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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Chemically aged and mixed aerosols over the Central Atlantic Ocean – potential impacts" *by* M. Astitha et al.

M. Astitha et al.

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Reply to the reviewer's comments

Interactive comment on "Chemically aged and mixed aerosols over the Central Atlantic Ocean – potential impacts" by M. Astitha et al. Z. Levin (Referee) zevlev@post.tau.ac.il Received and published: 8 March 2010

This work uses chemistry transport models in combination with in-situ and remote sensing data to discuss the composition and concentrations of continental aerosols over the central Atlantic Ocean. They report that in the hurricane genesis region, very low concentrations of accumulation size aerosols of natural or anthropogenic sources were found, while the concentrations of Aitken size particles dominated the size dis-

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tribution. By calculating the vertical distribution of the different aerosols they conclude that many of the aerosols interact with clouds but not always from cloud base. Some of the results point out that sulfate and mineral dust enter through cloud tops, through the side by entrainment and also through cloud base. Sea salt particles, which they find to mostly concentrate at lower levels, enter clouds through their bases. The issues addressed in the paper are relevant to our understanding of the role of aerosols in the formation of clouds in general and of tropical storms in particular. The paper is clearly written but before publication the following points should be addressed.

[Author]: We would like to thank the reviewer for the constructive and useful comments on the manuscript. We respond to the issues raised by the reviewer below and we carefully address his questions and suggestions in the revised version of the manuscript.

General points: There should be a short discussion and description of some of the main chemical processes that are included in the model (e.g. the mechanism by which dust particles get coated with sulfate in the clouds). Also it is not clear to me if the model includes scavenging of aerosols by precipitation size particles. This point needs to be clearly shown.

[Author Reply]: We have added a description of the mechanism for the heterogeneous chemical uptake of gases on the surface of dust particles (in section 2: Models and data used). The CAMx model includes wet deposition processes using separate scavenging models for gases and aerosols. The numerical method treats the uptake as a function of rainfall rate, cloud water content, PM size. Aerosols are irreversibly scavenged directly by all precipitation forms via impaction and by uptake into cloud water (liquid and ice) as condensation nuclei that are scavenged by all precipitation forms. These details have been added to the manuscript as suggested by the reviewer.

Specific comments: 1) Page 5187 –Line 8 – The reference to Lelieveld et al 2002 is not appropriate here because in that paper the discussion was about transport to the Mediterranean area. Although the effect of North America pollution on the Mediter-

ACPD
10, C3109–C3115, 2010

ranean is discussed, it is not about the transport of pollution from Europe and Africa to the Atlantic.

[Author Reply]: We have removed the reference as suggested by the reviewer.

2) Page 5187 –Line 14 – better references at this point could be Andreae et al, 1986 (Science), Levin et al 1996 (JAM) and Levin et al 2005 (JGR).

[Author Reply]: This is done in the manuscript.

3) Page 5187 –Line 16 – should be Levin and Cotton 2009 and references therein.

[Author Reply]: This is done in the manuscript.

4) Page 5187 – Line 16 to 19 – the sentence needs improvement

[Author Reply]: We agree with the reviewer and the sentence has been revised accordingly: "Anthropogenic aerosols can cause a direct radiative forcing of climate (Forster et al. 2007 and references therein), and, acting as CCN, IN or by locally absorbing solar radiation can interact with clouds and precipitation, thus causing indirect climate effects (Ramanathan et al. 2001, Denman et al. 2007)."

5) Bottom of page 5189 to the top of 5190 – Here the authors mention the fact that the two models do not interact, namely no feedback from the chemistry back to the SKIRON model. In some cases this point could be important. For example, changes in the chemical composition of the aerosols could affect the radiation and thus affect the temperature field and other meteorological parameters. For example, Alpert, et al., 1998. Nature 395 6700, pp. 367–370 showed that errors of a few degrees in the forecast of temperature could be caused by not including dust in forecast models. This possible limitation should at least be mentioned in the paper.

[Author Reply]: We agree with the above comment that the feedback effects of chemical composition into the atmosphere can be very important. Unfortunately, this can only be treated properly by the use of an online atmospheric-chemistry model and not with

ACPD

10, C3109–C3115, 2010

the modelling tools available for this study. We have added a short discussion on this in the manuscript as suggested by the reviewer.

6) Page 5192, line 14 – Although the concentration of Giant CCN is small, their effects on precipitation development could be large. This point needs to be mentioned with proper references.

[Author Reply]: We have added this information in the specified section as suggested by the reviewer (section 3:Results and Discussion).

7) Page 5192 – line 15 – I cannot see how the sulfate particles can act as IN. They are efficient CCN but not IN.

[Author Reply]: We fully agree with this comment, although Ramanathan et al. (2001) refers to it in the indirect radiative forcing section of the paper in Science (page 2121). We have made appropriate changes in the text to avoid any misunderstandings.

