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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.

Major comments

[1. The major emission sources on the Earth are exhaust, tire and brake emissions from automobiles, and power plant spewing. Information is not provided for such sectors. It was mentioned that the missing nitrate contributes to the model underprediction,

but the most significant sources may be fugitive dust from paved and unpaved road, agricultural operation, mining and construction (e.g., 88% of US PM10 emissions, see http://www.epa.gov/ttnchie1/trends/ for more details). It is not clear if the GEIA inventory has included these sources, and which version of the emission inventories is used. This sector may be responsible for the soil over eastern US where windblown dust is not very important. Similarly, it is unclear which year of sulfur emissions the GEIA inventory represents. Note that rapid SO2 reduction has occurred over the last decades due to tighter regulations and increased efficiency of control technology (see the above link).]

A: We agreed with the comment. The fugitive dust emissions were not considered in this study as there was no global emission inventory for the fugitive dust at the time when this study started. In a regional simulation, we have found the importance of fugitive dust contribution to the ambient PM levels in North America (Park et al. 2010). Therefore, this should be included in the future global aerosol models.

For the GEM-AQ/EC simulations, we used the version 1A of the GEIA inventory based on year 1985. This inventory does not include the fugitive dust emissions. 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.

[2. The under-prediction of organic aerosols seems linked to both the uncertainties in fire emission estimation and the missing representation of secondary organic aerosols. here is a gas chemistry module in the model. Does it interact with the aerosol module? Do you have emissions of active VOCs, such as isoprene and monoterpene? The underestimation of biomass burning will be more important in wintertime, while SOA formation plays a larger role in summer.]

A: Yes, the under-prediction of organic aerosols is likely due to the uncertainties in both fire emission estimation and production of secondary organic aerosols. The underestimation of biomass burning will be more important in wintertime, while SOA formation plays a larger role in the summertime.

In GEM-AQ/EC, the chemical mechanism module ADOM (the Acid Deposition and Oxidants Model, Venkatram et al., 1988) is used to model the gas chemistry, which interacts with the aerosol module. The aerosol module includes chemical transformation of sulphate and the production of secondary organic aerosols by chemical transformation from their precursors together with particle nucleation, condensation and coagulation, which are also implemented.

These precursors are linked on-line with a gas phase chemistry module within the GEM-AQ/EC. We have used the emissions of active VOCs, such as isoprene and monoterpene in modeling aerosols. In the revised version, we added the discussion on the underestimation of aerosol modeling and further clarified the model description.

[3. The method of fire emission estimation seems unsatisfactory, when the model performance of OC prediction is concerned. A more in-depth investigation of the underlying reasons may help future development of the model and supporting dataset. Could the bias caused by misplaced emission injection height or something else?]

A: Agree. Comments have been incorporated in Section 3 of the revised manuscript. The method to calculate emissions from boreal fires, presented in the current paper, does not take into account the variability in amounts of fuel consumed from month to month, except for Canada. Therefore, the method underpredicts emissions, including those of OC, from fires during drier than normal years. A new version of the boreal fire emissions is currently under development. It will include the influence of daily variation of weather conditions on fuel consumed amounts across the whole boreal vegetation. Regarding injection heights, aerosols from fire emissions are injected at different altitudes, depending on type of vegetation, weather conditions, and thus the nature of the

fire. The technique used in the present manuscript follows the vertical distribution of smoke in the model layers, already used in the AeroCom study (Dentener et al., ACP, 2006). The injection heights are set to constant values for various geographical areas; for example, 4-5 km in boreal North America and 2-3 km for other regions. The authors agree that this approach is coarse. Therefore, the bias of OC prediction could be caused by "misplaced" injection heights for specific years or months. However, considering the seasonality of injection heights would mean taking into account the weather conditions in the calculation of the plume heights over 10 years, which was beyond the scope of our modelling study. The next version of the boreal fire emission datasets is currently under development and will integrate the seasonal variability of the emissions heights following the method detailed in Lavoué et al. (Int. J. Wildland Fire, 2007).

[4. The estimates of dust and fire emissions from the USA seem to be much lower than previous studies. For instance, the annual dust emission is around 4 Tg/yr or 0.2% of global budget, compared to the 3% from Ginoux et al. (JGR, 2001). It was claimed that Russia and Canada are the main biomass burning source in the North Hemisphere, while other studies have found that US and Central America sources are more important (e.g. Wiedinmyer et al, AE, 2006). Because of the large uncertainties in emissions and the missing sources, caution has been taken when the authors proceed to quantify the relative contribution of natural and anthropogenic emissions.]

A: Thank for providing two reference papers. We also agree with the comments. There are large uncertainties in emissions and the missing sources for aerosols. In the revised manuscript, we cite the both papers of Ginoux et al. (JGR, 2001) and Wiedinmyer et al. (AE, 2006) in the discussion in quantifying the relative contribution of natural and anthropogenic emissions dust aerosol emissions in section 3.1. References: Ginoux, P.,Chin, M., Tegen, I., Prospero, J.M. Holben, B., Dubovik, O.and Lin, S.-J. : Sources and distributions of dust aerosols simulated with the GOCART model, J. Geophys. Res., 106, 20,255–20,274, 2001. Wiedinmyer, C. , Quayle, B., Geron, C., Belote, A., McKenzie, D., Zhang, X., O'Neill, S. and. Wynne, K.K.: Estimating

emissions from fires in North America for Air Quality Modeling. Atmos. Environ.„ 40, 3419-3432.2006. Ginoux, P., Prospero,J.M. Gill, T. E., Hsu ,N.C. and Zhao, M.: Global-scale attribution of anthropogenic and natural dust sources and their emission rates based on MODIS Deep Blue aerosol products, Rev. Geophys., 50, RG3005, doi:10.1029/2012RG000388,2012.

Minor changes [Title: evaluation] A: It is corrected.

[What is GEM-AQ/EC? Need to define it somewhere.] A: It is defined in the first paragraph of section 2. GEM-AQ/EC description. [P9288 L5: Gong 2003 not in the reference.] [P 9292 L5-6: Sentence not completed.] A: both are completed. [L19-20: What number did Mahowald (2003) get? That seems to be purpose of mentioning this work here.] A: The whole sentence of L19-20 is cut. [L21: What is tkm-2? Ton/km2?] A: yes. It was changed to ton km-2. [L24: change type-monthly to type monthly.] [Cross the text: change AeroNet to AERONET.] [Figure 1. the ten years of] [Figure 7. Behavior.] A: Thank the reviewer for the detailed check. We have done all the technical corrections.

[Figure 9. It seems that the emission data you are using do not perform well over the western US, even for sulfate is known to be best simulated by CTMs.]

A: The sulfate emission data used in the modeling study is from the GEIA emission inventory Version 1A and is based on year 1985. As it is mentioned in section 3.4, the accuracy in the emission inventory of anthropogenic sulphur species may have played a role in the fact that sulfate is not well modelled over western US.

[Figure 11. Surprisingly poor performance. Any clue of what is missing there?]

A: The increasing trend in PM-emissions has contributed to the discrepancy between model simulated and observed aerosol concentrations in Asia in Fig. 11. More accurate emissions in terms of spatial and temporal resolutions are needed to better simulate aerosol concentrations in Asia.

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