct reason?

In this manuscript, we only show that the higher monoterpene emissions estimated by the PSI model lead to higher SOA formation and the agreement between the modelled and measured OA improves. In order to understand whether the improvement is due to the biogenic emissions, source apportionment studies are needed. The modelled fraction of biogenic and anthropogenic OA were found to be closer to the PMF analysis of the measured data at Zurich (Canonaco et al., 2013; Daellenbach et al., 2017) when the PSI emissions were used. This is the topic of another manuscript in preparation.

Page 2, Line 38: "highest over all the model inputs" What kind of inputs? Specify.

We rephrased the sentence (Page 3, Line 7-9) as follows:

"Comparison between MEGAN and another widely used biogenic emission model, the Biogenic Emission Inventory System (BEIS) indicated that the influence of biogenic emission models on ozone simulation results over the United States is far greater than using a different photosynthetically active radiation (PAR) input (Zhang et al., 2017)."

Page 4, Line 3: "Initial and boundary conditions were: : :" What kind of IC and BC? Specify.

We modified the sentence (Page 4, Lines 23-25) as follows:

"The gridded initial concentrations of chemical species in each layer of the model domain, as well as at the domain lateral boundaries were obtained from the global model data MOZART-4/GEOS-5 (Horowitz et al., 2003) with a time resolution of 6 hours.…"

Page 5, Line 31: "The value of 0.1 is used for MT in MEGAN2.1, while the values are between 0.065 to 0.077 : : : in PSI model" Why the PSI model uses different parameters while it uses the same scheme as MEGAN?

Although similar response functions are used by PSI model and MEGAN, the PSI model uses species-specific parameters for different monoterpene species historically based on the experimental results reported by Tingey (1980) (e.g. 0.065 is for β-phellandrene, 0.077 is for β-pinene).

Page 6, Line 3: "canopy model" What's the impacts of different canopy models on the simulated light availability for PSI and MEGAN models?

Leaf temperature and PAR in a forest vary substantially within the canopy. Canopy models take into account this effect to calculate the leaf temperature and light for sunlit and shaded layers of the canopy. Since the canopy models of the PSI model and MEGAN are based on similar principles, the effect of different canopy models on the light is not expected to be significant. A BVOC reduction of about 20% due to the canopy model was reported for the PSI model by Oderbolz et al. (2013), however, the influence of different canopy models on BVOC were within the uncertainty range of observed fluxes (Lamb et al., 1996; Guenther et al., 2006). We added a sentence in Page 7, Line 3-5.

"A BVOC reduction of about 20% due to the canopy model was reported for the PSI model by Oderbolz et al. (2013). Although different canopy models could influence the modelled BVOC emission, such influence was within the uncertainty range of observed fluxes (Lamb et al., 1996; Guenther et al., 2006)."

Page 6, Line 30: "The variation of biomass density in MEGAN was simulated by the satellite data" How the satellite data simulates biomass density?

We are sorry about the incorrect wording in that sentence. The satellite data of leaf area index is used to quantify the amount and age of foliage for each grid cell via intermediate calculations of the canopy environment model and leaf age model within MEGAN v2.1. We revised the sentence (Page 8, Line 1-2) as follows:

"The biomass density in MEGAN was calculated by the canopy environment module based on the satellite data of the leaf area index (LAI, m^2 leaves per m^2 projected area) with a time step of 8 days."

Page 7, Line 27: "To demonstrate the seasonal differences". The word "demonstrate" is not appropriate, better to use "evaluate" or "quantify".

Done

Page 8, Line 7: "observed" This is not observation. Better to use "found"

Done

Page 8, Line 14: "In winter, highest isoprene emissions occurred in Central Europe for PSI model" Why there are isoprene emissions in winter when leaf biomass is set to zero.

We set the leaf biomass of only deciduous trees to zero but there are some coniferous species emitting isoprene, although the emissions are very low.

