Responses to the comments of anonymous referee #3

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

Review Summary

Jiang et al. simulated ozone and aerosol concentrations in Europe using two different biogenic emission models (PSI and MEGAN) to probe uncertainties in regional air quality models. They compared model results with ozone observations from the European air quality database, AirBase, and aerosol observations from eight different measurement locations with an Aerodyne AMS or ACSM. Results were generally consistent with previously published papers demonstrating that MEGAN tends to over-estimate isoprene and under-estimate monoterpene emissions. They also found that the simulated ozone mixing ratios between the model runs varied less than the isoprene emissions. This is also consistent with previous studies showing much of Europe's ozone production is NOx-limited rather than VOC-limited. Finally, their model comparison suggests higher monoterpene emissions lead to better comparison between simulated and observed organic aerosol. The authors acknowledge this could be due to compensating factors (e.g. they could be "right for the wrong reasons"). Overall, the scientific approach is reasonable and the scientific questions are appropriate for the scope of the journal. However, it is unclear what information this paper is adding to the scientific community that has not already been published in previous papers. There are also a number of gaps in the methods section that lack clarity. I recommend publication after the manuscript is revised to address the following comments.

General Comments

The authors should better clarify how this particular paper is filling in gaps that have not already been addressed in previous publications. All results sections generally state the results are consistent with work that has already been published, and so it is very unclear what the conclusions from this paper are adding to the growing body of scientific knowledge. The manuscript could better highlight how this work is filling in unique gaps in understanding.

Although our results are consistent with previous studies in general, we think that they provide much more additional information. To our knowledge, there are only a few studies comparing emissions from different BVOC models (Karl et al., 2009; Keenan et al., 2009; Steinbrecher et al., 2009), but comprehensive studies showing the impacts of using different BVOC emission models on secondary pollutants in Europe are scarce. Some studies report the effect of biogenic emissions with zero-out simulations (Sartelet et al., 2012) or with doubled BVOC emissions (Aksoyoglu et al., 2017; Ciarelli et al., 2016). Curci et al. (2009) compared effects of two different biogenic emission inventories, one based on Guenther et al. (1995) and one based on Steinbrecher et al. (2009), on ozone in Europe during 1997 to 2003. However, the limitation of ozone production might have been altered due to large emission reductions of the various precursors in Europe during the past decades.

Our main goal in this study is not just to compare two BVOC models but rather to show how using different BVOC emissions affect the modelled secondary pollutant concentrations and how the effects change spatially and temporally. We chose MEGAN since it is the most widely used biogenic model globally, and the PSI model (which was developed originally for Switzerland and updated for the European domain) to represent models developed specifically for a regional scale. We investigated the effects of using different BVOC emissions not only during summer periods but throughout the whole year. We believe that the OA evaluation with a wide coverage of existing ACSM/AMS measurements in Europe during the simulation period provides valuable information about the influence of BVOC emissions in different parts of Europe in different seasons. In this way, we also want to emphasize the need to harmonize the biogenic emissions as much as possible in model inter-comparison studies. Although their importance on air quality modeling results are well known, BVOC emissions are usually not prescribed in model inter-comparison studies (e.g. AQMEII, Eurodelta, MICS-Asia) making it very difficult to compare and interpret the results.

Furthermore, although the effects of different BVOC emissions on ozone have been reported in a few previous studies, it is important to keep the knowledge updated in the context of continuous reduction of anthropogenic emissions since 1990s, which could change the sensitivity of secondary pollutants formation to precursor emissions in some regions. We revised the Introduction to make the objective and novelty of this study clearer as follows:

Page 2, Line 9-18:
Although there are a few studies comparing different BVOC models (Steinbrecher et al., 2009; Karl et al., 2009; Keenan et al., 2009), comprehensive studies showing the impacts of using different BVOC emission models on secondary pollutants in Europe are scarce. Some studies report the effect of biogenic emissions with zero-out simulations (Sartelet et al., 2012) or with doubled BVOC emissions (Aksoyoglu et al., 2017). Curci et al. (2009) compared the effects of two different biogenic emission inventories, one based on Guenther et al. (1995) and one based on Steinbrecher et al. (2009), on ozone in Europe during 1997 to 2003. However, the limitation of ozone production might have been altered due to large emission reductions of the various precursors in Europe during the past decades. Understanding the potential influence of biogenic emissions on European air quality is therefore of great importance especially under the continuously reduced anthropogenic emissions since early 1990s.

Page 3, Line 22-24:
In spite of an increasing interest in understanding the influence of biogenic emissions on ozone and aerosols, limitations still remain: most of the studies focus on short periods (mostly in summer), while the potential influence of BVOC on SOA could still be high in winter at local scale, the evaluation of modelled OA is challenged by the scarcity of field measurements, and not much attention has been paid to the effects of BVOC on SIA by different biogenic models.

