Response to Referee 1

We would like to thank the referee for the detailed review of our manuscript and for useful suggestions. Below we provide a point-by-point reply to the comments. The referee's comments are in red, while the authors' replies are in black.

Reviewer: It should be stated that for some of the emission types (e.g. Wildfires) the choice of inventory is not optimal considering what is freely available to the modelling community from data portals at the current time.

Reply: We agree with the reviewer; some inventories used in the current version of DEHM are not the latest inventories introduced. The main focus of this study was to improve the model for the simulation of BVOC emissions, SOA and sea salt concentrations. However, we have a plan to further improve DEHM by implementing the newest inventories especially for natural emissions like lightning and wildfires in the near future. New updates to the model can always be conducted; however, we think that handling all the different source categories will be too comprehensive in one paper.

The paper has 15 figures included making it quite long and the quality of writing is fair although the analysis is not robust enough in a number of places. Comparisons are made against measurements mainly in the US and Europe showing that even after the updates DEHM exhibits a low bias in PM concentrations, although potential reasons are not discussed in much detail but simply listed.

We think that the 15 figures are necessary for evaluation of all the different results. We improved the text in section 3.3 according to the suggested points.

A second part of the paper then moves towards the contribution of different 'natural' emission sources on tropospheric ozone at the surface, which has already received attention in the literature over the past decade as part of many independent studies.

As mentioned in the manuscript, and following the reviewer's comment, some previous studies have examined the contribution of natural emissions to ozone concentrations, but one of the advantages of this study is investigating the contribution of different kinds of natural emissions in one study and using the same air quality model. This gives us the ability to assess and compare the relative importance of natural sources with respect to the production of ozone and also fine particles, on a large scale, as well as individual selected regions in the Northern Hemisphere.

There is no validation of the surface O3 distribution included therefore the reader is left guessing as to quality of the model performance when using non-yearly specific emission inventories for e.g. the wildfire component.

Evaluations of the simulated ozone concentrations (by DEHM) against measurements from both Europe and North America, in the model domain have already been done by Brandt et al. (2012), Zare et al. (2012) and Solazzo et al. (2012a, b), which are mentioned and cited in the manuscript. Since further development of DEHM in this study only modified the final result of fine PM simulations, we showed evaluation of the modeled total fine PM against observations. That is conducted for both the previous and new model versions to assess improvement of the model performance with the updated description of natural emissions.

For publication in ACP I would therefore like there to be more focus on the missing components towards explaining the PM2.5 bias at the expense of removing the tropospheric ozone component. One

suggestion would be to utilise the additional sensitivity studies in terms of understanding and quantifying the most plausible explanations for the significant PM2.5 low bias in DEHM. This could also be extended for other regions using the e.g. AERONET network. I am finding it difficult to take away a new message from this manuscript or a direction in which emphasis should be applied to tackle the underestimation of PM concentrations which occur across a number of independent models.

Following the reviewer's comment and as mentioned in the conclusion section of the manuscript, one of the biggest concerns of the CTM modeling community is the underestimation of fine PM concentrations. In this regard, we also tried to improve our CTM model for simulation of PM concentrations. In this study, we have specifically focused on simulation of two individual compounds of SOAs and sea salt. However, the DEHM model is still underestimating the fine PM concentrations. Therefore we think, in regards to PM simulation, the good messages that can be taken away from our study can be summarized as below (mentioned in the manuscript);

"Model evaluations for the individual compounds of particles demonstrate that the simulated sea salt and secondary inorganic aerosols (Brandt et al., 2012) are in a good agreement with measurements. Limitations of the current model performance for fine particles in Europe can partly be related to missing windblown dust from the Sahara desert. However, the dust aerosols contribute mainly to the coarse fraction of PM, and a minor part to fine PM. On the other hand, the underestimation of SOA in Europe was about a factor of 2. We may therefore conclude that better estimations of SOA may improve the ability of the model to simulate fine PM levels, at least in Europe."

General Comments:

(i) The grammar should be checked throughout the manuscript as there are many instances where it is not correct.

We have made a thorough review of the grammar in the paper and revised accordingly.

(ii) The use of ozone and O3 should be consistent throughout the manuscript.

Done

(iii) The use of references could be significantly improved by using more recent studies especially concerning the new parameterizations, emission estimates introduced into DEHM and the many previous works concerning the influence of emission type on regional and global surface ozone distributions.

Done

(iv) The unit of ppb is associated with mixing ratios rather than concentrations which typically have the unit of μg m-3.

We have modified the manuscript accordingly.

(v) Although the method of introducing variability in biogenic emission estimates is sufficient the choice of averages for wildfire distribution and emission is not satisfactory for capturing the well documented inter-annual variability in wildfire emissions. Especially considering the importance of this emission source to PM2.5 distributions in e.g. Scandinavia

Emissions from vegetation fires in this study are taken from the RETRO inventory (Schultz et al., 2007). The database provides the emissions for a long period of 1960 to 2000 with spatial and temporal

resolutions of 0.5 degree × 0.5 degree and monthly, respectively. However, the CTM modeling community are recently using GFEDv2 database (van der Werf et al., 2006). The GFEDv2 inventory is based on satellite data and available with an 8-day time step but just for shorter period of 2001 to 2005. Moreover, the reason that gives us more confidence for using the RETRO inventory here is the study by Schultz et al. (2008), showing that the geographical distribution and seasonal pattern of fires for one selected year agree reasonably well with the results obtained by GFEDv2. However, we are also working on implementing the GFEDv3 inventory (van der Werf et al., 2010) into the DEHM model.

(vi) Although SOA is introduced into the model is it interactive with the photolysis via additional scattering and absorption of photolysing light? Please provide details.
No – this process in not implemented in the model, yet.

