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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Aerosol simulation applying high resolution anthropogenic emissions with the EMAC chemistry-climate model" *by* A. Pozzer et al.

A. Pozzer et al.

apozzer@ictp.it

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We thank the referee #2 for the comments. We sincerely appreciate the comments, being motivated and precise, which will help to improve the scientific quality of the manuscript.

Two major issues are raised by the referee. Firstly the title is misleading because we do not really take advantage of the high resolution emissions (issue raised also by referee #1). Secondly, the analysis of the influence of the seasonal cycle of the emissions on gas and aerosol concentrations could have been influenced by the non CTM (Chemical Transport Model) behaviour of the EMAC model (which is based on

the General Circulation Model ECHAM5).

We indeed agree with both referees that the title does not present well the topic described the manuscript. As mentioned by referee #2, instead, the budget analysis provides new results with regard to atmospheric process which should be highlighted, because the topic is "...not considered in the title and not even discussed in the conclusions". This is a lack of coherence in the manuscript, which we intend to correct. Hence, with the approval of the editor, we would change the title of the manuscript in "Distributions and regional budgets of aerosols and their precursors simulated with the EMAC chemistry-climate model". We believe that the new title and the corrections/modifications suggested by the referee (see reply below to specific comments) will remove the incoherence which are actually present in the text and will improve the clarity of the manuscript.

Regarding the non CTM behaviour of the EMAC model, we completely agree with the referee about the issue. We apologise for not having mentioned in the manuscript the topic, which is extremely important and needed of clarification. In the simulations performed (ST and NS), the coupling between the radiation and the atmospheric composition has been removed (switched off). This implies that changes in the atmospheric composition calculated by the chemical mechanism (i.e. ozone, aerosols and greenhouse gases) do not affect the dynamical behaviour of the model, which is instead running based on a climatological concentrations of such components. Thanks to this approach, the EMAC model is behaving as a pseudo-CTM, where the nudging is forcing the model with the same intensity both in ST and in the NT simulations. More specifically in our case, the dynamics in both simulations are binary identical; the differences in the aerosols (and precursors) concentrations (mixing ratios) between simulation ST and NS calculated by the model are purely due to the different emissions and not by different meteorology. Hence we can reply to the referee confirming that we took care of the issue, and that the model "noise" (intended as different transport between the two simulations) is not present. We believe that the results obtained in Sect.5 are

reasonable results from well performed simulations, and we prefer to keep the topic in the manuscript, as also suggested by referee #1. We will add all these informations in the revised manuscript.

Specific comments: we have changed the text accordingly to the specific comments of the referee. Here below we answered the questions raised.

Page 25210, Page 8 The MECCA mechanism has been described in detailed in Jöckel et al. (2006), and the complete equation set is present in its electronic supplement. We will extend the description of the chemical mechanism adding also this reference in the manuscript.

Page 25212, line 6 For each mode, the total aerosol volume (all com- pounds including water) is calculated based on aerosol masses of all compounds and their respective densities. Then the volume fraction of each of the individual compounds is determined in a similar way (this guarantees that the sum of all fractions equals unity). The refractive index for each compound is weighted with its volume fraction to give the total refractive index for the aerosol of this mode.

Page 25212, line 14 The wavelength bands of the internal calculation are mapped to the wavelength bands of the radiation scheme, using a (constant) reference spectrum for the solar incoming flux at the Earth surface in which trace gas absorption based on climatological values has been considered. Additionally for arbitrary wavelengths (e.g. the 550nm wavelength of MODIS) diagnostics can be added to give the weighted values of AOD and other parameters at the specified wavelength following the same principles.

Page 25212, line 18 The actual mechanism for calculating the AOD is the following: for each mode the extinction coeïňČcient is calculated for a single particle; the extinction coeïňČcient is then multiplied by the number of particles per grid cell, giving the vertical integral of the extinction per mode and per layer. The AOD per

layer is obtained by adding all the modes in the layer, and the sum of all AOD values over the vertical model domain yields the total atmospheric AOD.

Page 25212, line 27 We corrected the manuscript. CIRCE stands for “Climate Change and Impact Research: the Mediterranean Environment”.

