Response to the Referee 2

We would like to thank the referee for the thoughtful comments. We have addressed all of the comments. Our responses are itemized below.

GENERAL COMMENTS:

Reviewer:

It is not clear in the Introduction why the authors choose to compare the GEIA and MEGAN emission inventories, when Arneth et al. (2008) found that they have similar global isoprene emissions. Are there large regional discrepancies? The GEIA inventory, which precedes MEGAN, has been replaced by MEGAN in most global chemical transport models. Are research groups still using the GEIA inventory to estimate isoprene emissions? Both inventories use empirical algorithms of the same heritage. It would be easier to justify a study that compares MEGAN and the process-based LPJGUESS inventory (Arneth et al., 2007) and the influence of these two inventories on ozone levels in remote, rural locations in Europe. I suggest that the authors provide a justification for their comparison and use appropriate references to back their approach.

Reply:

‘’Are there large regional discrepancies?’’ Yes. We had estimated the large discrepancies in the previous version in the Mediterranean region where the CTM model underestimated ozone concentrations and using MEGAN improved the result.

‘’Are research groups still using the GEIA inventory to estimate isoprene emissions?’’ Yes, in the atmospheric air pollution modeling community, the GEIA model has been widely used, and the general trend is that modeling groups are substituting the GEIA model with the MEGAN model, which is considered more extensive and more up to date. Therefore it is important to document whether a model performance improvement is obtained using the newer MEGAN model even though the two models are based on the same methodology.

‘’It would be easier to justify a study that compares MEGAN and the process-based LPJGUESS inventory’’. In the air pollution modeling community, implementing a much more comprehensive model like the LPJ-GUESS is considered to be the next step after using the MEGAN model. We also consider implementing LPJ_GUESS to be the next step and have a plan to do this comparison in the near future.

‘’ use appropriate references to back their approach’’: We have added appropriate references.

Reviewer:

Guenther et al. (2000) is quoted to substantiate that emissions of natural NMVOCs exceed emissions from anthropogenic NMVOCs. More appropriate references include Guenther et al (1995), where total natural NMVOC emissions are estimated, and Oliver et al. (1996), a global anthropogenic emissions inventory.

The authors reference studies that have used emission inventories in their analysis, but without any information about the relevance of those studies to their comparison and evaluation. For example,

the authors state that “A number of studies have also been conducted to evaluate results of the integrated BVOC models : : : with satellite, aircraft or ground-based observations : : :” on pg. 9251 line 5-7, but do not provide information about the major findings of these studies or at least the information from these studies that is relevant to their own.

Reply:

We have rewritten this section following the reviewer’s comment.

Reviewer:

The justification that the authors provide for evaluating the isoprene emission inventories in terms of ozone observations instead of isoprene concentration measurements is somewhat contradictory. They state on pg. 9248, line 5-6 that “Isoprene has a short lifetime, and hence it is very difficult to evaluate its emission estimates against measurements” and reiterate on pg. 9264 line 6 that “Due to the scarcity of observed isoprene data and the short lifetime of isoprene : : : an indirect evaluation was made based on measurements of ozone concentrations in Europe”. The authors use ozone data that was measured at the same sites in which isoprene concentrations were made (summarized in Table 1), implying that if isoprene data is scarce, so too is ozone data. Although the lifetime of isoprene is short, isoprene emission inventories could also be evaluated by comparing measured isoprene concentrations with simulated isoprene concentrations in DEHM using the two different emission inventories, as enhanced levels of isoprene is coincident with isoprene emissions. This reduces uncertainties in transport (as isoprene is short-lived) and isoprene oxidation chemistry (as loss of isoprene due to oxidation occurs in the first oxidation step, whereas ozone formation is complicated by local concentrations of hydrocarbons and NOx) in the model. My concern here is that, in order to compare simulated and measured ozone, transport and NOx concentrations in the model should be validated, or a validation paper using DEHM should be referenced in text.

SPECIFIC COMMENTS: How well does the model reproduce NOx conditions over the model domain and/or the EMEP measurement sites? This would be an important point in the discussion for isolating isoprene emissions as the major uncertainty in the model.

Reply:

“*Isoprene has a short lifetime, and hence it is very difficult to evaluate its emission estimates against measurements*” and “*Due to the scarcity of observed isoprene data and the short lifetime of isoprene:.. : an indirect evaluation was made based on measurements of ozone concentrations in Europe:*’’ We do not see any contradictions. Despite the fact that isoprene has a short lifetime and its simulation is accompanied with uncertainty, we have evaluated it to find whether the model results are in the right order of magnitude. But since there are just very limited measuring sites which have available isoprene data in the study period, we have compared simulated ozone and observation from the same network as observed isoprene database (EMEP). However, the number of stations with available ozone data in that period is much more (117 stations) than those for isoprene (9 stations). On the other hand, because the atmospheric data and the CTM model

configuration, except for the isoprene emission, remained the same for the two runs, the difference between the simulated ozone of the two runs must originate from the different isoprene emission models used. In other words, here we just want to figure out using which isoprene model results in a better agreement with observations compared to another isoprene model while other parts of CTM model are retained. Furthermore, Brandt et al. (2012) (cited in the paper) presented the evaluation of DEHM model results for the year 2006 compared to available measurements from the EMEP network for some of chemical species including NO_2 as well.