(vii) Wildfires are strongly influenced by agricultural practices in the tropics so this is cannot be defined as a Natural emission source as done throughout this manuscript.
In respond to the reviewer's comment, we should mention that biomass burning comprises the burning of living and dead vegetation. This includes domestic fires (like fuel wood and crop residue burning) as well as open vegetation fires (forest, shrub, grass and cropland burning). This study aims at quantifying natural emissions of air pollutants. Thus, only the open vegetation fire emission has been subject of this study and called wildfire emissions in the manuscript. However, wildfires can be controlled and uncontrolled burns of natural or anthropogenic origin in open vegetation (Schultz et al., 2007). For instance, Mollicone et al. (2006) estimate 87% of the forest fires to be caused by human influence at the boreal area. In general there are no distinctions between the origins of the wildfires in the inventory; therefore, in this study we consider roughly the open vegetation fires (wildfires) as a natural source of emissions.

Mollicone, D.; Hugh D.E. and Achard, F. (2006): Human role in Russian wild fires. NATURE, Vol 440, 23 March 2006.

Specific comments

Pg 16777,

- ln 4: Show that the exceedence of biogenic emissions over anthropogenic emissions occurs for the more recent emission estimates by updating references to e.g. Lamarque et al., 2010 and Guenther et al., 2012, which are used later on in the text. For what compounds does this occur?;
- ln 9: " : : : are expected to change in the future : : : ";
- ln 14-16: Update references e.g. direct comparisons of recent biogenic estimates for a number of BVOC species has been performed in Williams et al, 2013;
- ln 21: provide reference for modelling studies;
- ln 27/29: "natural O3" is O3 from natural sources? To help the reader please define what percentage changes are introduced.

Done

Pg 16779, ln 7-8: Use capitals before acronyms.
Done

Pg 16781,

- ln 2: The work of Price et al (1997) has been significantly updated in Ott et al (2010). For instance the estimate for the cloud-to-ground and cloud-to-cloud flash distribution has been modified for the northern hemisphere. Can the authors provide some reason for using the old method considering this is a recently implemented parameterization?

  In general, we agree with the reviewer and, as already mentioned, we are aware that newer inventories for LNO_x are recently released. In this study, the main focus is investigate and understand how an update of the BVOC emissions, SOA and sea salt concentrations will improve CMTs. We intend to implement the new inventories for LNOx and wildfires emissions in DEHM in near future.

  We also note that Price et al. (1997) is still the most commonly used method in the CTM community; e.g. in GEOS-Chem (Hudman et al., 2007), MOZART-4 (Emmons et al., 2010), and CAM-Chem (Lamarque et al., 2012). Apart from that, another recently published work (Koshak et al., 2010) states that CG lightning produces ten times more NOx per flash than IC lightning, same as the estimated ratio by Price et al. (1997).

  - *Emmons, L. K.,Walters, S., Hess, P. G., Lamarque, J.-F., Pfister, G. G., Fillmore, D., Granier, C., Guenther, A., Kinnison, D., Laepple, T., Orlando, J., Tie, X., Tyndall, G., Wiedinmyer, C., Baughcum, S. L., and Kloster, S.: Description and evaluation of the Model for Ozone and Related chemical Tracers, version 4 (MOZART-4), Geosci. Model Dev., 3, 43–67, doi:10.5194/gmd-3-43-2010, 2010. 3495.*
  - *Hudman, R. C., Jacob, D. J., Turquety, S., Leibensperger, E. M., Murray, L. T., Wu, S., Gilliland, A. B., Avery, M., Bertram, T. H., Brune, W., Cohen, R. C., Dibb, J. E., Flocke, F. M., Fried, A., Holloway, J., Neuman, J. A., Orville, R., Perring, A., Ren, X., Sachse, G. W., Singh, H. B., Swanson, A., and Wooldridge, P. J.: Surface and lightning sources of nitrogen oxides over the United States: magnitudes, chemical evolution, and outflow, J. Geophys. Res., 112, D12S05, doi:10.1029/2006JD007912, 2007. 3495.*
  - *Lamarque, J.-F., Emmons, L. K., Hess, P. G., Kinnison, D. E., Tilmes, S., Vitt, F., Heald, C. L., Holland, E. A., Lauritzen, P. H., Neu, 5 J., Orlando, J. J., Rasch, P. J., and Tyndall, G. K.: CAMchem: description and evaluation of interactive atmospheric chemistry in the Community Earth System Model, Geosci. Model Dev., 5, 369–411, doi:10.5194/gmd-5-369-2012, 2012. 3495.*
  - *Koshak, W. J., Peterson, H. S., McCaul, E. W., and Biazar, A.: Estimates of the lightning NOx profile in the vicinity of the North Alabama Lightning Mapping Array, Proc. of the 2010 Int. Lightning Detection Conf., Orlando, Florida, 2010.*

  Can the authors discuss what impact using a 8 year average for have on predicting the inter-annual variability in LNOx during 2006. ;

  In this specific study, we only look at the annual mean value of LNOx emission to assess its contribution to annual mean ozone concentrations in the model domain; therefore we have not gone through the inter-annual variability here. Since the inventories for LNOx generally are very uncertain, using an 8 year average for the year 2006 can probably be as good as most other estimates.

- ln 8 The range given in Schumann and Huntrieser (2007) is 5±3 Tg N/yr rather than 1-20 Tg N/yr. Please correct.;

Even though, the annual LNOx emission estimated by Schumann and Huntrieser (2007) is 5±3 Tg in global, they reviewed many studies and showed that the best estimation from the majority of studies since the mid-90s assumed a value of about 5 Tg yr^{-1}, with a range of 1–20 Tg yr^{-1}. We clarified it in the manuscript.

- ln 20: replace "the model" with DEHM;
  "The model" in this sentence refers to the empirical model of soil-biogenic NO_x emission introduced by Yienger and Levy (1995). We clarified it in the manuscript.