Page 25213, line 7 We corrected the sentence.

Section 2.3 The data were downloaded from (with references):

CASTNET : http://www.epa.gov/castnet/data.html

EMEP : http://www.nilu.no/projects/ccc/emepdata.html, Hjellbrekke and Fjæraa (2011)

EANET : http://www.eanet.cc/product/index.html, ACAP,Asia Center for Air Pollution Research (ACAP) (2011)

Please note that we did not used the entire the observational dataset, but we extracted the tracers/aerosol compounds of interest which we used in the manuscript (NO_3^-, SO_4^{2-}, NH_4^+ and Na^+). The observational datasets include a much larger amount of aerosol compounds/tracers which are not used in this work and we refer to the web sites for additional informations.

Section 3.1 We agree with the referee about this point. Nevertheless we believe that we were cautious in drawing the conclusions, as the general overestimations/underestimations were corroborated with additional references and motivations (e.g. global emissions of Particulate Organic Matter and Black Carbon from biomass burning). Following also the comments from referee #1 we reformulated part of the text.

Section 3.1, Figure 1 Referee #1 also raised a similar comments. We added a figure with the absolute value of the model.

Page 25219, line 10-11 The information requested by the referee is missing and very important in this calculation. For standard deviation we used the root mean square difference (in space) from the spatial average, each estimated from the set of multi year monthly value. Hence in our case the standard deviation represents the spatial variability of the data from each set of temporal means (month).

Page 25220, line 10-11 As mentioned at the beginning of Sect.3, we used only monthly values in our calculations. Additionally all level 3 data from MODIS and MISR are in monthly means. Hence also the correlation was based on monthly averages. We clarify this in the MODIS/MISR description and in this part of the text.

Page 25221, line 13 and Page 25221, line 16 As mentioned by the referee, we gave at the beginning of Sect.3.2 a short description of the figures 4-11. We apologise for the unclear description and missing reference to the figure. This is corrected following also the suggestions of the referee.

Page 25222, line 4 Some precisions have to be made: the number of stations used in the comparison is very similar in each network (30 stations), but the EANET network covers a much larger region than CASTNET and EMEP networks. As correctly mentioned by the referee, in Asia the number of stations (compared to the area covered) could produce a rather weak statistics, and not be very significant for the region. We add to Tab.3 the number of stations used in the comparison, so to have an indication of the quality of the statistics.

Page 25222, line 9-11 We agree with the referee with his/her analysis. The Taylor plots were misinterpreted and we corrected this error in the revised version.

Page 25223, line 3 We agree with the referee and we reformulated the sentence.

Page 25223, line 18-24 The referee is indeed correct and we apologise for the wrong wording used in the text formulation. We made the appropriate changes in the

text by mentioning that here we refer to the spatial distribution. Nevertheless a low spatial correlation is indeed present during summer month, which could indicate a possible evaporation of nitrate from the filter, although it is not a confirmation. As the Taylor plots were not correctly interpreted, we will revise and correct the text.

Page 25224, line 2-3 We will reformulate the sentence : "As shown in Fig. 8, high NH_4 concentration are found over continental regions, especially over India and China and over Central Europe, which agrees with the findings of Clarisse et al. (2009)".

Section 3.2.3 As mentioned by the referee, also NH_4^+ can evaporate from filters and this could bias the observations. Because this effect is present during the hot and dry season (Summer), the real concentrations for NH_4^+ could be even higher than observed in July, August and September. In this case, the differences between model results and real concentrations could be even higher, and the discrepancies described in the model represents a lower limit. This, to our opinion, corroborates the findings of this section.

Page 25224, line 14 We agree with the referee that the spatial distribution is not extremely well reproduced. Nevertheless, the temporal correlation (Fig.8 lower panel) is above 0.5 for almost all stations, i.e. with a much better agreement with respect to the CASTNET network..

Page 25225, line 20-22 We respectfully disagree with the referee. Fig.10 (upper panel) clearly shows that the boxes (multi year means of the observations) shows a darker/greener colours (i.e. lower values) than the underlined colour value (multi year means of the model results). Hence Fig.10 is in agreement with Fig.11. We miss the discussion of Fig.11, which is added in the revised version.

