Atmos. Chem. Phys. Discuss., 10, C1660–C1666, 2010
www.atmos-chem-phys-discuss.net/10/C1660/2010/

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.

Anonymous Referee #4

Received and published: 15 April 2010

GENERAL COMMENTS:

The contribution shows a simulation of a chemistry-transport modeling system (SKIRON-dust/CAMx) for the month of August 2005. It is focused in the distribution of the aerosol number concentration and its chemical origin-composition (desert dust, sodium from sea salt, sulphate from anthropogenic sources and sulphate on dust particles) over the tropical north-eastern Atlantic. Validation of the model performance is proposed by comparing:

1) Simulated 24-hour averages of the Total PM10, Total PM2.5 and Sulphate PM10 mass concentrations, with observations at a selection of surface stations distributed in the whole domain.

2) Simulated 2-hour averages of the number concentrations at different size ranges, with observations at a selection of 4 European stations (3 of them at the top of mountain sites)

3) Simulated 24-hour averages of the aerosol total mass load, with observations by the Moderate Resolution Imaging Spectro-radiometer (MODIS) aboard the NASA Terra and Aqua satellite platforms.

The work follows the series of articles (mentioned in section 1-Introduction) which are drawing the attention into the impact of the European pollution (and dust) in the Tropical Atlantic and its eventual intercontinental transport into America.

The article has its interest in showing aged anthropogenic aerosols (sulphate), with a likely European-Mediterranean origin, mixed with desert dust, arriving at the Eastern Tropical Atlantic. Dust coated with sulphates, after chemical reactions during the transportation process, has been simulated by the modeling system, after including the heterogeneous uptake of gases on dust particles in CAMx. This mixture of aged aerosols and dust, with an important number concentration in the Aitken mode, as shown by the model simulation, could be playing an important role in the cloud formation, tropical storm generation and their possible westward transportation/developing into Hurricanes. In my opinion, this contribution could be published in ACP, but a more rigorous approach is necessary to show and explain the results. I hope the following observations may help to improve the manuscript before it is finally accepted.

My main objections to the contribution are related with the following points:

a) Evaluation of the modeling system, which could be clearly improved

b) Presentation-interpretation of the main results: some of them are difficult to understand following the explanation of the authors

c) Lack of clarity in the figure depiction, which has to be made more comprehensible for the reader: this point is directly related with the preceding one.

SPECIFIC COMMENTS:

a) Evaluation of the modeling system

I completely agree with the comments of Referee #3 on the comparison of the simulated number concentrations at different size ranges, with observations at a selection of 4 European stations shown in Figures 2 and 3. I will add that the low performance of the evaluation of the number concentration by the modeling system (Figure 3), which clearly overestimates at low concentrations, is casting a shadow of doubt to the number concentrations of the different species (desert dust, anthropogenic sulphates, sulphates on dust and sodium) mentioned in the description of results (section 3.4) and in the conclusions (section 4) of the manuscript. I will ask the authors to estimate the uncertainties (interval estimations) of those results and include them within the results and the conclusions of the manuscript: at a first glance, it seems that the uncertainty could be as high as the estimation value for most of the modeling results in the tropical Atlantic region. The authors have to realize that this is a key question for the reliability of their results. However, contrary to the opinion of referee 3, I think that the heights of the selected stations (which have to be included in the manuscript), are not in the origin of the mentioned low performance: the Melpitz station, shown in Fig. 3b, is at 86 m a.s.l. and it shows the highest Y interception value (845 cm-3). In addition, the selected resolution for the Skiron/Dust modelling system (shown in Table 1) is enough to resolve the mountain meteorology of the other three stations shown in the comparison.

A contrary behavior of the modeling system is found when evaluating 24-hour averages of the Total PM10 and Total PM2.5 mass concentrations (shown in Fig. 2a-b and Table 2): the model seems to underestimate at low mass concentrations, except for the sulphate PM10, and shows a tendency to underestimate at high concentrations. One of the main problems of this validation is that all the 10 sites have been grouped, and following the same arguments explained by referee 3, I will suggest to show regional differences in the model performance, using spatial plots of NMB or MB using color codes. The authors can also plot bias and errors against average concentrations in

order to show the model's ability to predict at low and high concentrations. Alternatively time series plots of model and predicted daily concentrations will be of a great help to understand the model behavior.

The third group of the model validation tasks encompassed by the authors refers to the daily averages of the aerosol total mass load, which have been compared with observations by the Moderate Resolution Imaging Spectro-radiometer (MODIS). This comparison adds no much information for the reader of the article. The MODIS data show just a plume of desert dust over the ocean with no continuity over the continental land mass of northern Africa. As acknowledged by the authors, the modeling system has problems to locate correctly the aerosol plume (line 25-28, page 5199) over the ocean, which they attribute to temporal mismatches between satellite and simulations. At this respect, the authors should explain why they did not select a "more simultaneous" model output "more coincident" to the satellite passes instead of representing daily averages. Model dislocation/under-estimation of the MODIS aerosol plume over the Canary Island is observed in Figure 4 (15 and 16 August), as stated by the authors, but in Figure 5 (20 and 28 August) the model has also problems in estimating the plume location and/or the total column mass load over the Canary Island (20 August) and SW of Cape Verde (28 August), and this is not mentioned in the article. AERONET data in Cape Verde and Canary Islands will help to confirm/explain this type of differences. As the authors know, dust is the major aerosol source for the observed light extinction in the target region. The Skiron/Dust estimated optical depth can also be used to validate the AERONET or satellite AOD data not only over the ocean but over the continental land mass. This has been already used by two of the authors of the manuscript in a recent contribution published in Environmental Fluid Mechanics (Astitha and Kallos, 2009). Thus, this reviewer can not understand why the authors are reluctant to use this source of data to comparison (lines 15-17, page 5199).