- ln 25: How does the global annual emission compare to the estimates given in Steinkamp et al. (2009)?
  Yienger and Levy (1995) calculated the global annual SNO_x emissions of 5.5 TgN with a range of 3.3-7.7 TgN, while Steinkamp et al. (2009), by using an algorithm based on the Yienger and Levy (1995) parameterization with slight modification, estimated the global emissions of 9.74 TgN/yr which is larger than the estimated range by Yienger and Levy (1995). However, both estimations are in the ranges from 4 to 21 Tg(N) mentioned by Steinkamp et al. (2009) from other studies.

Pg 16782,

- ln 3: why not use the NH3 anthropogenic estimates provided in Lamarque et al (2010) in order to maintain a consistent set of emissions;
  In fact, we did, we have used the RCP dataset (Lamarque et al (2010)) for all anthropogenic sources. Now, we have modified the text to clarify this matter.

- ln 13: what is the reference for the Wild Animal NH3 emissions totals and what is the corresponding value for Domestic animals?
  As mentioned in the manuscript, global emission value of NH_3 from the source of wild animals is provided by the inventory introduced by Bouwman et al. (1997). According to this inventory, the corresponding value for domestic animals is estimated to be 21.6 TgN yr^{-1}.

- ln 15: replace "released" with emitted, plural of wildfire
  Done

Pg 16783,

ln 4-9: Most state-of-the-art global CTM's use monthly mean distributions from wildfires for specific years from e.g. GFEDv2. Why do the authors not use these readily available estimates for 2006?
This matter is responded above.

What type of injection heights are used for introducing fire emissions (e.g. Dentener et al., 2006)?
The fire emissions are vertically distributed unevenly up to 5km above surface with main part between 1000m and 3500m and with a maximum at 2000m.

The year 1997 had a strong signal due to El-Nino in the intensity of fires therefore may not be directly applicable to a more normal year such as 2006.

The year 1997 in this sentence is referred to the Schultz et al. (2008) study, in which they showed that in this year results for geographical distribution and seasonal pattern of fires from RETRO inventory agree reasonably well with the results obtained by GFEDv2.

Pg 16784, ln 25: ": : :HTs, Henze and Seinfeld (2006) : : :."
Pg 16786, ln 5: remove comma after salt and DEHM
Done

Pg 16787,

- ln 19-20: Mention the effects on the strong seasonal cycle in NH biogenic emissions on the annual surface averages (i.e.) are the maximal mixing ratios similar during boreal summertime?;
  We have modified the text according to the reviewer's comment.

- ln 25: replace summation with 'cumulative total".
- Define the latitude and longitude values of the domain.
  Done

Pg 16788,

- ln 3: A range of global monoterpene emissions is provided in Arneth et al., 2008. How does the annual total in DEHM lie within this range?
  Arneth et al. (2008) provided an overview of studies related to global monoterpene emission estimates, in a table, which shows that the majority of studies (5 from 9) calculated the total annual values in range of 103-127 Tg, although the values of 32, 33, 72 and 99 Tg are also estimated by other studies. Considering our model domain (from 10°S to 90°N), we can say that our annual estimated value of 88.4 Tg for monoterpenes emissions is consistent with most of previous studies overviewed by Arneth et al. (2008).

- ln 9: replace "validate" with compared;
- ln 35: ": : : with corresponding the measurements "
  Done

- ln 10-11: provide latitude and longitude for each site as done for Thompson Farm;
  Although they are provided in the table 2, we have addressed them in the manuscript as well.

- ln 13: "in time periods". This should be replaced by "days of the year" as time periods infers a specific year;
  In fact, we wrote the phrase "in time periods" on purpose, because not only the days of years with available measurements are not similar in these sites, but also the years are different. Therefore, we did the evaluation for each site during the periods for which measurements were available.

Pg 16789,

- ln 1-2: Although the relative magnitudes are similar a correlation co-efficient of 0.34 shows that the timing of the emission is quite different. Please add a sentence regarding this in the text.;
  Done

- ln 15: the simulation is for 2006 therefore the comparison is not for corresponding years as currently described in the text;
  Although most parts of this modeling study have been carried out for the year 2006, but in this part (comparison of simulated monoterpene concentrations with the observations), due to lack of measured data, we had to simulate and do the evaluation for the periods which correspond to each time period with available data at each site. These comparisons are done for Hyytiälä in 2006, for Vielsalm in 2010 and for Petersham in 2005. However, for the station in Durham there are available measured data for many years from 2004 to 2009, therefore the comparison is done for all the years (in table 2) and we only showed the temporal variations for the year 2008 as an example (figure 3).

- Introduce Figure 3, and the related discussion, immediately after Tables 1 and 2. Move more general reasons for the mismatch to the end of Section 3.1.;
- ln 23 replace "East Asia" with South-East Asia, replace "on both" with during;
- ln 25: Higher SOA in the NH due to higher temperatures, the tropics only has a small seasonal cycle.
  We have modified the text accordingly.

Pg 16790,

- ln 1: What is the difference in temperature in the tropics between January and July? There is little seasonal cycle in monoterpene emissions in the tropics.
  Zare et al. (2012) made available mean temperature patterns of the lowest model layer provided by MM5v3.7 in 2006 for both summer (JJA) and winter (DJF) times which are used in this study as well. In addition to displaying the differences in mean of temperatures between these two seasons, Zare et al. (2012) indicate that there are differences in the modeled BVOC emissions (by MEGAN) between these two seasons, even in some parts of tropics.

- ln 3: there is a stark contrast between Africa and South America with respect to SOA concentrations. Can the authors explain why considering the distribution of vegetation and monoterpenes (probably related to POA distribution)?
  Following reviewer's idea and as also discussed in the manuscript and shown in figure 5, we can say that although the high SOA concentrations are seen in the tropical regions, both in Africa and South America, with the highest emissions of precursors, the availability of POAs as a surface for condensation of SOAs also plays an important role in the distribution of SOA concentrations. In other words, since the POA concentrations are high in regions with strong wildfires such as Africa's savannah, the SOA concentrations are higher in these regions compared to South America.

