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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Response of fine particulate matter concentrations to changes of emissions and temperature in Europe" *by* A. G. Megaritis et al.

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Main comments:

1. *The paper presents the results of a series of sensitivity simulations with the PMCAMx model. The information provided in this paper is of interest to the community and especially for those that work on emission control strategies. However, I feel that the paper can be largely improved through an improved model evaluation, an emphasis on the broader picture instead of all the separate runs and the discussion of the results e.g. in comparison to others.*

We would like to thank the reviewer for the constructive comments and for the resulting

improvements of our paper. Several sections of the manuscript have been now reexamined and rewritten. The section describing the model sensitivity results has been rewritten and we have added information on the model evaluation that has been discussed in other publications. The section where the results of this study are presented was reorganized focusing on the effectiveness of each emission control strategy. We have also extended the comparison of our results with other studies in Europe and the US.

2. *The evaluation for PMCAMx is extensively performed for the US. However, for Europe hardly any evaluation is available. As climatological regimes and emission regimes are quite different from the US and input data are differently defined in European practices a thorough evaluation seems necessary, prior to using the model for detailed sensitivity studies. The mentioned AMS campaigns cover only a few weeks in total and do not cover all PM components. Moreover, they do not provide a good coverage across Europe. I wonder why a comparison to chemical data from the EMEP network and PM data from AIRBASE is not presented for the period at hand. Inclusion of a standard statistical is therefore advised.*

We agree that while the performance of PMCAMx in the US and Mexico City has provided useful information about its capabilities and limitations (Karydis et al., 2007; Murphy and Pandis, 2009; Karydis et al., 2011; Tsimpidi et al., 2010; Tsimpidi et al., 2011) that additional evaluation is needed for Europe for the periods that are the focus of the present study. This detailed model evaluation has been presented in other work (Fountoukis et al., 2011). In that study, we compared the model predictions against hourly average ground measurements, taken at four European measurements stations, as well as airborne measurements from aircraft flying over Europe. The evaluation was based on approximately 8500 measurements and examined the performance of the model on an hourly basis over an extended area. The area covered (thanks to the airborne measurements) and the number of data points for the various PM components make this one of the most extensive evaluations of a CTM over Europe. Our evaluation

focused on the AMS measurements and not on routine PM10 measurements because of the issues involved in the interpretation of the latter (Rees et al., 2004; Tsyro, 2005). These issues involve problems with the significant amounts of water that is often involved in the PM10 measurements in Europe and the significant contribution of the difficult to simulate dust in these concentrations. We do agree with the reviewer that additional information about its performance is needed and we have added a summary of the evaluation results in the main paper and some additional summary evaluation statistics in the Supplementary Information.

3. *Chapter 4 is very lengthy. For each simulation the results are discussed per component. This is also shown in the conclusion section. I would favour a more condensed presentation of the results and an interpretation of the comparison of the different control options and the fact that these may have different impacts and order of effectiveness in different parts of Europe.*

We followed the reviewer's suggestion and rewrote Section 4 reorganizing the presentation of the results and condensing the corresponding information presented there. Some of the details have been moved to the Supplementary material. We have also rewritten the corresponding parts of the Conclusions section avoiding the details and focusing on the most effective strategies.

4. *The study only shows control strategies for single components. It should at least be shortly discussed which combinations of pollutant reductions might be promising considering the outcome of this study and the nonlinearity's that may occur.*

This is a valid point. We have added some discussion about the combination of strategies and the potential nonlinearities. We do plan to examine these as parts of realistic strategies in future work.

5. *Many absolute and relative changes are mentioned. These may not show the same*

patterns and maybe should be separated a bit more.

We have done our best in rewriting Section 4 to separate (or contrast where appropriate) the corresponding absolute and relative changes of the PM concentrations in order to avoid any confusion.

6. *A discussion of the results is missing. Now and then other studies are named but their outcome is not given and is not set in relation to the outcome of this study. No short comings of this study are discussed, nor are recommendations given. Is the coupling of the different components in the atmosphere 100 percent represented by the model? Are all needed components modeled? What are the most uncertain parts of our knowledge and the model description? Which results are robust, which are less certain, which are contradicting other studies?*

There a lot of good questions in the above comment. We have added a paragraph in the end of the manuscript discussing the weaknesses of the model application (emissions, model uncertainties for example in the treatment of OA) and the corresponding confidence of the predictions. We have also added some discussion about potential missing couplings and processes. Finally we have extended the discussion of the comparison of our results with other studies in the same domain but also in other areas.

7. *The selected periods are not typical periods for winter and summer conditions in Europe. I think both may also be characterized as spring. A discussion on the representativity of the simulations for longer periods appears to be needed.*

Both periods included a variety of meteorological conditions and pollution levels. For example, the first half of May was characterized by a blocking anticyclone leading to stable meteorological conditions and high pollution levels over Central Europe. Hamburger et al. (2011) have provided an extended analysis of the synoptic and pollution situation over Europe during this period. The high temperatures observed in most of northern and southern Europe are characteristic of early summer. The winter period

was relatively typical. A summary of the meteorological conditions and references to the corresponding EUCAARI papers summarizing the conditions during the corresponding campaigns have been added to the revised manuscript.