Page 3, Line 25-29:
Biogenic emissions in Europe were estimated by two BVOC emission models with different land cover and emission factors; MEGAN as a widely used model globally and the PSI model to represent models developed for a specific region. The BVOC emissions from the two models were then used as input for the regional air quality model Comprehensive Air Quality Model with extensions (CAMx) to simulate gaseous and particulate pollutant concentrations in 2011.

Specific Comments
METHODS: SECTION 2.2.1 EMISSION RATES Authors state they estimate reference emission rates of isoprene and monoterpenes based on Lamb et al., 1993 but then go on to say Norway spruce isoprene emissions were estimated to be 10% of alpha-pinene. It is unclear why Norway spruce was handled differently and why it is singled out to be described separately from the other plant species. Please clarify.
The PSI model was originally developed in the early 90s only for Switzerland using a very detailed tree inventory. Norway Spruce covers almost half of the Swiss forests (49%) and it is also an abundant forest type in the other European regions, it was therefore treated explicitly using some explicit data available in Europe at that time (Schürmann, 1993; Steinbrecher, 1989).

The PSI model emission rates are species-specific except for "pasture" and "crop" (Table 1). How much variability would you expect between different types of "pasture vegetation" and "crop vegetation" based on the literature? What proportion of the total area covered in the model is characterized as "pasture" and "crop"? Is it a significant portion of the land that could drastically impact results or is it minor?
Although the coverage of crop and pasture is large (see Fig. 1 below), their contribution to total BVOC emissions is small because of their low biomass density, emission rates and short vegetation period compared to forests (Simpson et al., 1999). Therefore, we believe that the impact of different types of pasture and crops on the results is minor.

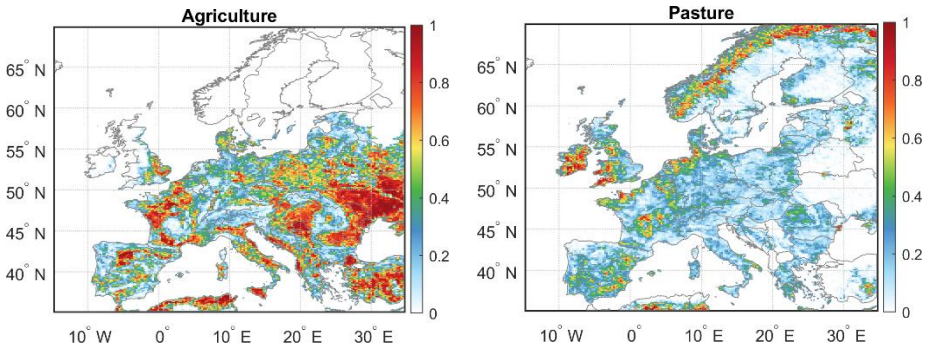

Fig.1: Fraction of agricultural land (left) and pasture (right) in the model domain.

Sesquiterpene emissions: authors state that sesquiterpene emissions were assumed to be 5% (by weight) of monoterpene emissions based on field measurements from various studies, but then cite a single paper that is actually a modelling paper and not a review or synthesis of measurements. Please cite the original literature from which this "5% (by weight)" reference is derived.

Thank you for this comment. The approximation of sesquiterpene emissions as 5% of monoterpenes was estimated using the data compiled from various emission databases containing both monoterpene and sesquiterpene emission rates for 116 tree species (Steinbrecher et al., 2009, Suppl.). We revised the sentence about sesquiterpenes (Page 5, Line 25-27) as follows:

"…SQT emissions were treated only as pool emissions and assumed to be 5% (by weight) of the monoterpene emissions based on the emission rate data for 116 species compiled from various studies as given by Steinbrecher et al. (2009)."

METHODS: SECTION 2.2.2, RESPONSE FUNCTIONS If sesquiterpenes are being treated as pooled emissions as stated in the previous section, then they will be treated similar to monoterpenes. However, the authors do not discuss what value they used for sesquiterpenes. In Guenther et al., (2012) the empirically-derived temperature correction coefficient, for sesquiterpenes was 0.17. Was that the value used in this study as well? Please clarify.

As we stated before, sesquiterpene emissions in PSI model were not calculated explicitly, but their emission rates were scaled to the monoterpene pool emissions. Therefore, they were treated similarly to the monoterpene emissions (5% (by weight) of the monoterpene emissions, as stated above).

Again, the authors single out Norway spruce emissions being handled a bit differently than other plant species. In this case, the Norway spruce monoterpene emissions have some light-dependent fraction estimated based on a study in 1993. Why is Norway spruce being singled out for more detailed emission estimation? Is it the dominant species in the modeling domain? This should be clarified.