- ln 5-6: remove anthropogenic from the discussion if negligible. This could be confusing and misinterpreted as a link between anthropogenic emissions and SOA.
  Done

- ln 10: Has POA been defined?
  Yes, that has been already done in page 15785, line 10.

- ln 18: “tropic” should be “equatorial regions”;
- ln 10-24: the discussion here is not clear and should be re-written.
- ln 29: repalce “the model” with DEHM
  Done

Pg 16791,

- ln 3-4: Given that the total global SOA in DEHM is near the lower limit of 2 Tg/yr surely it is not so surprising that the model underestimates the measurements. What is the corresponding global total of POA’s and how does this compare with the literature values?
  Hoyle et al. (2007), using an inventory for emissions from fossil/biofuel combustion and GFEDv2 inventory for the emissions of OA from biomass burning, estimated the total global value of approximately 23 TgC/yr for POA emissions. However, another modeling study by Heald et al. (2008), using fossil fuel emissions from the EDGAR v2.0 inventory [Olivier et al., 1996] and Biomass burning emissions in the tropics and the extratropics, calculated the global value of 45 TgC/yr for the POA emissions. Considering our model domain (from 10°S to 90°N), we might be able to say that our annual estimated value of 24.4 TgC for POA emissions, in the DEHM model, is in the estimated range by some previous studies.

- Ln 5-6: Is this probably due to good meteorology driving MEGAN?
  Yes, it could be. We have modified the text to state this matter too.

- Ln 3-14: The findings related to OA could be interpreted in a better way by segregating the US into e.g. East and West and showing the corresponding correlations. This would add to the discussion as POA will be higher in the East than the West. This could be done by introducing an additional Table which focuses on the correlation co-efficient and biases in different states similar to what is done for Europe.
- Fig 7: Using a total average across the US masks the fact that the agreement in the West seems better than in the East.
  Following the reviewer’s comment, we have plotted two different figures that show correlation coefficient and fractional biases as well as annual mean of OA simulations and observations for individual sites (see figures 1a and 1b, below). We have classified the stations to eastern, western and central US stations. Corresponding averages of the parameters are mentioned at the top of each plot. In general, the plots do not help us conclude “that the agreement in the West seems better than in the East”. Moreover, underestimation of OA concentrations in majority of the western sites are more pronounced than in the eastern sites. However, the simulated OA concentrations in the west seem to follow the observations better with fairly higher correlation coefficient. Therefore, we do not think that including this matter in the manuscript will provide more information.

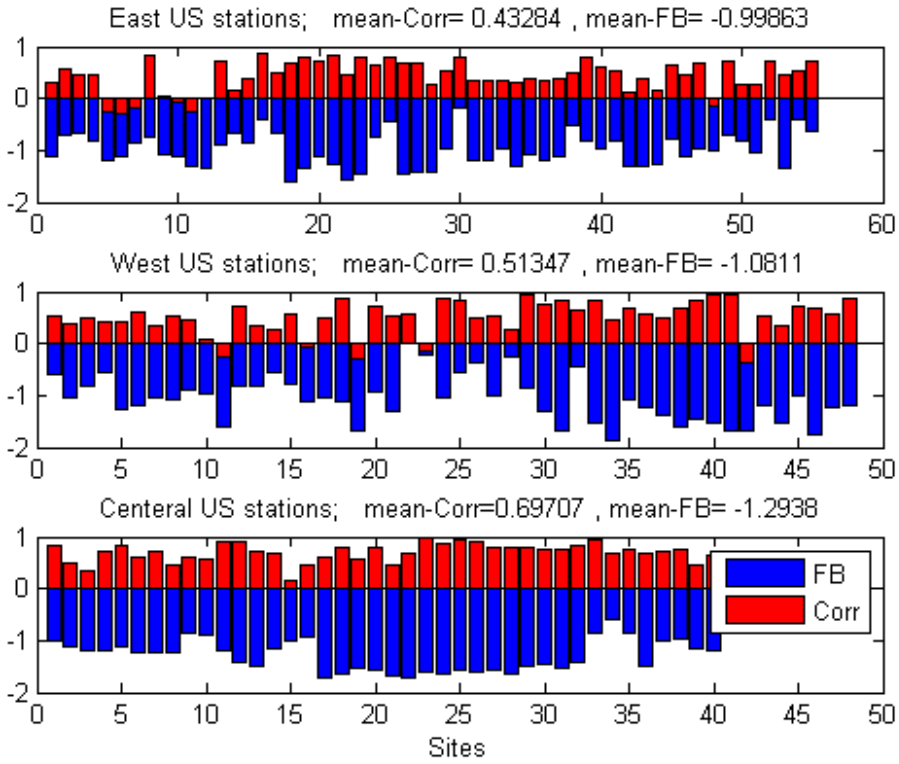

Figure 1a: Correlation coefficient and fractional biases for individual sites from eastern, western and central US between annual of OA simulations and observations.

