Responses to the comments of anonymous referee #1

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*.

Jiang et al. describe a modeling study comparing two biogenic VOC emission models, MEGAN and PSI, and their effects on modeled ozone and aerosols in Europe using a chemical transport model CAMx. The two BVOC models mainly differ in vegetation classification and reference basal emission rates. PSI predicts much lower isoprene emission but 3 times of monoterpene emissions higher than MEGAN. Such emission differences result in relatively small differences in ozone (<10%) but very large differences in SOA. The manuscript is well structured and generally clearly stated. The study focuses on the impact of different BVOC inputs which is one of the fundamentals of atmospheric chemistry. I recommend publication of this manuscript in ACP after minor revisions.

My major concern is that the two BVOC models predict very different patterns and magnitude of isoprene and monoterpene emissions, but readers have no idea about how good they are compared to real observations. I suggest to add a section comparing the PSI and MEGAN results with at least some in-situ measurement of isoprene and monoterpene emissions. Validation of only ozone and SOA are not enough to fully understand the strengths and weaknesses of the two models, as many other factors may contribute to the formation of ozone and SOA and they may compensate each other.

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 the reader 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."

Other comments:

P6 L1: Are those factors including soil moisture and CO_2 dependence "turned on" in your simulations?

Since we are using the offline version of MEGAN v2.1, the soil moisture and CO_2 dependence corrections were not included (Emmerson et al., 2016). We used the default parameterization where these factors were set to 1. The CO_2 inhibition effect might be significant in regions with high CO_2 and isoprene emissions. Studies using global coupled land-atmosphere models reported that accounting for CO_2 inhibition has little impact on predictions of present-day global isoprene emissions but might have larger effects on future emissions (Heald et al., 2009, Tai et al., 2013). We rephrased the sentence in the revised manuscript (P6 L23-L25) as follows:

"In addition to the light and temperature response, MEGAN v2.1 covers also some other factors such as leaf age and leaf area index (Guenther et al., 2012). Since the correction of soil moisture and CO_2 dependence are not included in the offline version of MEGAN (Emmerson et al., 2016), we used the default parameterization where the correction factors were set to 1."

Figure 2: Font of legends should be consistent.
Corrected

Figure 3: How to interpret different MT peak time in MEGAN and PSI (in summer, bottom panel), even though they adopt a similar T-dependent function and use the same meteorology input?

In addition to the T-dependent pool emissions, both the PSI model and MEGAN include species having both light and temperature dependent synthesis emissions. Different fractions of the light-dependent MT emissions of the two models could lead to different MT peak times. We added the diurnal variation of T and PAR to Figure 3 in the revised manuscript to show the different T/PAR dependence of MT emissions of two models. We updated Section 2.2.2 (P6 L19-L22) to clarify the influence of light-dependent response to MT emissions as follows:

"The light-dependent synthesis emissions of MTs were considered in MEGAN v2.1 as described in Guenther et al. (2012). Depending on different MT species, the light-dependent fraction of MT emissions ranges between 0.2 to 0.8 for MEGAN. In the PSI model, the light-dependent emissions from Norway spruce are calculated for each monoterpene species as a function of PAR based on the data of Schürmann (1993)."

We also added an explanation about the different MT peak times in Section 3.1 (P9 L27-L31) as follows:

"Comparison of monoterpenes emissions (Fig. 3b) with temperature and photosynthetically active radiation (PAR) (Fig. 3c) indicates that monoterpene emissions by the PSI model are mostly temperature-dependent while the influence of light is stronger for the MEGAN–MT emissions. For instance, the highest PSI–MT emissions in summer occurred at the same time of the highest temperature (13:00–14:00 UTC), while the occurrence of highest MEGAN–MT is close to the PAR peak (10:00–12:00 UTC)."

We revised the sentence (P10 L25-L28) as follows:

"Studies comparing different models with each other, as well as with measurements suggest that MEGAN tends to overestimate isoprene emissions especially in Scandinavian countries and south-west Europe and to underestimate monoterpene emissions by more than a factor of 2 (Bash et al., 2016; Carlton and Baker, 2011; Emmerson et al., 2016; Poupkou et al., 2010; Silibello et al., 2017)."

