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Dear Editor,

We are submitting the revised version of the manuscript in which we addressed comments and corrections pointed out by the reviewers. Please find below a list of the main changes made in the manuscript as well as the responses to the reviewers. We hope you'll find the current version of the manuscript suitable for publication in Atmospheric Chemistry and Physics.

On behalf of all authors I thank you for your considerations.

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List of main changes

- Values of the flux measurements in the Amazon were revised and corrected. Error bars were added to Figure 15 in order to illustrate uncertainties of the measurements. As suggested by Reviewer #1, the negative fluxes from both measurement campaigns (in the Amazon and Borneo) were excluded from the calculation of statistical comparison between model and observations, and values presented in Table 6 were corrected.
- In order to address comment of Reviewer #2, we performed additional model run evaluating the impact of use of climatological LAI instead of actual LAI values. Comment on uncertainty of the emissions resulting from the use of climatological LAI was added to Section 2.2.2.
- As a response to the comment of Reviewer#2 we have added a paragraph to Section 5 discussing the light dependence of monoterpene emissions in the Amazon.
- As suggested by the Reviewer#2 we extended the Conclusions with several recommendations on what we think are the important tasks to be addressed in order to better assess the biogenic VOC emissions.
- Two equations describing the use of light dependence fraction (LDF) factor in the MEGAN model algorithm were added to Section 2.1 (suggested by Reviewer #2).
- For better orientation in the graph, the monthly total emissions of isoprene and monoterpenes were added to Figure 5 (comment of Reviewer #2).

We would like to thank both reviewers for their reviews. Our responses to the comments and questions follow, point by point. The review comments are written in *italic*.

REVIEWER #1

The manuscript has been largely improved. It is suitable for publication after addressing these comments:

1. P10735 L5: "Yuan et al. (2011) processed the MODIS Collection 5 LAI data with a quality control algorithm in order to diminish uncertainties and inconsistencies". Has this method been used in this study to process the MODIS LAI?

• In our study we used the MODIS LAI data provided by the authors of the Yuan et al. (2011) paper. The data were processed following the method described in the paper. In order to be clear on this in the manuscript, the sentence on P10735, line 3 now reads: "We have used the 8-day high spatially resolved LAI data processed by Yuan et al. (2011) from global retrievals of the Moderate-Resolution Imaging Spectroradiometer (MODIS, Collection 5) for the period of 2000-2009. Yuan et al. applied a quality control algorithm in order to decrease uncertainties and inconsistencies."

2. P10738 L4: Since "As previously shown by Guenther et al. (2006) and Arneth et al. (2011), isoprene emissions are very sensitive to the driving meteorological data, especially temperature and solar radation", why the MS only considered the sensitivity of isoprene emissions to PAR. Moreover, "radation" should be "radiation".

• In the paper we wanted to evaluate the calculation of PAR from shortwave radiation: this approach is often applied due to the fact that PAR variable is scarcely available from meteorological preprocessors. We agree with the reviewer that the sensitivity of biogenic emissions to meteorological inputs, most importantly temperature and solar radiation, could be investigated more. However, we think this deserves another study. We are currently working on a paper that will follow up with the Guenther et al. and Arneth et al. papers and will evaluate impact of different meteorological data on biogenic emissions.

The typo was corrected.

3. In Section 3.2.1-3.2.6, authors separated the different input data with results. It is easy to confuse readers. So I suggest to combine the description of different input data and results in the one section.

- Since we compare the results of the different studies in 3.2.6, we have preferred to analyze all the results in one section. We could indeed discuss the results in each section, but this would have led to a lot of repetitions, and we would then need a section on the synthesis of the results. We think that this would make the full 3.2 section difficult to read.
- 4. The same as sensitivity analysis. Section 4.1 can be removed. Moreover, Table 3 is not

needed while Table 4 is used.

• We think that information given in Section 4.1 is different from that in Section 4.2. Section 4.1 provides a more general overview of global and regional studies on isoprene, while Section 4.2 focuses on more detailed spatial and temporal analysis of global isoprene estimates.