Section 4 As suggested by the referee, we improved the entire section, adding more emphasis on the budgets and their analysis in the manuscript.

1. We added a map of the specific regions used for calculating the budgets.
2. A figure which summarise Tab. 5, as suggested by the referee, will be included. A first attempt to formulate the figure is shown here in Fig.1, where the information contained in Tab.5 is visually presented.
3. The discussion requested by the referee is essential. This section is partially reformulated, mostly by adding new findings that could be inferred from Tab.5 and the new figure. Being this the main focus of the manuscript (as suggested by the referee) we put additional effort in improving the section.
4. We included the findings in the conclusions at the end of the manuscript.

Figure 2 The triangle shows that the model-remote sensed data comparison present a rather similar statistic (during the winter period) as inter-satellite comparison. This is connected to the comment of the referee that "the satellite data products, however, can have large uncertainties", and can give an indication of the real performance of the simulation. We mention this point in the revised manuscript, although we are reluctant in going into detail in the subject. In fact, inter-satellite comparison is a rather large topic which should be covered in detail, which is not the scope of this work.

Fig.5 We added the network names in the figures as suggested.

References

ACAP,Asia Center for Air Pollution Research (ACAP). Eanet data on the acid deposition in the east asian region. Technical report, Network Center for EANET, 2011.

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J. Lelieveld. Evaluation of the atmospheric chemistry gcm ECHAM5/MESSy: Consistent simulation of ozone in the stratosphere and troposphere. *Atmos. Chem. Phys.*, 6:5067–5104, 2006.

Figure 1 label: Budget of different aerosol (and precursors) species. The colour code denotes the process, while the positive (negative) value is associated with a source (sink) of the component. In the abscissa the regions are listed: North America (NA), Europe (EU), East Asia (EA), Central Africa (CA) and South America (SA). For non bulk species the gas-phase and aerosol contribution to the budget are also shown with patterned and solid colour, respectively. Additionally, in the non bulk aerosol species, the emissions where removed for clarity.

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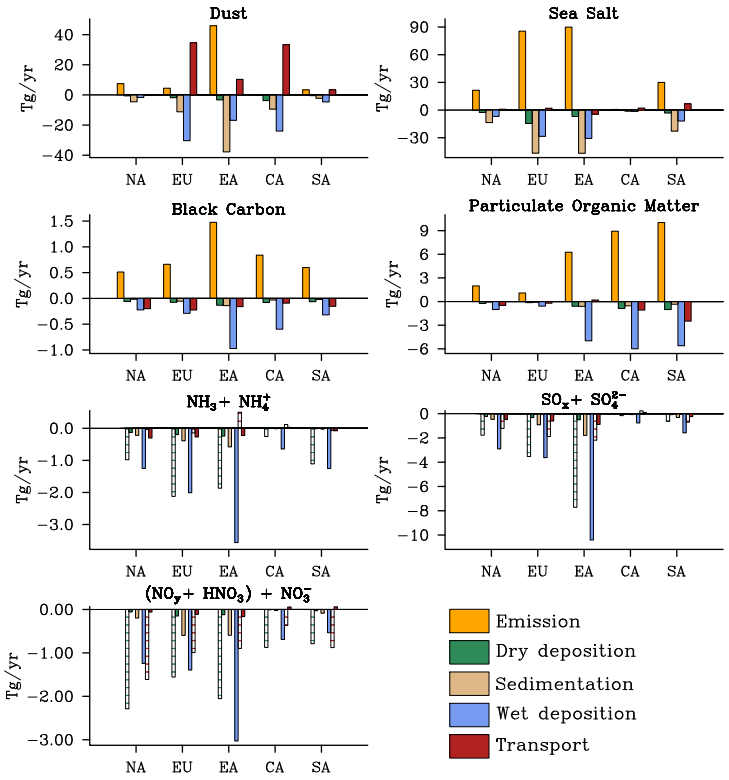

Fig. 1. See end of response to referee#2 for the label (Page C8)