We rephrased this sentence (P11 L21-L23) to clarify it as follows:

"The spatial distribution of the ozone difference, i.e. (PSI-O_3) – (MEGAN-O_3) (Fig. 6, right panel) is very similar to that of the difference in the isoprene emissions (Fig. S2a)"

Several European studies reported that ozone formation in most regions is NO_x-sensitive in general except around the English Channel, Benelux and Po Valley regions, where NO_x emissions are higher and the response to a change in the VOC emissions is relatively stronger (Beekman and Vautard, 2010; Aksoyoglu et al., 2012; Oikonomakis et al., 2018). We added some discussion in P11 L34 to P12 L11.

"The main reason for the weak effect of the isoprene emissions on ozone is the stronger sensitivity of ozone formation in general to NO_x emissions rather than VOC emissions in Europe. 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 (Oikonomakis et al., 2018; Sartelet et al., 2012). Several European studies reported that ozone formation in most regions is NO_x-sensitive except around the English Channel, Benelux and Po Valley regions, where NO_x emissions are high (due to intensive anthropogenic NOx emissions from both land and shipping or geographical characteristics leading to high accumulation of pollutants) and the response to a change in the VOC emissions is relatively stronger (Aksoyoglu et al., 2012; Beekmann and Vautard, 2010; Oikonomakis et al., 2018). However, the sensitivity of ozone formation to its precursor emissions might change as a result of large NO_x emission reductions in Europe since 1990 according to the Gothenburg Protocol. On the other hand, emissions from shipping activities are not regulated as strictly as land emissions and have been increasing continuously especially in the Mediterranean, affecting both ozone and particulate matter concentrations (Viana et al., 2014; Aksoyoglu et al., 2016)."

We totally agree that showing the contribution of biogenic versus anthropogenic sources would be more straightforward. However, this is a topic of another manuscript (in prep.) in which we focus on the source apportionment of organic aerosols, therefore we prefer not to show such figures in this manuscript. In order to reply the referee's question however, we show below (Fig. 1) the anthropogenic and biogenic OA concentrations (stacked) modelled using the PSI and MEGAN emissions at two sites. These figures show that the contribution of biogenic emissions to OA is higher with the PSI emissions at both sites. The modelled fractions 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.
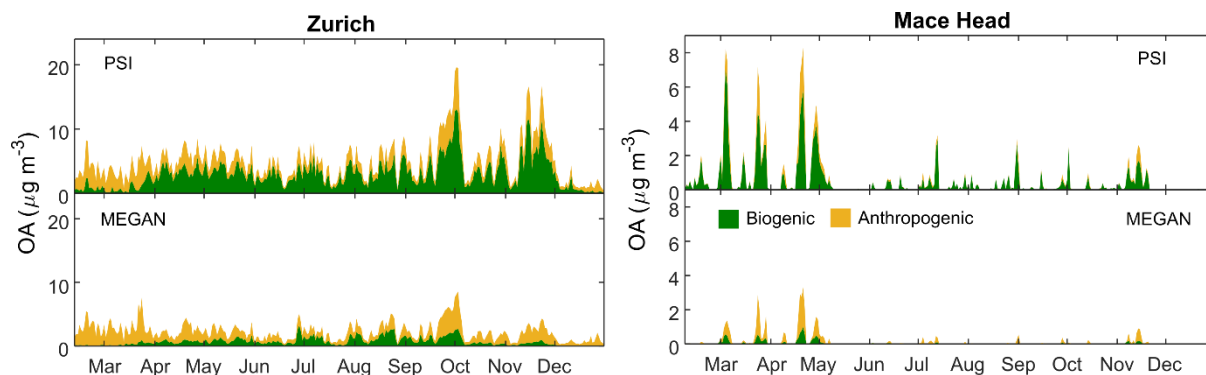
Fig. 1: Time series of anthropogenic and biogenic OA modelled by using PSI and MEGAN BVOC emissions at Zurich (left) and Mace Head (right).

P11 L27: "vertical distribution of elevated emissions" should be "vertical ventilation"?
What we mean is the injection of point-source emissions into the vertical layers of the model domain. We revised the sentence (P14 L14-L15) to make it clearer as follows:

"The precursor gases SO_2 and NO_x from anthropogenic point sources (continental, shipping) (Fig. S8) might be accumulated too much in the surface layer since all emissions were injected into the 1st model layer, leading to too high SIA formation. "

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