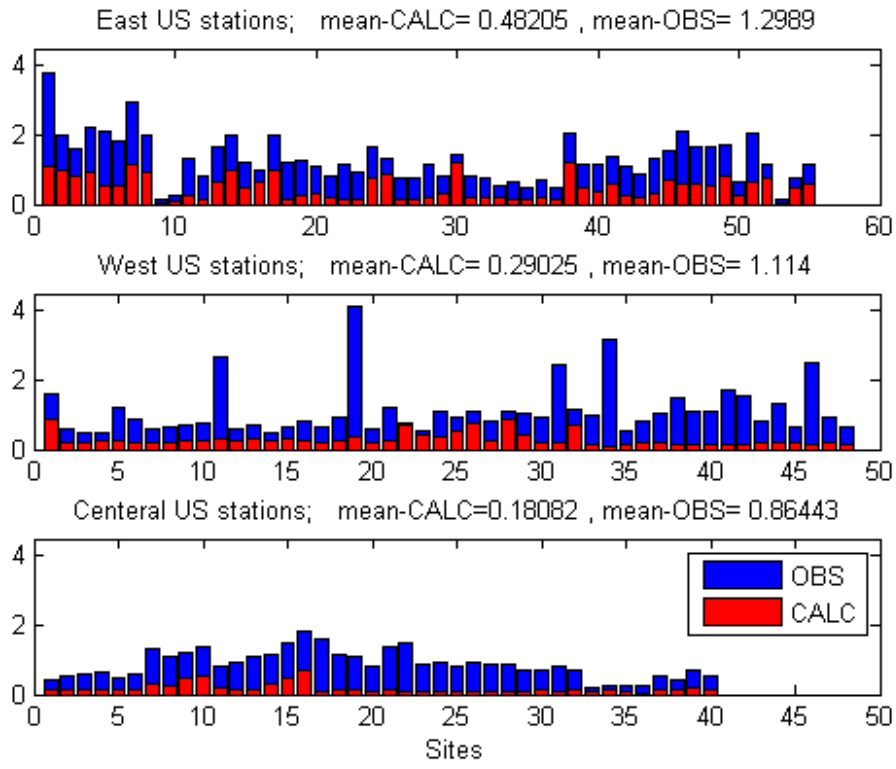

Figure 1b: Annual mean of OA simulations and observations for individual sites from eastern, western and central US.

Pg 16793,

- ln 1-7: The authors should comment on the differences in agreement between seasons and potential causes of the significant underestimation during the wintertime. From figure 9 it can be assumed that the sea-salt distribution is not the cause of the under-estimation but rather the other PM components in the system. It has been shown before that SOA contributes between 1-8% of OA therefore the missing component seems related to one of the other aerosol types. For instance how good are the BC emissions? Further discussion should be added on such details.
  The text has been modified according to the reviewers comment.

- Sect 4: Has there been some past validation of the surface O3 distribution in DEHM over Europe in previous work? Please add a few sentences related to this.

Yes, ozone evaluation has been done in some previous studies such as Zare et al. (2012) and Brandt et al. (2012) which are mentioned in the manuscript (page 15801, line 24). However, we now revised the text to make it clear in page 15779 line 5.

- Ln 11: "the latter simulation" : : : is this the BASE simulation?
  It refers to "control simulation" which considers all emission sources including both anthropogenic and natural source types. We revised the text to make it clear.

Pg 16794,

- ln 10: Unless you are able to differentiate between wildfires and biomass burning then the burning component is not strictly natural but mostly related to agricultural practises;
  This matter is responded above.

- ln 22-25: there have been many studies related to the influence of lightning on surface O3 such as Zhang et al. (2003), Aghedo et al (2007), Holmes et al. (2013) so this sentence is not correct and should be modified.
  Although we cannot see any contradiction with the results of the studies mentioned by the reviewer and our results, we modified the text slightly and also cited the suggested references to the manuscript. Below you can see some parts of the reviewer's referenced studies which can prove our claim:
  Aghedo et al. (2007): "*The impact of lightning on surface ozone is negligible, while anthropogenic emissions yield a maximum of….*" Or "*Lightning has a small impact on surface ozone concentration, and only accounts for a maximum of 1 ppbv over a small area in Africa, and is therefore not presented.*"
  Zhang et al. (2003): "*Summer lightning is shown to play a dominant role in controlling NO_x and O_3 concentrations in the middle and upper troposphere, …*", Or " *The largest lightning impact on the upper tropospheric O_3 budget reaches ≈30% in August. Below 5 km O_3 production is dominated by surface NO_x emissions, with the lightning contribution reaching 20% in August*."

Pg 16798: The observational data in Fig 10 could be segregated into different regional components e.g. Scandinavia and the different sensitivity tests related to e.g. wildfires used for a better quantification of the contribution. The inter-annual variability in wildfire incidence will be important for capturing events though.

The reviewer's suggestion regarding different sensitivity tests is interesting. However, as already mentioned, our main focus is updating the DEHM model for simulation of individual compounds of only SOA and sea salt, and thereafter improving simulated fine PM results. In other words, in Fig. 10 (a) and (b), inventories of other natural emissions such as wildfires and lightning are kept identical, and simulated fine PM results compared to observations are improved only due to updating simulations of SOA and sea salt aerosol.

To assess the contributions of these two updated individual aerosol components in the improvement of simulated PM results, especially in different regions, we have repeated the evaluation of the simulated total $PM_{2.5}$ concentrations against measured data at different stations in the EMEP network. **Figure 2-5** shows the evaluations of simulated $PM_{2.5}$ concentrations from **(top)** the recent, and **(middle)** the revised versions of DEHM with updating just sea salt simulation, and **(bottom)** also with updating both sea salt and SOA simulations, for four different stations in different countries as examples.

In fact, we could not find any specific and consistent results, for sites located in the same regions, with regards to the contributions of updating sea salt or SOA simulations in improvement of the simulated total fine PM results. However, first with updating sea salt simulation in DEHM and then implementing a scheme for SOA simulation too, the improvement of simulated total fine PM results show increasing rate.