The Norway spruce (*picea abies*) is indeed the most typical tree species in northern and central Europe. As we explained above, the PSI model was originally developed in the early 90s for Switzerland. Norway Spruce covers almost half of the Swiss forests (49%) and it is also an abundant forest type in the other European regions, it was therefore treated explicitly using data from Norway spruce studies.

Also, more details should be included about how the light-dependent emissions were estimated instead of just referring to the 1993 paper with no summary of what information was taken from that paper and used in this study. Finally, does this section then imply that all other monoterpene emissions were light-independent? Can this be stated more clearly and justified? If all monoterpene emissions are being treated as light-independent (except for some unstated fraction of Norway spruce monoterpene emissions), then this should be justified because it is well known that a substantial fraction of monoterpene emissions are

lightdependent; for example, in MEGANv2.1 the light-dependent fraction of monoterpene emissions ranges from 40-80%! (see Guenther et al., 2012, Table 4).

We apologize for the ambiguity in this issue. We referred to Guenther et al. (2012) in Page 6, lines 1-2 for MEGAN. In the PSI model, light-dependent MT emissions were calculated as a function of PAR for all the individual monoterpenes emitted from Norway spruce. We updated this section in the revised manuscript (Page 6, Line 19-22) 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)."

METHODS SECTION 2.2.3 INPUTS OF DRIVING VARIABLES Unclear how GlobCover 2006 data is being used to derive species-level distributions. How did the authors go from fractions of "needleleaf, broadleaf and mixed forests" to plant species distribution using the profiles from Simpson et al., 1999? There is missing information here that links the two.

We added the detailed procedures describing the calculation of the species-level distribution based on GlobCover 2006 data and Simpson's profile in Page 7, Line 27-32.

"The original 35 forest species in Simpson et al. (1999) were grouped into 10 classes (including 5 coniferous and 5 broadleaf species), and the ratio of each species class to the total coniferous forest and broadleaf forest was calculated (Table S2). The ratio of "other trees" were proportionally added to the 10 tree species. As the "other trees" are mainly found in a few Mediterranean countries, their influence on the whole domain is small. The species-coverage was then generated by multiplying the forest coverage from GlobCover with the country-specific tree species profile."

How much of variation between PSI and MEGAN emissions was driven by differences in normalizing emissions to leaf surface area (MEGAN) versus leaf biomass (PSI)? Are there potential biases that could vary between plant types for comparing total canopy-scale flux that arise from how surface area versus biomass are scaled up?

The PSI model estimates the plant-specific emissions and it uses the biomass densities (g m^{-2}) to convert the emission factors of specific plant species (in μg g^{-1} h^{-1}) based on Steinbrecher et al. (2009) to emission rates in μg m^{-2} h^{-1}. On the other hand, MEGAN emission factors for each PFT are given directly in μg m^{-2} h^{-1}. At the end, the emission rates in both models are in μg m^{-2} h^{-1}. A direct comparison is not possible because of different modeling approaches. Differences might arise also from using different land use data, different emission factors and different biomass densities. However, a detailed comparison of BVOC models is out of scope of this manuscript. We focus here on the effects of using different BVOC models on modelled secondary pollutant concentrations.

How did the authors ensure they were making meaningful comparisons between the models with the emissions normalized differently? Figure 2 was clear: authors graphed the emission rate per model grid cell. It was less clear how this comparison was done in Figure 3 where the graph simply shows the emission rate. Was this also per model grid? Per entire modeling domain? This should be stated more clearly.

The Figure 3 was based on the average emissions per grid cell in the entire model domain. We revised the units of the y-axis labels (kg $cell^{-1}$ h^{-1}), and updating the caption to *"Diurnal variations of average grid-scale isoprene and monoterpene emissions in the model domain"*.

Figure 2: right axis label is cut off on third row.

Corrected.

Figure 4: Figure caption should be re-worded. Currently states, "Mean bias of surface O3 mixing ratios in the afternoon 12:00-18:00 UTC) for each bin of observed ones in July 2011." I suggest revising to more clearly describe what is meant by "observed ones".
The Figure 5 (in the revised manuscript) caption was updated as:

"Mean bias of surface O_3 mixing ratios in the afternoon (12:00–18:00 UTC) for each bin of observed hourly average ozone in July 2011."