Reviewer:

The authors should make more of an effort to determine what makes the largest contribution to discrepancies between the two emission inventories. The authors conclude the results section by stating that "The better performance using MEGAN can be attributed to a better parameterization of environmental activity factors or more accurate emission factors in MEGAN." Considering that those are the only inputs for estimating isoprene emissions, this is an obvious statement that leaves the reader dissatisfied with the analysis. I recommend that the authors determine the largest contributor to the discrepancy (i.e. emission factors due to different land type maps used for example; or the scaling factors – that is the temperature or radiation or leaf age etc. used).

Reply:

Following the reviewer's comment, we have compared the activity factors of temperature and PAR separately for the two models (not shown in the manuscript). However, because of the difficulty in separating effects of LAI and PAR in the GEIA results, we have compared the environment activity factors (combined influence of changes of LAI, temperature and PAR on the isoprene emissions) of the two models. Figure 4b is added which shows the result and the text is reformulated accordingly.

Reviewer:

There are grammatical errors that should have been picked up, such as the use of "nitrate proxy" instead of "nitrogen oxide" on pg. 9259, line 18.

The figures include captions and colorbars that are only legible if the manuscript is viewed at 200-300 times its printable size.

Reply:

Typos/grammatical errors are now corrected and figure captions and colorbars are enlarged throughout the manuscript.

SPECIFIC COMMENTS:

Reviewer:

Why mention other isoprene emission inventories in the Introduction (BEIS series, se- BVOC and BEM, for example)? The paper focuses on evaluation of MEGAN and GEIA. It is not clear what inclusion of these emission inventories adds to the Introduction or analysis that follows.

Reply:

We have made the revisions suggested by the reviewer.

Reviewer:

The statement ": but not evaluated with observations." on pg. 9251, line 18-19 is not correct. Curci et al. (2010) compared satellite-derived isoprene emissions with MEGAN in Europe.

Minor COMMENTS: Pg. 9251, line 7: The list of references presented here should include Curci et al. (2010), which compares satellite-derived isoprene emissions with MEGAN in Europe.

Reply:

We thank specifically the Referee for this comment. We have changed the text in the Introduction, Result discussion and References sections.

Reviewer:

Be careful about using decisive statements such as that on page 9251, line 23. Arneth et al. (2008) carry out a detailed comparison of numerous isoprene emission inventories. Does your analysis offer the advantage of a hemispheric analysis (and here the authors should define what they mean by hemispheric, i.e. the model domain), with particular focus on the sensitivity of O3 in remote regions to changes in isoprene emission?

Reply:

We have rewritten this part of introduction and also mentioned the domain of study area in the text.

Reviewer:

On pg. 9254, line 12-13 the authors choose to not use the soil moisture parameterization in MEGAN, despite inclusion of water-limited regions such as the Sahel. They comment on the role soil moisture plays in increasing the discrepancy over these waterlimited regions (pg. 9257, lines 7-9). Please include a justification for exclusion of the soil moisture parameterization.

Reply:

Following the reviewer's comment, we have implemented soil moisture factor in the revised manuscript and modified the text and figures accordingly.

Reviewer:
On pg. 9255, line 15 the authors provide the annual isoprene emissions for the study area covering the northern hemisphere and much of the tropics. The emissions from MEGAN are close to the upper limit of the global estimates reported from previous studies (pg. 9255, line 16). What contribution does the excluded region in the southern hemisphere make to global emissions in the DEHM model? Wouldn't this lead to emission estimates that are higher than 770 Tg isoprene y-1?
Reply:
In the previous manuscript, we had an error to read LAI input data that caused this high value. The error is fixed in the current manuscript. Moreover, we have implemented soil moisture in the new version that has helped the total value to decrease. New simulated annual isoprene emission is 592 Tg/y which could not exceed 770 Tg/y when considering contributions of the excluded regions.

Reviewer:
Are the authors implying that on pg. 9257, line 19-20 MEGAN base emissions of shrubs are higher than those in GEIA, thus contributing to discrepancies in the African savannah and the subtropics? This sentence should be made clearer.
Reply:
We have decided to skip this conclusion because the evidence is not clear.