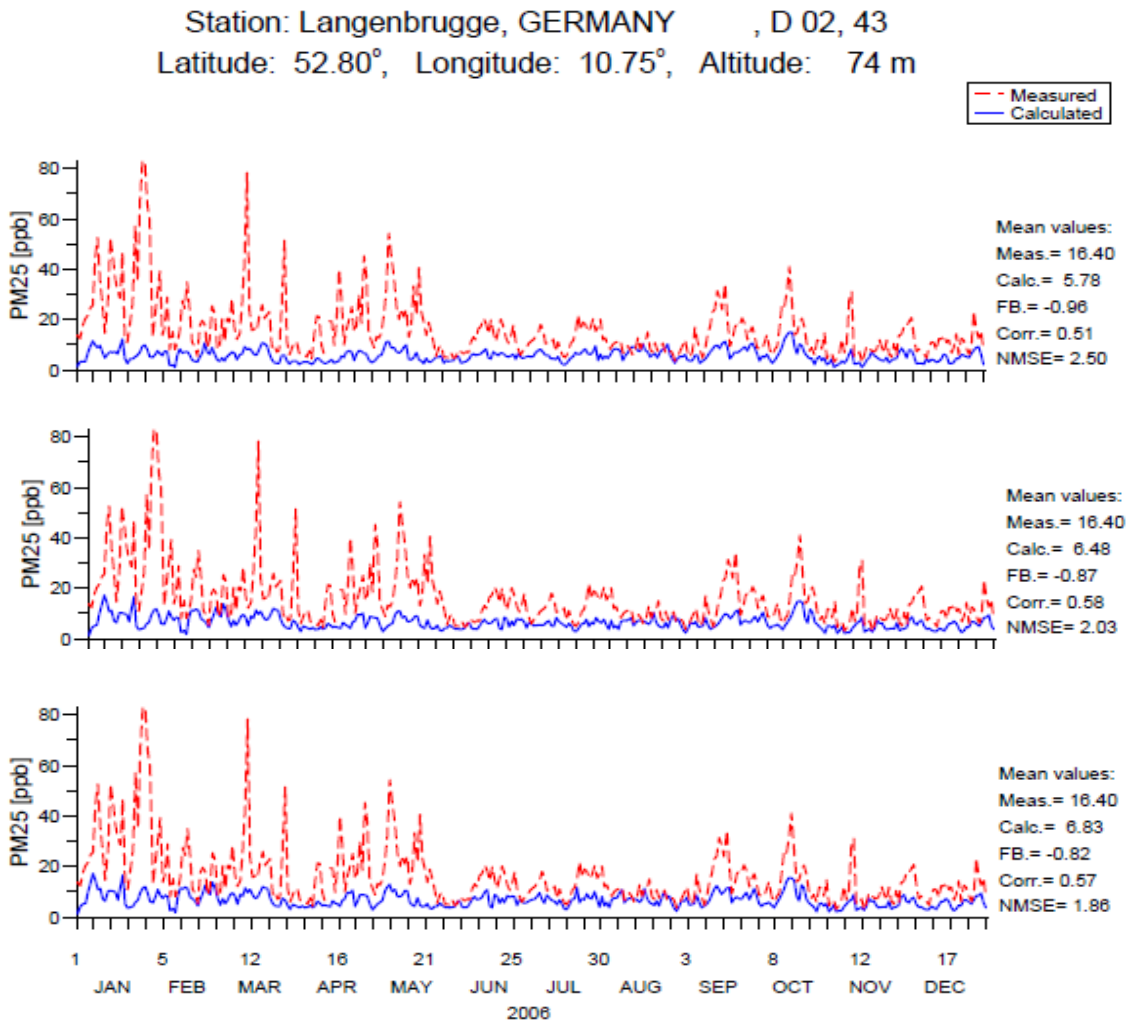

Figure 2: Evaluation of the simulated total PM2.5 concentrations against measured data for Langenbrugge station in Germany. **(Top) S**imulated PM2.5 concentrations are provided by the recent versions of DEHM, and **(middle)** the revised versions of DEHM with updating just sea salt simulation, and **(bottom)** with updating both sea salt and SOA simulations

