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Atmospheric Chemistry and Physics Discussions

Interactive comment on "GEM-AQ/EC, an on-line global multiscale chemical weather modelling system: model development and evaluations of global aerosol climatology" *by* S. L. Gong et al.

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We thank the reviewer for the instructive comments which have improved the quality of the paper. The manuscript (acp-2011-979) has been revised following the comments. To help the readers of this reply, we have quoted the questions of the reviewer in brackets.

[1 The description of emissions, in particular Section 2.3, should be improved. It is not clear which anthropogenic emissions are kept constant during the simulation and which are varying during the 10 yr simulation. In particular more informations should be provided about sulphur emissions, which are an important component of the aerosols.

I would suggest to the authors to summarize in a new table or expanding table 2 with sulphur emission budget. About sulphur species, in Section 2.2 there is no mention about DMS emissions, which are a significant source of sulphate. Some comments should be provided on the choice made by the authors to use quite old and coarse resolution datasets (GEIA and Liousse 1997, 4.4 x 7.5 degrees).]

A: We have revised the Section 2.3 to clarify the emission descriptions. The GEIA sulfur emission inventory used in 10 year-simulation include the seasonal averages of SO2, SO4 and DMS for the year 1985. The Table 2 summarized the annual aerosol emissions by primary source types in the northern and southern hemispheres, and globally over the 10 years of study. Since sulphate is mainly a secondary aerosol from SO2 and DMS, depending on the oxidation reactions, we did not add the sulphur emission budget of SO2, SO4, and DMS in the Table 2. Due to the limitation of the available sources of sulphur aerosol emission inventories at the time of the beginning of the study (mid-2000's), the sulphur emission inventories were based on 1985 GEIA data and were integrated in the model without any inter-annual variation during the whole 10 years of simulation. Regarding biomass burning sources, the original 4.4o$\times$7.5o inventory from Liousse et al. (1996) was interpolated onto a 1o$\times$1o grid and a land-sea mask was applied to constrain emission fluxes to the land areas along the coasts. We agree that the GEIA emissions used in this paper are quite old. However, the purpose of this paper was to implement plausible emission inventories in the first version of the aerosol modeling system GEM-AQ/EC. The simulation for each year took 1-2 months to complete. That represents 1 to 2 years for the simulations only. When the work started mid-2000s, we had to use best guess emissions available at that time. And all "background" static emissions had to stay constant for the 10 years of study. That's the reason why we used older emissions. Since the model has been developed, the next version of GEM-AQ (called GEM-MACH) will include state-of-the-art emission inventories with daily temporal resolutions and the combination of various inventories for different world regions.

[2) Section 3.3: the description of the comparison between AERONET data and model results should include more details. For example, how the stations were selected? There was any filter applied to the stations in other to calculate the monthly average? It is not very clear the sentence on the significance of the correlation coefficient between the observed and simulated AODs. How is it calculated? I don't think that we can observe from Figure 7 the spatial and temporal distributions of the AODs. The under-overestimations should be analyzed region by region in more detail and better related to the possible causes (e.g. anthropogenic or natural emissions).]

A: To evaluate the model performance, monthly modeled AODs were compared to surface based AOD observations from AERONET during 1995-2004. AERONET data cover the whole 10 years of simulation period. Simulated AODs were distributed in the horizontal model grid at the global uniform resolution of 1o$\times$1o. We were able to select the model grid cells corresponding to AERONET sites. We then compared the simulated and observed AODs in the same grid cells. No filter was applied to the AERONET station data to calculate monthly averages. The significance of correlation could be calculated, based on the t-test, to assess whether the means of both groups were statistically different from each other. Both groups of simulated and observed AODs were built from the same spatial and temporal distributions of AERONET sites during the 10 years of simulation. The correlation between the two groups is shown in Fig. 7 to evaluate the AOD modeling against the AERONET observations.