Reviewer:
Pg. 9258, paragraph 2: A description or figure of the sites listed in Tables 1 and 2 relevant to isoprene emissions would be useful. What vegetation types dominate these sites? What is the average leaf area index (annual or summertime) for these sites? Or are these sites described in a paper that the authors can reference in this section?
Reply:
As we mentioned, these measuring sites are from EMEP measuring network and have been addressed in the manuscript (http://www.nilu.no/projects/ccc/emepdata.html). Nevertheless, we have added a map of LAI in summer time for Europe and shown the location of the site on the map.

Reviewer:
On pg. 9258 line 28 the authors state that ": : :MEGAN results have a very small difference from observed mean values". However, in Table 1 the discrepancy is much larger for Peyrusse Vielle (France). The authors don't discuss this exceptional site. What contributes to the large discrepancy for this site?

Reply: We agree with the reviewer. The simulated mean values has the a larger different from observation that could be because of a few episodes of enhanced isoprene in France (Curci et al., 2010)

Reviewer:

Pg. 9262, line 12-14: MEGAN and GEIA are evaluated in Figure 12 in terms of their correlation with measurements. However, Figure 12 shows that the strength of MEGAN is its ability to capture day-to-day variability of daily average O3 and daily maximum O3. This should be included in the discussion of Figure 12.

Reply: We agree with the reviewer and have reformulated the text.

MINOR COMMENTS:

Reviewer:

Pg. 9249, line 12: ": : : inhabitant of agricultural products, : : :"does not make sense. Rewrite sentence or replace clause with ": : : crops, : : :".

Pg. 9251, line 31: Replace ": : : showed : : :" with ": : :presented: : :".

Pg. 9252, line 9: Provide latitude and longitude boundaries of the model domain.

Pg. 9253, line 13-14: This sentence does not make sense. Remove or alter.

Pg. 9253, line 27: Define PCEEA (i.e. provide the full name and a brief description).

Pg. 9254, line 29: The authors state that they focus on 2006 to coincide with the analysis year of Guenther et al. (2006). However, the analysis year in Guenther et al. (2006) is 2003. The publication year is 2006. Please fix this inconsistency.

Pg. 9257, line 26: Please specify the upper limit (in km or hPa) of the "lowest model layer".

Pg. 9258, line 25: Place the temporal variability information "3h" and "twice a week" earlier in the paragraph when the authors distinguish between Rigi, Switzerland and the other sites, so that it is clear what the authors mean by "time variant" on pg. 9258, line 15.

Pg. 9261, line 18: MEGAN is only marginally better than GEIA in the average statistics that are reported. This should be stated in text when describing Figure 11.

Figure 9: "Noisoprene" is not one word. Please fix.

Figures 1, 3, and 5: Please change the color scale of difference maps to that shown in Figure 9 for better visual clarity.

Figures 1, 3-7, 9-10, 12: Please increase the relative size of colorscales so that they are legible.

Figure 7: Please provide the altitude of the lowest model layer of DEHM in the title of this figure.

Reply: We have modified the manuscript according to the suggested points.

Reviewer:
Pg. 9262, line 4-6: Please justify why these two sites were selected for further analysis in preference to any other background sites.
Reply:
We have analyzed all of the 117 stations but had presented those two stations as an example on the Mediterranean region where the MEGAN had estimated better results. However, after the correction for reading LAI and implementation of soil moisture factor in the new version, we have not seen more differences on that area, so the figure has been substituted for average of all stations.

FIGURES:
Reviewer:
Figures 1 and 4: The unit "KT/grid/year" is not defined in text and not consistent with units used in other isoprene emission studies. Please change this to "atoms C cm-2 s-1" or "mg m-2 day-1".
Figure 2: Similar to the above comment "KT/day" is not comparable to other studies. Please change to be either "atoms C s-1" or "mg day-1".
Reply: we agree with the reviewer and have changed the unit in Figure 1 to $Tg/m^2/y$ and in Figure 2 to Tg/day.

Reviewer:
Figure 9: Bottom two left panel labels are ambiguous. Are the authors showing the percentage contribution of isoprene to ozone in DEHM after running DEHM with and without biogenic isoprene emissions? The labels imply that the bottom two left panels are difference plots. Please fix the labels and provide a clearer title for this figure.
Reply: We have clarified it in the figure legend

Reviewer:
Figure 11: (1) Start the x- and y-axes at _25-30 ppbv so that the data occupies more than 20% of the plot. (2) Is it necessary to plot labeled data points when it isn't possible to distinguish labels? Just include symbols in this figure.
Reply:
we agree with the reviewer about the start points of x- and y- axes and have fixed. Concerning the labeled points: they show the abbreviations of EMEP stations and we have also discussed about the result at some of the sites in the manuscript.