Tables 3 and 4 contain different information. Table 3 shows the comparison of the results from this study with previous regional estimates, while Table 4 summarizes the 5 global datasets that were used for more detailed comparison of isoprene emissions.

5. As for REA data, why there are negative values? Authors should explain the reason and the negative emission fluxes of isoprene and a-pinene should be excluded during the calculation.

 The REA measurement data were revised and error bars were added in Fig. 15 in order to illustrate the uncertainties on the measurements. The negative values are a result of subtraction of updrafts and downdrafts sampled by the DEA system meaning that the extend ensured down into the

the REA system meaning that the actual overall flux was oriented down into the canopy. There is no evidence of a technical or processing failure, which would indicate a necessity of rejecting these values. However, measured negative fluxes are now excluded from the calculation of statistical characteristics for the comparison in Table 6. This is because the MEGAN model simulates only the net primary emission from the ecosystem and does not account for a downward flux into the canopy. Similarly, values for OP3 study in Table 6 were recalculated taking into account only positive emission fluxes.

6. As shown in Fig.13, the results obtained by bottom-up method tend to be higher than that obtained by top-down method. More discussion should be given to explain it.

• Isoprene emissions calculated by bottom-up methods are indeed higher than those calculated by a top-down method for the datasets used in Figure 13. However, as can be seen in Figure 10, top-down methods can also give higher values.

7. Eq (2) cannot be found.

• Eq. (2) is located on page 10731, line 15.

REVIEWER #2

General comments:

The authors present the results of model simulations performed to produce a dataset of biogenic emissions for the 30-year period of 1980-2009. Sensitivity studies are conducted to attempt to quantify the effect of altering model driving data or parameters on modelled isoprene emissions estimates. The authors compare their inventory with global and regional emissions estimates generated in previous studies, and evaluate their modelled data with flux measurements made during field campaigns in the Amazon and Borneo. Their results indicate the relative importance of certain geographical regions, modelling assumptions and driving data to estimates of biogenic emissions derived from MEGANv2.1.

The data generated by this study are available to the community via the Emission of atmospheric Compounds and Compilation of Ancillary Data (ECCAD) website

(http://www.pole-ether.fr/eccad). While such an inventory is welcome, and in many ways long overdue, it would be good to see how the authors envisage it could be used. Do they intend for it to be used in place of the GEIA emissions for use with models that do not generate biogenic emissions on-line, or for evaluation of model output? Furthermore, is it the intention of the authors to extend this study to produce emissions inventories for future scenarios?

• The data presented in this study could be used for different purposes. The so-called GEIA emissions for BVOCs were built in the 1990s using much less detailed emission models. The presented dataset provides updated BVOC emissions with inter-annual variability. The algorithm and the emissions discussed in the paper are being used in the development of Atmospheric Service of the Copernicus European Program. The dataset is also used as emission input for models, which do not calculate biogenic VOC emissions on-line: for example, several models participating in the Chemistry-Climate Model Initiative (CCMI) are using the dataset. The dataset is publicly available and can therefore be used for model evaluation and inter-comparison. Producing emissions for future scenarios is part of our future work plan.

While the methodology of the study is rigorous and relatively comprehensive, their analysis of it would benefit from further discussion of the sources of discrepancies between their results and those of previous measurement campaigns and model studies, and the uncertainties involved in the parameterisations within the model. Having conducted such in-depth comparison and evaluation, the authors are well placed to make recommendations for the best targets of future research (both experimental and modelling) for the biogenic emissions community to narrow uncertainties and constrain emissions estimates. I recommend this article be accepted for publication in ACP subject to the authors satisfactorily addressing the comments and concerns outlined below.

Major concerns:

- In absence of available data the authors have used an average LAI derived from MODIS data for 2000-2009 for all other years (i.e. from 1980-1999 and 2010). There is no evidence that they have applied any weighting or scaling to these average data, and yet climate conditions varied markedly during the 1990s in particular which would be expected to affect GPP and hence leaf density. At the very least, the authors should conduct a further sensitivity study alongside S1 to indicate the potential error the use of average LAI may introduce to their inventory.