Due to the sparsely distributed AERONET sites for AOD observations, we have not analyzed the differences between simulated and observed AOD region by region and we have not determined the potential impact of different source types (e.g. anthropogenic or natural emissions). The under- and over-estimations could be caused by the uncertainties characterizing aerosol emissions and atmospheric transport patterns. In the revised version, we clarified the methodology used to perform the comparison and added some sentences in response to the reviewer's comments in section 3.

[3) The description and the analysis of the comparisons between observed and simu-

lated PM10 and PM2.5 is not always clear (Section 3.4). The authors should provide more details on the methodology used to perform the comparison. For example, how many years and which periods were available for each measurement network (Improve, EMEP, ..)? How many stations were considered and how they were selected? Any filter was applied to select the stations? How were calculated the statistics (correlation coefficients), for example what doest it mean "0.65 for the 10-yr averaged PM10 and PM2.5"? Also the authors should comment about the impact of aerosol species which are not included in the model, such as nitrate and secondary organic aerosols, when comparing to PM observations. In principle the model should always underestimatePM10 and PM2.5 observations. The authors should also link what seen in the PM2.5 and PM10 observations with the discussion on AOD. For example it is seen a model overestimation of dust over NA, which can be also seen in an overestimation of AOD over the Atlantic (Figure 6). The sum of the aerosol components results in a simulated PM10 and PM2.5 generally within a factor of 2 with the observations, on the other hand in Figure 9 we can see that sulphate and dust are overestimated by the model, while BC and POM underestimated. The authors should comment more on this and also how the simulated aerosol chemical composition is influencing the simulated AOD.]

A: Because the aerosol module accounts for five aerosol species: sea salt, sulphate, soil dust, black carbon and organic carbon in 12 aerosol size (diameter) bins from 0.01 to 40.96 μm for modeling results, we can calculate the simulated PM10 and PM2.5. From the 10-yr modeling result. The simulated PM10 and PM2.5 values are distributed in the horizontal model grids with the global uniform resolution of 1$\times$1. From the locations of IMPROVE and EMEP sites, we could select the model girds covering the observation sites. Therefore, we compare the simulated and observed PM10 and PM2.5 in the same grids. No filter is applied on the AERONET station data in other to calculate the monthly averages. We have used all the data from the IMPROVE and EMEP stations that fall within the time period of model simulations, i.e. 1995 to 2004.

The significance of correlation could be calculated based on the t-test, which assesses whether the means of two groups are statistically different from each other. The two data groups of simulated and observed PM-values are built from the same spatial and temporal distributions of IMPROVE and EMEP sites over the time series with the available observation data. The correlation coefficients between two data groups are calculated based on the classical statistics to evaluate the PM modeling against the observations. For example, the correlation coefficient of 0.6528 in Fig. 8b for the 10-yr averaged PM2.5 is high passing a significance level of 0.01. The impact of nitrate aerosol species which are not included in the model has been discussed in the comparisons to PM observations. The overestimates in PM may also result from the discrepancies in both the aerosol emissions used in the model and the simulated meteorology governing the aerosol transport and other processes. It might be a good idea to link the AOD with PM. However, we cannot quantitatively link them as the AOD is a column integrated properties while the PM is for surface only. The comparison between modeled and observed AOD is a rather complicated process as both the chemical compositions and size distributions have impacts on the model values. The reviewer has raised a very good question which we may not able to complete in this study as we may need both chemical composition and size distribution measurement to validate the comparisons.

[4) The conclusions should be expanded. The authors should summarize the results in more detail and discuss the limitations/advantages of using a global model for air quality studies. Probably, they should also introduce in the conclusions the companion paper of Zhao et al., 2012, which contains further analysis of the same simulation.]

A: Following the suggestion, we expanded the conclusions of the revised manuscript with a more detailed result summary, with a discussion on global models of air quality and with an introduction to the companion paper (Zhao et al., 2012).

Minor comments:

[Page 9284, line 6: "at scales from global to urban domains". The authors, also in the

title, define the model as a multiscale model, but they don't give any description of the model features for this specific point. Please clarify what do you mean for "Multiscale" model and provide the correspondent model description, for example in Section 2.]