Detailed Comments

8. *General: Please write out all abbreviations at first usage (OA, POA, VOC, etc).*

Done. In the revised manuscript we have defined all abbreviations used.

9. *Abstract, last sentence: Please rephrase as temperature is not a controlled variable.*

The last sentence of the abstract has been rephrased.

10. *Introduction, page 3, line 3-24: The text on the impact of climate change on air quality focuses on ozone completely, whereas PM is under emphasis in the paper. There is information on particulate matter too. Could you incorporate this too and maybe shorten the ozone discussion a bit?*

We have followed the reviewer's suggestion and we rewrite this paragraph. In the revised manuscript we added more information on the impact that climate may has on particulate matter, mentioning results from recent studies. We have also shortened the discussion about the effect of climate change on ozone levels.

11. *Introduction, page 4, line 29: here it is claimed that PMCAMx can accurately and efficiently describe the physical and chemical transformations of gas and aerosol species. This sounds very confident as many challenges are left in the modeling of PM and precursor species. Moreover, when very efficiently why not model a 1-5 year period? Other models can. So please rephrase or remove this part of the sentence.*

To avoid any misunderstandings about the abilities and weaknesses of PMCAMx, we have deleted this sentence. The revised manuscript now mentions that "The main

objective of this study is to quantify how fine particulate matter (PM2.5) responds to emissions changes of its precursors and how a change on temperature would influence its concentrations. For this purpose we apply a three-dimensional CTM (PMCAMx-2008) over Europe. Three-dimensional CTMs are well suited for this purpose because they link emissions and meteorology to PM2.5 concentrations through descriptions of the physics and chemistry of the atmosphere. The PMCAMx-2008 model includes state-of-the-art organic and inorganic aerosol modules which make it well suited for the purpose of this study. . .".

12. *Section 2.1: please also mention modules for sea salt and dust. Please include removal mechanisms. Does the dry deposition scheme include temperature dependent stomatal conductance description?*

Sea salt emissions were calculated using the marine aerosol emission model developed by O'Dowd et al. (2008). This is now mentioned in the revised manuscript in the section on modeling domain and inputs. The Saharan dust emissions are not modeled explicitly but are included as boundary conditions in the domain. The dust emissions from Europe are included in the corresponding inventories. This information has been added to Section 2.2.

PMCAMx simulates wet deposition using a scavenging coefficient approach in which the local rate of concentration change within or below a precipitating cloud is equal to the product of the concentration of a pollutant and its scavenging coefficient. The scavenging coefficient is determined differently for gases and particles, based upon relationships described by Seinfeld and Pandis (2006). For dry deposition, PMCAMx determines a deposition velocity for each land use type, for each given species, particle size, and grid cell, and then linearly combines them according to the fractional distribution of land use. The deposition flux is used as the lower boundary condition in the vertical diffusion algorithm. For the gas phase species, the resistance model of Wesely (1989) is used. The deposition velocity is calculated from three resistances

in series: the aerodynamic resistance, the quasi-laminar resistance, and the surface resistance. The surface resistance is further expressed as several more serial and parallel resistances that depend upon the physical and chemical properties of the surface in question. Many of these resistances, such as the stomatal resistance are temperature dependent on the model. Over water, the surface resistance is based on some improvements adopted by Kumar et al. (1996) in UAM-AERO, following Sehmel (1980). For aerosol particles the resistance approach of Slinn and Slinn (1980), as implemented in UAM-AERO (Kumar et al., 1996), has been adopted in PMCAMx. This information has been added in the revised manuscript.

13. *Section 2.2: Are the WRF data for Europe evaluated? Please add reference or summarise results.*

The performance of WRF for Europe has been the topic of a number of recent studies (Jimenez-Guerrero et al., 2008; De Meij et al., 2009; Im et al., 2010; Argueso et al., 2011; Garcia-Diez et al., 2012) against observed meteorological variables has been subject to other recent studies. These studies are now mentioned in Section 2.2

14. *The GEMS emission dataset is quite old and was replaced by the MACC dataset. Why was this set not used? How do you treat emissions as function of time, height, VOC split, etc. Please provide a reference for the EC-OC emission database.*

The inventory used in this study was developed by the TNO team during the EUCAARI project (Denier van der Gon et al., 2010) as a continuation of the work in GEMS and MACC. The major improvement was the development of the Pan-European Carbonaceous Aerosol Inventory improving the emissions estimates of anthropogenic particulate organic and elemental carbon (Kulmala et al., 2011). The interested reader is now referred to Denier van der Gon et al. (2010) for the details of the inventory. This point is clarified in the revised manuscript.