• We have performed additional model run using the climatological LAI for the period of 2000-2009 and comparing the results with the reference, in which the non-averaged MODIS data were used. The difference between the two runs is about 5%. The following sentence has been added to the manuscript in section 2.2.2:

"Using climatological LAI instead of the actual LAI values can lead to about 5% emission difference."

Minor concerns:

- Why stop at 2010? Is it the intention of MACC/CCMI to extend this dataset to include future projected emissions estimates for biogenic compounds?

• The period of 1980 – 2010 was selected in order to accommodate the reference hindcast simulations with specified dynamics of the CCMI project. Nevertheless, the dataset will be updated with most current years and the data will be posted on the

ECCAD database. We plan to make a new version of the dataset in 2015 using the new GMAO meteorological data product (GEOS-FP).

- Why have the authors chosen to compare their estimates against those of previous datasets for the year 2007 when one of the previous studies only has estimates available for 2003? Surely it would make far more sense to evaluate data from the same year for all studies? As the climatology will be different for the different years, it would be expected that, particularly, on a regional basis, emissions would likely be very different.

• We agree that it would be ideal to make comparison using the same year for all datasets. However, the BISA-top-down dataset is available only after the year 2007 (Table 4 in the manuscript). Since this was the only dataset among studies selected for comparison prepared by the top-down approach, we gave preference to the year 2007 instead of 2003.

- While the presentation and quantitative discussion of the results of this study are rigorous and detailed, they lack any real qualitative discussion and conclusions. In particular, I would like to see more attribution of the discrepancies between datasets (both modelled and measured). Why, for example, should there be large differences between MEGAN-MACC estimates and regional studies for Europe (which has also been demonstrated in previous work)? Why do different LAI datasets produce such different emissions estimates in Australia? Why do modelled Amazon fluxes differ in absolute value from measurements when they capture the seasonality so well? And on p10751, the authors write "other factors than meteorology are likely to play an important role in driving the emissions"; these other factors should be outlined here.

• The paper includes indeed an analysis of the differences between different datasets. Section 4.2 discusses the differences between four available isoprene emission estimates in different parts of the world, i.e. the BISA bottom-up and top-down emissions, the GUESS-ES dataset, and the MEGANv2 emissions. More analysis could indeed be done, but we think that detailed analyses for many different regions would make the paper too long. Such very detailed analyses are planned for the coming months/years with colleagues from different regions, who have a detailed knowledge of region-specific issues.

As pointed out by the reviewer, there are other factors than meteorology that are likely to influence the emissions, and we have added a list of some of these other factors in the text. The sentence on p10751 now reads:

"Finally, other factors than meteorology are likely to play an important role in driving the emissions, e.g., the representation of ecosystem composition which leads to the determination of emission potentials and leaf area index, biotic stress, abiotic stress such as oxidative capacity of the ambient air."

- In their conclusions, the authors assert that comparisons of their modelled emissions against measured fluxes show good agreement. This hardly seems the case for Borneo, where the authors appear to have (rather arbitrarily) scaled their emissions estimates by a factor of 1.7 to bring them in line with measured fluxes (see the further comment below regarding Fig.16); the authors themselves go on to highlight this discrepancy (rather inconsistently with their assertion of good agreement).

• We have changed the conclusion to make clearer that the comparison with observations shows discrepancies with the results of some observations. The conclusion now says:

"The comparison shows relatively good agreement in the Amazon. The modeled monoterpene daily mean emissions for Borneo correspond well to the measurements; however isoprene estimates are on average a factor of 1.7 higher than observations in this region."

Comment on this discrepancy is given further in the Conclusions (p10755, line 28): "Recent flux measurements obtained during the OP3 study over the tropical forest in Malaysia (Langford et al., 2010) suggest that MEGAN model calculations in Southeast Asia might be overestimated due to utilization of emission potential value, which was found to be up to factor of 4 higher than the one measured in the field. Emission reduction in this region has also been supported by the study of (Stavrakou et al., 2014) who constrained isoprene emissions using the inversion of formaldehyde satellite retrievals."

- Please consider including a comparison with Barkley et al.'s more recent Amazon emissions estimates, as per the Short Comment.

• Comparison with Barkley et al. (2013) was included in the paper.