A: In the first paragraph of section 2, we added the following model description: The Global Environmental Multiscale model GEM, as the host meteorological model for the on-line implementation of air quality processes could be run in global uniform, global variable and limited area configurations for modeling chemical weather at scales from global to urban domains.

[Page 9285, lines 10-12: "South east Asia . . . pollution loads". It is not clear if the sentence is still referring to the paper of Koch et al, 2007 or is form another reference.]

A: This sentence is still referring the paper of Koch et al, 2007. We have moved the citation of "(Koch et al, 2007)" to the end of this sentence.

[Page 9285, line 18: "primary sources", but sulphate is mainly a secondary aerosol.]
A: Yes, sulphate is mainly a secondary aerosol. The aerosol emission data include the primary sulphate sources.

[Page 9286, lines 7-11: Consider to revise the entire sentence "For instance, . . . transport models", as it results difficult to read.]

A: It has been revised.

[Page 9286, line 17: the authors stated that the goal of the study is to develop a comprehensive emission inventory of both natural and anthropogenic aerosol for a 10 year simulation. On the other hand anthropogenic emissions as described in Section 2.3 are mainly based on existing inventories previously developed. Page 9287, Section 2.1 could be more detailed. Which are the couplings between the gas and aerosol phase, in particular regarding SO2 and DMS oxidation?] A: In the revised manuscript, we have made the changes to the description of the goal as "to develop a comprehensive emission inventory of natural aerosols for ten consecutive years from 1995 through 2004"

as this is more accurate to reflect the objective of the paper. We have also revised the Section 2.3 and made more clear what was developed and what was just used from other studies. In terms of the coupling between gases and aerosols, we have the oxidation of DMS by OH and O3 to SO2, which, together with the SO2 emissions from anthropogenic sources, is oxidized by OH into gaseous H2SO4. Then the gas-to-particle conversion schemes are activated to turn H2SO4(gas) into H2SO4(particle), i.e. sulphate. The detailed description of the scheme used in this paper can be found in Gong et al. (2003).

[Page 9289, Line 7: "SF" is "SU" for sulphate?]

A: “SF” is changed to “SU” for sulphate.

[Page 9290, Lines 9-11: "This nudge setup . . . reanalysis meteorology". Consider a revision of the sentence, as difficult to follow. Also about nudging, which variables are nudged?]

A: The meteorological variables of wind, temperature, pressure and water vapor are nudged. Following the reviewer’s suggestion, we have modified the sentence.

[Page 9290, Line12: a spin-up of 3 months was performed, but which initial concentrations were used to start the spin-up? 3 months are probably enough for aerosols but what about the gas-phase chemistry?]

A: We have clarified the description: The initial concentrations of aerosols are zero, and but the initial concentrations of gas chemistry are from the global distribution of October 1 averaged from the 5-year GEM-AQ simulation (Kaminski et al., 2008, ACP). Therefore, a spin-up of 3 months starting on Oct.1, 1994 is enough for aerosols and also for gas-phase chemistry.

[Page 9290, Line 17-19: here the authors state that the anthropogenic emissions are constant, which is not completely clear from Section 2.3. Some emissions are varying (e.g fossil fuel) some are fixed (e.g. bio fuel).]

A: There are seasonal variations in some emissions of anthropogenic aerosols with the monthly data (e.g fossil fuel), and but no inter-annual variability over 10-years for all anthropogenic aerosol emissions. We have added the more explanations.

[Page 9292, Line 5-6: "Global SS within the Canadian GCMIII integrating CAM (Gong et al 2002)." Sentence not completed?]

A: It has been completed.

[Page 9292, Line 10: the meaning of "r80" is not explained in the text.]

A: This is the particle radius at relative humidity at 80%. We have added this into the text.

[Page 9292, Lines 1-12: sometime units of sea salt emissions are given as kg and not as kg yr-1.]

A: Line 7-8: the annual emissions of 5.8–7.5$\times$1012 kg for global sea salt aerosols.