8) Throughout the paper the authors use the term “concentration” when referring to mass. This is confusing to me. I suggest to either use mass concentration or mass loading to differentiate it from number concentration.

[Author Reply]: This is done in the manuscript according to the above suggestion.

9) Page 5195 last line and first line on the next page – The size should be referred to as geometric mean diameter.

[Author Reply]: This has been added in the manuscript.

10) Page 5196 – line 16 - The assumed density of the particles should be mentioned here.

[Author Reply]: The density of the particles used for the simulation has been added to the text accordingly. (Dust=2.6 g/cm3, PNa=2.0 g/cm3, PNH4=PNO3=PSO4=DSO4=DNO3=1.5 g/cm3).

ACPD

10, C3109–C3115, 2010

11) Page 5200 – first paragraph – AOD over the land suffers from much bigger errors than AOD over the ocean. Could this be one of the reasons for the differences between model and observations? Please elaborate.

[Author Reply]: We would like to thank the reviewer for his idea, on which we have elaborated and concluded the following: For this study we have used the MODIS Collection 5 dataset based on recent aerosol algorithms and calibration coefficients. According to studies comparing this satellite product with previous ones and AERONET (Remer et al., 2008 (JGR); Papadimas et al., 2009 (ACP)) the discrepancies over land have been reduced. Nevertheless, there is a need for caution in addressing the differences between model and satellites as a result of the AOD errors over land. Moreover, a quick qualitative comparison with AERONET data at Izana and Tenerife for these specific days (15-17 August) did not reveal large differences with the MODIS AOD. Based on the above we shall keep the discussion on the differences (page 5200) as it is.

12) Page 5200 line 26 – the Liquid water content, LWC, in convective precipitating clouds could be much higher than 0.3 g/m3. In some cases, such as in deep convective clouds over the ocean they can exceed 3 g/cm3.

[Author Reply]: We have added this information to the manuscript.

13) Page 5201 line 14 – You should specify (as you do in the figure captions) that the unit micrograms/cm2 is an integral over height.

[Author Reply]: This is done.

14) Page 5203 – line 2 – The model results that high concentrations of sulfate particles are found above clouds agrees with some observations in other parts of the world where pollution particles are located above dust storms and above clouds (e.g. Levin et al 2005, JGR).

[Author Reply]: We have added this information in the manuscript as suggested by the reviewer.

ACPD

10, C3109–C3115, 2010

15) Page 5206 line 23 – high concentrations of coated dust, suggests that these could also affect the warm portion of the clouds by modifying the size spectra of the cloud drops, leading to more rapid development of precipitation.

[Author Reply]: The specific paragraph in the conclusions section refers to the dust particles, without any coating from sulphates or nitrates. The very important suggestion by the reviewer will be a speculation for the conclusions of this specific work and we have been careful in not concluding beyond the modelling capabilities.

16) Page 5207 – last paragraph – this paragraph is confusing and needs to be revised and clarified. The role of aqueous chemistry in the clouds is not included here, while it is well known that such chemical reactions could increase the number of coated dust particles. This is especially true if a number of cycles through clouds occur (e.g. Wurzler et al, 2000 [JGR]).

[Author Reply]: We have tried to clarify this paragraph as suggested by the reviewer. Aqueous phase chemistry is part of the modelled processes (aqueous sulphate and nitrate formation in resolved cloud water) for the anthropogenic species. What is missing is the interaction between aerosols and clouds leading to an explicit treatment of the CCN and/or IN produced. One important note here is that the percentages of sulphate on dust (calculated as DSO4/DUST*100) in terms of number distribution, refer only to the findings in the 6 point locations (P1-P6) in the Atlantic Ocean. The percentages do not cover specifically the cloudy areas but the whole vertical distribution in each location point. The aim of making such calculation was to show the relative abundance of the sulphates produced on dust compared to the dust particles available, solely by the use of the described methodology. Our model is not suited to address the cloud microphysics and cloud-aerosol interactions and we have mentioned this in the manuscript. We fully agree with the reviewer on the role of aqueous chemistry in the clouds, but we feel we cannot expand speculation far beyond the model capabilities in this study.

17) The role of coating in reducing the ice nucleating efficiency does not apply uni-

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10, C3109–C3115, 2010

versally. There could be some coatings that could increase it. This de-activation may not take place if the coated particles are immersed in the water drops. The soluble component is expected to be dissolved, leaving the mineral dust free of coating. The coating may reforms on the aerosols when the drops evaporate. To my knowledge, not too much work has been done on this last point.

[Author Reply]: We thank the reviewer for his valuable thoughts on this matter. The short mention about the ice nucleating efficiency in the conclusions has been removed.

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 5185, 2010.