- On p107521, the authors discuss the comparison of modelled and measured nighttime monoterpene emissions. Their results appear to suggest that South American monoterpene emissions are light and temperature controlled, and if that is the case, can they recommend an appropriate light-dependent factor to be applied to monoterpene emissions calculations for S America?

• The current version of the MEGAN model takes into account both a) light and temperature and b) temperature only driven monoterpene emissions. In case of α-pinene, MEGAN sets 60% of its emissions to be light and temperature dependent, and 40 % to depend only on temperature. The MEGAN model currently assigns the light dependence fraction (LDF) factor to specific monoterpene species disregarding the geographical location of its source. The comparison of modeled and measured monoterpene emissions in the Amazon would suggest that the dependence of monoterpenes on temperature only in this region is either very limited or not present as the emissions fall to zero over night. This is also supported by several experimental studies performed in the Amazonian tropical forest (Rinne et al., 2002; Kuhn et al., 2002; Karl et al., 2004). According to these results, the LDF varies with geographical location and for monoterpenes emitted in the Amazon should be set close to 1.

The paragraph on p10751, line 13, now reads:

"On average, MEGAN simulates the α -pinene emissions well (Table 6), however, it does not capture the high frequency fluctuations of measured fluxes and tends to keep monoterpene emission levels above zero during night (Fig. 15). The MEGAN model algorithm for estimation of monoterpene emissions combines monoterpene dependence on light and temperature (similarly to isoprene) and to temperature only. As a result, modeled monoterpene nocturnal emissions never fall to zero and remain fairly constant during nighttime. The model defines the fraction of emissions dependence fraction (LDF)

factor. For α -pinene LDF is set to 0.6, which means that MEGAN expects 60% of α -pinene emissions to be both light and temperature dependent, and 40% of the emissions to depend on temperature only. There is an increasing experimental evidence (Fig. 15; Rinne et al., 2002; Kuhn et al., 2002; Karl et al., 2004) that monoterpene emissions in the Amazonian tropical forest are strongly light dependent and the results suggest that the LDF factor for monoterpenes emitted in this region should be set close to 1."

- Finally, the authors should make specific recommendations of where further research (modelling and experimental) would be of most benefit in constraining emissions estimates.

• As suggested by the reviewer, we have changed the last line of the conclusion, which now says:

"Nevertheless, more observations on isoprene and other BVOCs in different parts of the world are needed for a better emission quantification and for a better understanding of the different factors driving the emissions of all these species. We see a great potential in using the satellite observations to constrain or derive biogenic VOC emissions. However, previous studies have shown inconsistencies in emission estimates suggesting that use of satellite data for these purposes is still connected to large uncertainties. In order to increase accuracy of these methods, more investigation is needed in the validation of data from different satellite instruments, in evaluation of dependence of the method on a priori emissions and on chemical scheme applied"

Technical corrections:

p10728, L7-11 – please make it clear that their impact on the atmosphere has been identified by MODELLING studies;

p10728, L11-13 – tropospheric ozone also has climate impacts via radiative forcing;

p10728, L23 – please give the version number of MEGAN used (i.e. v2.1);

p10729, L2 - please state the units of the flux, F;

p10729, L11 – replace "on isoprene and only temperature" with "on isoprene emission rates but only the temperature";

p10729, L28 – replace "development" with "developmental";

• Corrections were applied in the manuscript.

p10730, L7-9 – please include the equation that Sakulyanontvittaya et al introduced to better explain the inclusion of the light dependent factor;

• Two equations describing the use of light dependence fraction factor for γ_P and γ_T were added on p10731, line 23. The sentence now reads: "Both γ_P and γ_T are divided into the light dependent (LD) and light independent (LI) parts using the light dependence fraction factor (LDF) as defined by Sakulyanontvittaya et al. (2008) and Guenther et al. (2012)

 $\gamma_{P} = (1-LDF) + LDF \gamma_{P_LD}$ (3) $\gamma_{T} = (1-LDF) \gamma_{T_LI} + LDF \gamma_{T_LD}$ (4) "

p10730, L16 – replace "compounds" with "compound groups";