RESULTS O_3: results are consistent with previous literature demonstrating that O_3 production in most of Europe is in a NO_x-limited regime as opposed to a VOC-limited regime and thus the isoprene differences between the two models do not translate into large differences in ozone. Not a novel result. Can the authors comment on how this study is different from previous ones that have published the same result?
Although in the past ozone formation was more sensitive to NO_x in most of Europe, we think that it might be changing and would be different on a local scale as a result of large emission reductions since the 1990s. Our results suggest that the regions that are affected more by higher isoprene emissions from MEGAN are especially around the coastal regions in the south (see Fig. 5, right panel) where isoprene emissions are relatively higher than in other regions, but also where NO_x emissions from shipping are still high (not regulated as land emissions by the revised Gothenburg Protocol). It is therefore not clear how ozone formation will evolve with reduced land emissions while ship emissions continuously increase especially around the Mediterranean. We deepened the discussion by adding the regional analysis based on previous studies in P11, L33 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 (Aksoyoglu et al., 2016; Viana et al., 2014)."

OA comparison: the study does not have a single AMS/ACSM location in the Northern Europe region. Surely there are measurements at Hyytiälä, or some other boreal forest site in northern Europe. Can you justify why no measurement sites were included for northern Europe?
We agree that comparison with measurements in Northern Europe (where the difference in emissions between the two BVOC models is largest) is important. However, OA measurements in Northern Europe are quite scarce. Although the AMS/ACSM stations of Hyytiälä and Vavihill did not have data available for the period of interest, there was one dataset available from a campaign at SMEAR II station at Hyytiälä between 15 March and 20 April 2011 (Kortelainen et al., 2017). The comparison of modelled OA by both PSI and MEGAN emissions with that dataset showed that the modelled OA could capture the temporal variation of measurements and PSI emissions led to a better agreement between modelled and measured OA (see Fig. 2 below). The comparison of daily OA at SMEARII were added to Figure 7, and the statistical results of the new stations were added to Table 3.

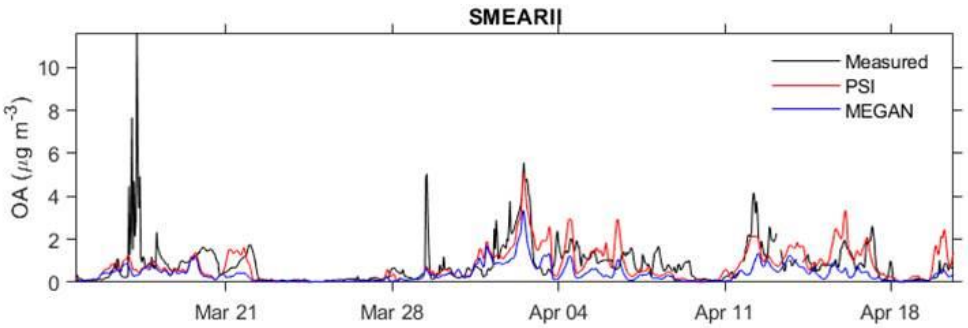

Fig. 2: Comparison of measured and modelled OA at SMEARII Hyytiälä station.

SECTION 3.2.3: INORGANIC AEROSOLS Authors do not set up a rationale in the introduction for investigating the impact of changing biogenic emissions on inorganic aerosols. Why would differences in biogenic emissions substantially alter inorganic aerosol? Without this rationale in the introduction, this section does not fit with the rest of the paper.

Isoprene and monoterpenes react with oxidants such as OH, ozone and NO_3 in the atmosphere and therefore they might lead to changes in oxidant concentrations, which are also involved in the formation of secondary inorganic aerosols such as ammonium nitrate and sulfate. Although such effects are smaller than the effects on organic aerosols, we think that it is worth including them. We have updated the introduction (in Page 3, Line 17-20) to highlight the rationale to study the impact of BVOCs input on inorganic aerosols.

"Moreover, the 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%."

DISCUSSION Authors end the paper by saying, "In future studies, BVOC emission models with more regional specific adaptation in vegetation types and emission factors are urgently needed to reduce the uncertainties in BVOC emission estimates in order to improve air quality modelling." Why is this the recommendation rather than simply improving the emission factors for the plant functional types in MEGAN? It is not reasonable to model the emissions from every single plant species on the planet. I don't agree that the results from this study emphasize the need for more plant specificity because even this paper only used a sub-set of 10 specific plant species (with an additional two general classes for "pasture" and "crop"). It seems to me that the major finding from this paper, consistent with published papers before it, is that the MEGAN emission factors could be updated and improved.

We agree with the referee that it is not reasonable to model the emissions from each single plant species. Our point is that the emission factors need to be improved based on the regional information such as vegetation types (for MEGAN). However, this suggestion is not specific for MEGAN, but also for similar species-specific models like the PSI model. As the referee noted, only 10 specific trees were included in the PSI model (they were originally selected according to the forest composition in Switzerland, they are however typical also for Europe), which should be improved in the future. We have revised that paragraph in Page 15, L24-26.

"In future studies, emission factors should be improved in BVOC models to include more regional specific vegetation types to reduce the uncertainties in BVOC emission estimates and to improve air quality modeling results."

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