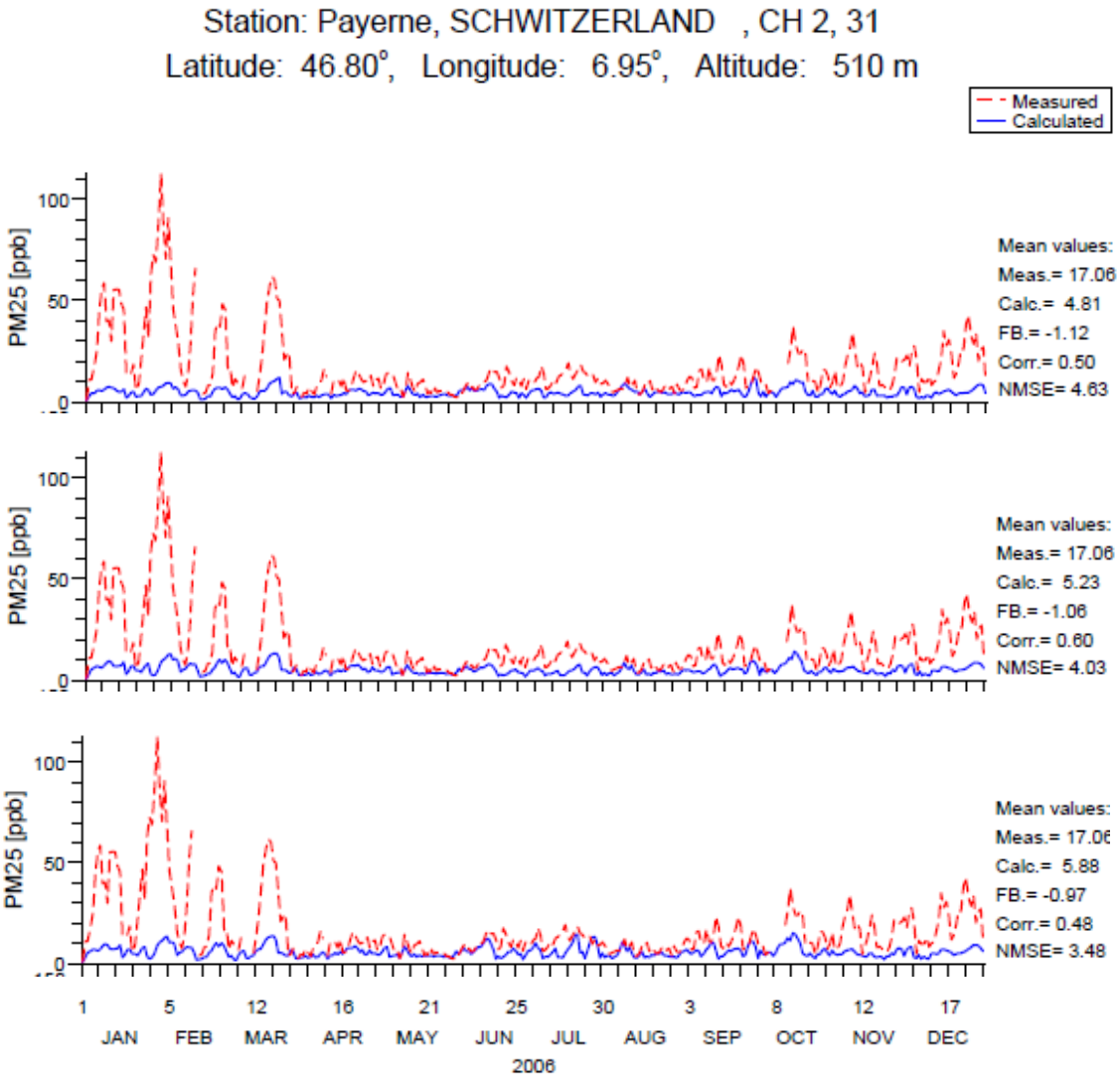

Figure 3. : Evaluation of the simulated total PM2.5 concentrations against measured data for Payerne station in Switzerland. **(Top) S**imulated PM2.5 concentrations are provided by the recent versions of DEHM, and **(middle)** the revised versions of DEHM with updating just sea salt simulation, and **(bottom)** with updating both sea salt and SOA simulations

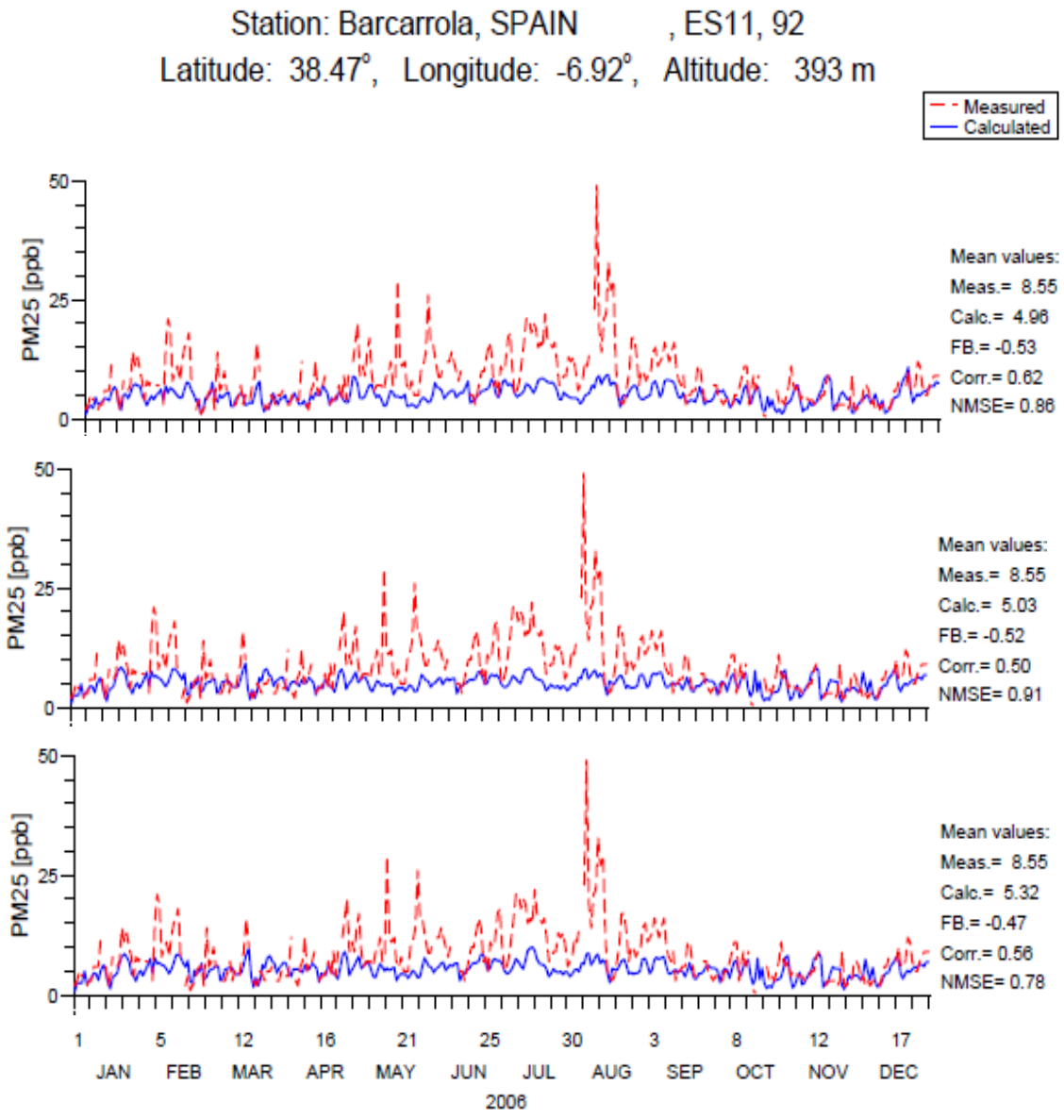

Figure 4: Evaluation of the simulated total PM2.5 concentrations against measured data for Barcarrola station in Spain **(Top) S**imulated PM2.5 concentrations are provided by the recent versions of DEHM, and **(middle)** the revised versions of DEHM with updating just sea salt simulation, and **(bottom)** with updating both sea salt and SOA simulations