• Correction was applied in the manuscript.

p10732, L8 - I would suggest explaining why the CO2 factor is 1 for other compounds. Perhaps something along the lines of "In view of the lack of clear experimental evidence of an effect, gammaCO2 is set to 1 for all other species.";

• Suggested sentence was added to the manuscript to the description of the γ_{CO2} factor on p.10732, L8.

p10732, L10 – insert "a" between "for" and "canopy";

p10732, L11-16 – this sentence does not make grammatical sense. Perhaps the authors could replace "Additionally" with "Further standard canopy conditions include" which would be slightly better;

p10733, L3 – please insert "the" between covers and modern;

p10734, L25 – replace "potential" with "potentials";

p10735, 3&7 - I assume that by "8 daily" the authors mean 8 days rather than 8 times per day, in which case "8 daily" should be replaced with "8-day";

p10735, L17 – replace "using a difference" with "based on changes", and refer Guenther et al, 2012 here;

p10735, L20 – insert "the" between "to" and "spatial";

p10736, L21 – insert "The" before "dominance" and "the" between "of" and "south"; p10737, L7 – add "of mean annual monoterpene emissions." at the end of the sentence;

p10737, L10 – insert "The" before "graphs";

p10737, L11 – insert "," after "species" and "the" between "and" and "higher";

p10738, L4 – insert "the" between "and" and "species";

p10739, L2-3 – insert () around "described in : : :: : : Eq. (3); insert "and" between "2.2.2" and "calculated"; remove the "the" before "Eq.";

p10739, L9 – move "," from after "Amazon" to after ")";

p10740, L1 – Heald et al would be a better reference here as Heald used the same

(Wilkinson) parameterization whereas Arneth used the Possell algorithms;

p10740, L10 – replace "equals to" with "is";

p10741, L5 – move "gammaSM" to after "factor";

p10741, L8 – replace "limit" with "limiting";

• Corrections were applied in the manuscript.

p10741, L12 & p10742, L19-20 – Why do sensitivity studies S4 and S5 also have acronyms (MEGAN-MACC-SM and MEGAN-MACC-SW) when none of the other sensitivity studies do?

• The acronyms for S4 and S5 (MEGAN-MACC-SM and MEGAN-MACC-SW) were removed from the manuscript.

p10743, L9 – remove "in global total" as this has already been made clear; p10745, L11 – replace "at" with "in"; p10747, L16 – insert "the" before "Northern Hemisphere's"; p10747, L28 – replace "update of" with "updating"; p10748, L2-3 – replace "Difference" with "Differences" and "originates" with "originate"; p10748, L27 – this should be a single sentence: ": : : regional totals, except for: : : "; p10749, L22 – "fluxes" should read "flux"; p10752, L4 – insert "the" before "dry season"; p10752, L5 – insert "the" before "wet season"; p10752, throughout – "during end of dry (wet) season" should read "during the end of the dry (wet) season"; p10752, L16&17 - replace "in the end" with "at the end"; p10752, L25 – insert "the" between "both" and "wet"; p10752, L28 – insert "the" between "during" and "dry"; p10753, L19 – "accual" should read "actual"; *p10753*, *L22* – *replace* "can be" with "is"; p10753, L23 – "malaysian" should read "Malaysian"; p10754, L1 – replace "can" with "could"; p10754, L6 – remove "being emitted"; p10754, L14 – replace "equals to" with "is"; Table 3 – this table would be far easier to read and understand if the data of previous studies and this study were presented in 2 separate columns rather than being separated by "|";

• Corrections were applied in the manuscript.

Fig. 5 – please consider giving some indication of the total emissions for each month on the graph as these fluctuate, either with a third bar or using a line graph;

• The monthly total values of isoprene and group of monoterpenes were added to the graph in Fig. 5.

Fig. 16 - please justify the apparently arbitrary scaling factor applied to modelled emissions of 1.7 - the emission potential used in the model is 7 while the emission potential estimated from measurements was 1.6, which would suggest a much higher scaling factor should be applied.

• The factor of 1.7 is a mean ratio between the measured and modeled isoprene values at the OP3 study site.

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