15. *You use the VBS approach. Do you assume additional (condensable) emissions that are not inventoried in the emission database? If so, please specify.*

As we mention in Section 2.1 on model description we followed the approach of Tsimpidi et al. (2010) and Shrivastava et al. (2008) in which IVOC emissions are added to the emissions in the inventory. These emissions are assumed to be proportional to those of the primary OA and are not included in the existing inventories. We assume that the IVOC emission rate is 1.5 times that of the original POA emissions. This approach has been followed by a number of previous studies (Robinson et al., 2007; Murphy and Pandis, 2009; Hodzic et al., 2010; Tsimpidi et al., 2010). The IVOCs can be oxidized by OH and may go to lower volatility and condense. We have clarified this point in the revised text.

16. *Please specify the boundary conditions.*

The boundary conditions used in this study were taken from Fountoukis et al. (2011) (shown in Table 1 of that paper) and are based on measured background concentrations in sites close to the boundaries of the domain (e.g., Zhang et al., 2007; Seinfeld and Pandis 2006). This information has been added to the revised manuscript.

17. *Section 2.3: This evaluation is too limited to be able to understand the model performance and short comings that are important to interpret the results (see above).*

We do agree with the reviewer that additional information about its performance is needed and we have added a summary of the evaluation results in the main paper and some additional summary evaluation statistics in the Supplementary Information. For additional information please see also our response to comment 2.

18. *Section 3: please explain the reason for the maxima above sea. The distributions for winter and summer are a-typical for some species (e.g. nitrate – no broad*

distributions across continental EU in winter, no Po-Valley maximum), OA – Germany minimum, PM2.5 lack of high concentrations over south eastern Europe) which may be due to the special events occurring in these periods.

The maxima of total PM2.5 above sea are due to several reasons. During the May period the high PM concentrations in Northern Europe (an area extending from Southern Scandinavia to Western Ireland) are due mainly to high ammonium nitrate concentrations. These predictions are consistent with the extended airborne measurements in the area as well as the measurements in Cabauw and Mace Head (Fountoukis et al., 2011). The high levels over the Mediterranean (central and eastern) are mainly due to high sulfate levels. The predicted sulfate is consistent with the measurements in Finokalia. Dust (in the Mediterranean) and sea-salt also contribute to these elevated levels over water. The elevated levels in the winter are due to a combination of high ammonium nitrate, organic aerosol and sea-salt. These are now discussed in the revised paper and the reader is referred to Fountoukis et al. (2011) for additional details.

The model predicts high nitrate concentrations over most of Europe during the winter and the levels in the Po Valley are relatively high (Figure 2f). These are average periods over a month including periods of accelerated removal (e.g., rainfall); the concentrations during specific episodes are a lot higher. A brief discussion about the ammonium nitrate fields has been added to the paper.

The predicted OA over Germany is indeed low compared to the available observations in Melpitz. There is strong evidence that the wood-burning emissions in this area are underestimated. We have found similar underestimation of the emissions in Sweden and Switzerland and this is the topic of a forthcoming publication. This important issue is discussed in the revised paper.

19. *Some words on the representativity of the conditions in EU during the periods is needed I think. March 2009 was summerly hot for instance in western Europe.*

Both periods had quite variable meteorology with a number of different meteorological regimes and air pollution episodes. We have added some discussion and references to publications discussing the meteorology during the simulated periods. This is also discussed in our response to comment 7.

20. *Section 4.2: Ozone increase in western Europe in case of NOx reduction is presumably due to less titration in this VOC limited area. The increase of ozone in urban centers is well known and has been shown many times, and is certainly not new.*

We agree that this is not a new result. The main objective of this study was to investigate the sensitivity of fine PM levels to anthropogenic emissions. However, these changes affect also the ozone levels. We think that the difference in response of ozone and the fine PM to NOx in urban areas should be stressed to avoid misleading the readers about the overall effectiveness of this strategy in improving air quality.

21. *Chapter 5. First paragraph: so temperature is increased everywhere. Does this also include the dry deposition and additional stomatal closure at high temperatures?*

The stomatal resistance in the dry deposition scheme used is temperature dependent (see also our response to comment 12). This is now mentioned in the revised text.

22. *Page 19, line: 25: Paris is mentioned here for POA impact. Considering the MEGAPOLI campaign results, is this realistic result? How does it translate to other urban signals in the pictures?*

The results presented here are relatively consistent with the conclusions of the MEGAPOLI campaigns. High regional concentrations of OA were observed in the area and they were due mainly to residential wood burning. Most of the emissions were in the surroundings of Paris and their effect was extending over a large area. The high OA levels in a lot of European major urban centers in the winter (Figure 2b) are in general

consistent with the available measurements and they are caused by a combination of weak vertical mixing and higher emissions from home heating. We have added a brief discussion in this point in the manuscript.

23. *Conclusion section: Final paragraph s very weak. It is mentioned that Control strategies may play a crucial role!!! This key. Please rephrase.*

We have followed the reviewer's suggestion. In the revised manuscript the last paragraph in the conclusions section has been rephrased.

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