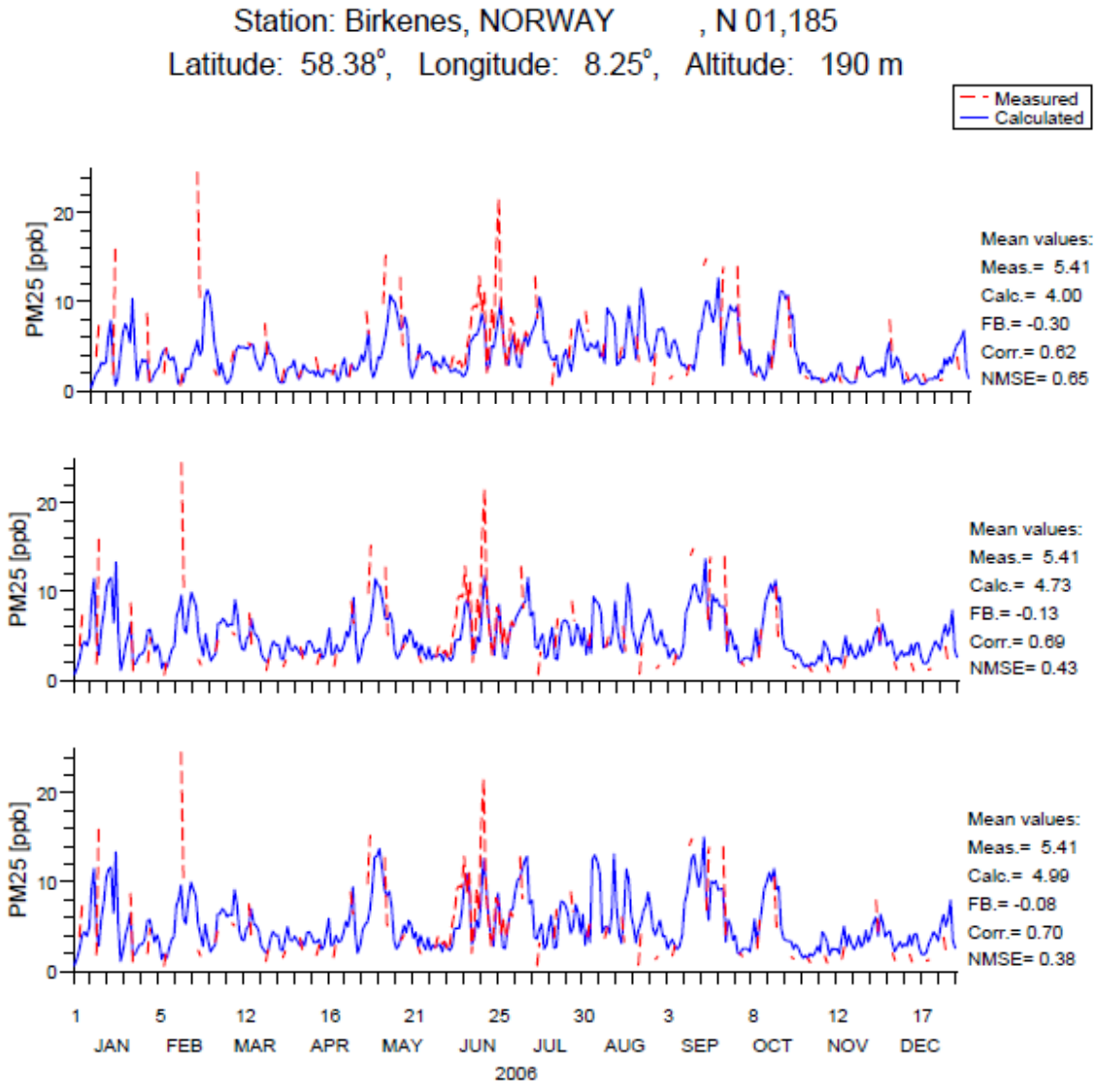

Figure 5: Evaluation of the simulated total PM2.5 concentrations against measured data for Birkenes station in Norway. **(Top) S**imulated PM2.5 concentrations are provided by the recent versions of DEHM, and **(middle)** the revised versions of DEHM with updating just sea salt simulation, and **(bottom)** with updating both sea salt and SOA simulations

Pg 16799,

- ln 1-2: Please include the latitude and longitude limits of each region;
  Although we also liked to do it, in fact we think that inserting all latitudes and longitudes for the subdomains in the text, especially for Africa with 8 corners, may not provide further information for the reader. Therefore, we tried to display the extent of these regions by figure 15.
- ln 23-next page: This section can be significantly shortened to a few sentences as the effects of non-linear O3 chemistry are well known in global modelling.
  Done

Pg 16801,

- ln 22-24: This sentence should be moved out of the conclusions to the discussion.
  Done
- Can the previous comparisons be valid for these simulations which adopt different emission inventories for NOx?
  Yes, the referenced studies of Brandt et al. (2012) and Zare et al. (2012) in the manuscript have used the same inventory for NO_x emission as the one used in this study.

Pg 16802, Final paragraph: Again there is new discussion arising related to the underprediction of PM2.5 in Europe which should not be in the conclusions but in the appropriate section.
Although we already discussed the underestimation of PM in the results section, the aim of authors to bring this phrase here, as the last paragraph of the manuscript, is to express their motivation for future works regards to promote the underestimation of PM in Europe.

Tables:

- General: Replace "NO. data" with 'Number of observations'.
- Table 4: replace "Lightnings" with "Lightning"
  Done

Figures

- Fig 2: Use a logarithmic Y-axis to allow better visibility between the different types of monoterpenes. The units should be Tg species/yr.
- Fig 4: The colour scale needs changing to 0.1 between 0.1-0.7 as there is currently too much green which hides the variability in e.g. Europe.
- Fig 9: Legend ": : : a combined source functions : : : "
- Fig 11: Add "No-NE" to the panel relating to the sensitivity simulation and 'Control" to the BASE panel relating to the simulation. Also "relative percentage differences" .
  Done

- Fig 7: Using a total average across the US masks the fact that the agreement in the West seems better than in the East.
  This issue has already been answered above.