

## Response to Referee 2

We are grateful to the Referee for the positive evaluation of our paper. All comments and suggestions are carefully addressed in the revised manuscript. Below we provide our point-to-point responses. The referee's comments are in red, while the authors' replies are in black.

### General comments:

Despite I am not a native speaker, I have detected some mistakes in the grammar, that has to be consistently checked throughout the manuscript.

We have thoroughly gone through the document to correct the grammar.

My main concern is related to the validation of the simulations. The authors have performed an interest study on how to improve the annual simulations of air quality over the northern hemisphere; however, to make sure that there is a real improvement in the simulations, an extensive validation against measurements have to be performed. I know the availability of measurements is very scarce for several species (e.g. monoterpenes), but I have strong doubts that considering just 2 stations in Europe and 2 in the United States can provide the validation needed for this type of study.

We completely agree with the reviewer and of course we wish we were able to evaluate our simulation results with more observations, but unfortunately the number of sites with available measured data for BVOC emissions is very limited. We are really thankful to Karl B Haase (University of New Hampshire), Bernard Heinesch (University of Liège), Taina Ruuskanen (University of Helsinki) and Karena McKinney (Amherst College, MA) for sharing the measurement data of monoterpenes for this study. The data are for different years and periods and we had to do this evaluation for time periods corresponding to the periods of measurements in the individual sites.

Moreover, I did not find any validation of tropospheric ozone in the manuscript. I have no doubts about the improvement in the emissions included in the DEHM model, but I have no information about whether these improvements lead directly to an improvement in ozone predictions or not.

Evaluations of the simulated ozone concentrations (by DEHM) against measurements from both Europe and North America, in the model domain have been already carried out and the results are presented by Brandt et al. (2012), Zare et al. (2012) and Solazzo et al. (2012a, b); all cited in the manuscript. Concerning further development of DEHM, we only modified the result of fine PM simulations in this study (due to simulations of SOA and sea salt), and only showed the results of comparison of the total fine PM concentrations against observations, connecting to this model improvement.

Emissions included in DEHM come from different sources, use different methodologies, are gridded at different resolutions: : : How could you assure the consistency between those different databases and methodologies?

We are using the best available emission data for each resolution and area in the model. We do not consider the different resolutions a matter of concern, since all emissions are integrated into our model projection, with mass consistency. Furthermore, all emission sectors are only represented once, due to consistency in the common SNAP emission definition.

In addition, I am not sure if using emissions inventories representative for a climatological period (that is, non-yearly specific emission inventories) can lead to an improvement in the simulations for a specific year of study (year 2006). I know it is computationally demanding, but including a longer period would contribute to remove the yearly-specificity in the results presented.

We generally agree with the reviewer that some of the inventories used in this study are non-yearly specific inventories, but with regards to further development of DEHM in this study, we have improved the BVOC, SOA and sea salt simulations for which we used the yearly specific emission inventories.

Last, for PM<sub>2.5</sub> the authors find a reduced bias when implementing the aforementioned developments in DEHM, but I really miss a detailed discussion on how this bias is reduced. So the reader is left with a lack of information on what the motives for improved simulations are. In this sense, conclusions are not very conclusive. I would expect a deeper analysis of the implications of this study for the state of the art and not a summary of what I've read before in the manuscript. The authors should elaborate on this a little bit more.

We enriched the text in section 3.3 according to the suggested points.

Minor comments:

The authors have to be very careful when using ppbV, since this is not a concentration unit, but is used for expressing mixing ratios. So I would use micrograms per cubic meter instead of ppbV when referring to concentration of trace gases.

We have modified the manuscript according to the suggested points.

The authors should be consistent with the use of "O<sub>3</sub>" and "ozone" throughout the text.

Done

The figures and tables in the manuscript are also relevant, but the quality of some figures should be improved before final publication (as stated below).

- Figure 2 is really hard to read; have the authors considered using a double Y-axis (or a logarithmic Y-axis) so the readers can appreciate the behaviour of those monoterpenes with low annual fluxes?
- The colour scale in Figure 4 should be changed, since the readers cannot appreciate any colour differences between 0.1 and 0.7 (concentrations of SOA).

Done

I cannot find any reference on the text to Figure 15; so if the authors want to keep this Figure, as it defines the different domains analysed, I would cite in the methodological part, together with the description of the modelling/analysis features

The figure has been pointed to in the text in page 16799 line 2 where we described the extent of each continental regions studied in tables 4 and 5.