[Page 9292, Lines 14-20: the recent paper from Huneeus,ÂËŸaN. et al 2011 (Global dust model intercomparison in AeroCom phase I, Atmos. Chem. Phys.) should be included in the discussion of dust simulation.]

A: Thanks for the reference information. The ACP-paper has been included in the discussion of dust simulation.

[Page 9293, Line 24: "Korea" instead of "Korean".]

A: It has been changed to “Korea".

[Page 9294, line 19: the acronym BOTE was not explained before in the text, and it is mentioned only one time in the entire paper.]

A: It is removed from the manuscript.

[Page 9294, line 20: Southern Europe is "west" of the Ural Mountains.]

A: it is corrected with “west”.

[Page 9295, line 9: "except in 1996".]

A: “expect” is changed to “except”.

[Page 9295, Line 22: ".. primary aerosol source strengths". Sulphate aerosol is mainly a secondary aerosol which is one of the main component in determining the total AOD.]

A: the “primary” is deleted there.

[Page 9295, Line 27: "model simulated AOD summarizing the contributions from . . .". This sentence is not very clear, as Figure 6 shows the simulated AOD calculated as the sum of the single aerosol species. I don’t think that this can be described as the summary of the contribution from all the species.]

A: Following this comment, this sentence is modified as “model simulated AOD as the sum of the single aerosol species of sulphate, sea-salt, black carbon, organic carbon and soil dust. . ...”.

[Page 9295, Figure 6: I would suggest to the authors to improve Figure 6, by using a different color scale, or maybe providing both the MODIS and simulated seasonal maps, instead of over plotting contour lines. For example it seems that the AOD is overestimated by the model for all season over the Atlantic (dust outflow).]

A: To and directly compare the MODIS- and simulated AOD, we plotted Fig. 6 containing the filled contours for MODIS-AOD and the contour lines for the simulated AOD together, which also reduces the figures. In response to the reviewer’s suggestion, we have double enlarged Fig.6 so that the contour lines and the underlying shading can be more clearly seen.

[Page 9300, lines 4-7: it is not clear here and in the emission description if a seasonality is included in the used emission dataset.]

A: The seasonality is included in some emission dataset.

[Page 9300, line 15: from dust observations in the Atlantic (Figure 13. Barbados, Rsmas, and Bermuda) it is seen that the model underestimates the observations during the summer peak, while previously the authors showed an overestimation of mineral dust transport from Africa (Figures 6 and 9).]

A: There are two different observation data sets. Data of Barbados, Rsmas, and Bermuda are not from the IMPROVE.

[Page 93001, line 5-6: A reference is not given for the review mentioned in the text.] A: Added

[Page 93001, Lines19-20: Figure 14 is not properly described. It is not given both here and in the figure caption the source of the satellite data.]

A: Yes. We have added a description for the Figure 14.

[Page 93001, lines 26-28: what is exactly represented by the contour lines in Fig. 14b. Is it the inter annual variability or the seasonal variability? My understanding of theFigure is that the seasonal variability of sea-salt is large, as shown by Figure 3, but the inter annual variability is rather small, about 1-2% (Figure 14b) in the souther hemisphere below 40S (where the main SS emissions occur). The larger Std Deviation over the Oceans at higher latitudes, might be due to the variability of anthropogenic aerosol continental outflow.]

A: The Fig. 14b caption was modified: “Global distribution of the averaged percentages of natural aerosol contributions to total aerosols (shading) with the standard deviations (contour lines) of the percentages from the 10 yr modeling”. It is the inter- annual variability (not the seasonal variability). The larger Std Deviation over the Oceans at higher latitude might be due to the inter-annual variability in atmospheric circulation controlling the anthropogenic aerosol continental outflow.

[Figure 1: Caption ". . . red dots are the stations ARE from GAW .."] A: “ARE” was removed in the revised version.

[Figure 8: The figures in panel b are the same of panel a (only IMPROVE is shown).]

A: Agree. The caption was rewritten.

[Figure 13: Caption " . . . soil DUCT . . ."]

A: Corrected.

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