Page 9, Line 13: "observed", again not observation. Better to use "calculated"

Corrected

Page 9, Line 28: "background", where is the background ozone from?

The background ozone is the fraction of ozone that is not attributed to local anthropogenic sources and might originate from both natural and anthropogenic sources, such as stratospheric intrusion, long-range transport of ozone from distant areas or production from methane emitted from swamps and wetlands (Vingarzan, 2004). In our simulations, background ozone in the model domain was provided as initial (in

the domain) and boundary concentrations (at the lateral domain boundaries) by the global model MOZART. We revised the sentence in Page 12, Line 2-3.

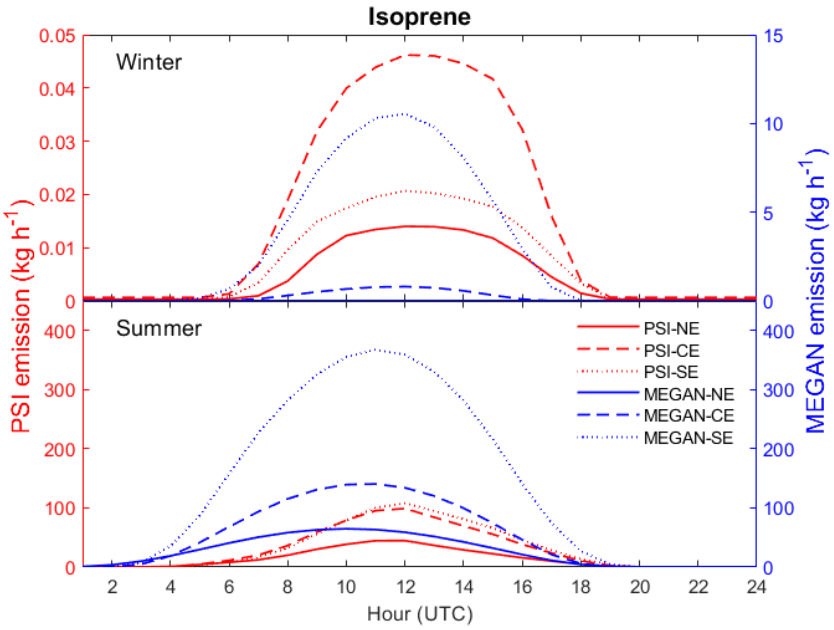
"An additional reason might be the rather low ozone production compared to the background ozone where the latter is not affected by local European emissions."

Page 10, Line 11: "and" should be "but"
Corrected

Page 11, Section 3.2.3: Not sure whether this section is necessary as BVOC has minor impacts on SIA
Although the overall influence of BVOC emissions on SIA is much smaller than on OA, it could reach up to 15% for particulate nitrate on the local scale. Their effect could be even higher in hourly time resolution (analyses were based on monthly average in this study) and under different meteorological conditions (Aksoyoglu et al., 2017). Therefore, we think the results are still important to understand the possible factors influencing the model performance on SIA simulation. Also, we added a sentence about the necessity to investigate the effects of BVOC emissions on SIA in the Introduction (Page 3, Line 17-20).

"Moreover, BVOCs also influence the secondary inorganic aerosol formation by changing the oxidant concentrations (Aksoyoglu et al., 2017; Karambelas, 2013; Sotiropoulou et al., 2004; Zhang et al., 2016). Aksoyoglu et al. (2017) found that doubled BVOC emissions in Europe led to an increase of particulate inorganic nitrate concentrations by up to 35%."

Figures 3 and 4: Why in CE, isoprene is much higher for PSI but ozone is still lower than MEGAN?
We think that there might be some misunderstanding. Isoprene by the PSI model in CE is lower than MEGAN in Figure 3. Please note that the scale of the left axis (for PSI emissions) is different from that of the right axis (for MEGAN emissions). To avoid misunderstandings, we updated the figure to have the same scales for the left and right axes for isoprene in summer, where it is seen that MEGAN-isoprene is higher than PSI-isoprene.



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