My last objection on the validation methodology and its results comes from an important absence: the meteorology simulated by Skiron/Dust has not been compared with observations. The authors can argue that it is not longer necessary this type of evaluation when the concentrations fields have been adequately estimated (which is not the case). I will tell the authors that it is a good practice to make this type of evaluations using surface and upper air data distributed in the whole domain of simulation: it is the only way to know if the over- or under-estimations of the modeling system (Skiron/Dust and CAMx) is due to emissions, meteorology, or general selection of model setup and input parameters. Cloud cover, precipitation and column water vapor are also satellite products that can be used for evaluating the meteorology at selected days, as those used to discuss the "typical aerosol number concentrations" in the tropical Atlantic (16 and 27 August 2005). None of this work has been done. I mentioned explicitly cloud cover and precipitation, because the first item is used by the authors for case selection in section 3.3 and the second is a key variable driving the aerosol wet deposition. At this moment we do not know if the clouds represented in Figure 6, which are shown to justify the two days used to discuss the main results, are real or a model dream.

b) Presentation-interpretation of the main results

I will comment here the results and conclusions presented in section 3.4 and section 4, respectively. Section 3.4 is devoted to describe the Figures 9 to 14 of the manuscript, where vertical profiles of condensed cloud water concentrations and selected aerosol number concentrations are shown at 2 different size ranges and 2 different days (2-hour averages, at 4 different times throughout each day): 16 and 27 August. As acknowledged by the authors themselves, the locations were selected with the criterion of simultaneous presence of clouds and absence of rain (page 5202, lines 12-13). However, Figure 1 of the manuscript shows both the simulated Skiron/Dust mean sea level pressure and the 12 h accumulated precipitation, showing rain from 00UTC to 12 UTC at the target area. The authors must explain this point.

I would suggest first, before going into the discussion of results, to confirm the presence of clouds over the area (I personally checked this point and found no clouds at the coastal area of Western Sahara for both days: 16 and 27 August) and the absence

of rain, both using simulations and satellite or "in situ" data (evaluation of the meteorology). After this, the arguments given to justify changes in the concentrations of the vertical profiles of aerosols have to be changed (Lines 20-24 in page 5203).

After my personal experience in analyzing data from the target region, over the western coast of Africa, to the north of the region of the West African Monsoon, and coincident to the target region of the manuscript, we can find a relative cool marine boundary layer (MBL) topped by an inversion layer (the trade wind inversion). The relative warm Saharan air layer (SAL), above the inversion, transports the main fraction of the African dust plume with the easterlies blowing at the SAL. Thus, because of the permanent presence of the temperature inversion above the MBL, an important wind shear can be found at the region, with northerly winds at the MBL down to latitude 15-20 N and the easterlies at the SAL. This is the main reason why to properly explain the origin and evolution of the different layers that can be seen in figures 9 to 14, the corresponding temperature and wind profiles are also to be drawn. This could help to understand the differences observed in the vertical profiles of dust (CRST) in Figure 12 and Figure 13. The authors explain the 'anomalous pattern' in the vertical CRST profiles at P4 (Fig. 12) by the effect of the continental proximity (lines 19-22 in page 5204), but P2 and P3 are equally close to the continental area and they show a completely different vertical distribution of dust, more similar to that observed in locations P4 and P5

Although the authors mention the aerosol nitrate (anthropogenic and formed on dust) in the abstract and in the description of the composition of PM10 and PM2.5, their distribution is neither displayed nor discussed in section 3.4, focused in the target area. However, their presence and distribution is important for any reader interested in the ocean and rain forest fertilization. In my opinion, they should be included in that section.

c) Lack of clarity in the depiction of Figures

1) All panels included in Figures 4, 5, 6 and 7 should be redrawn in order to show latitude-longitude units.

2) Figure 3a has no information of the aerosol size range (panels b and c do have). Heights of all stations have to be included in Figure 3.

3) Please, plot the points P1 to P6 in Figure 7, in order to interpret this figure together with the vertical profiles shown in Figures 9 to 14.

4) As comment above, authors should include at least temperature and wind in Figures 9 to 14.

OTHER SPECIFIC COMMENTS:

The title of the manuscript should be changed to include the word modeling, otherwise the reader could understand that aerosols have been measured over the Atlantic Ocean, and this is not the case. In addition, the target region is not the Central Atlantic Ocean, mentioned in the title, but the tropical north-eastern Atlantic, in agreement to referee 3.

Table 1:

a) What type of nudging has been used in the SKIRON/Dust modeling system: Did the authors include just the boundaries of the domain or the whole domain? Considering the large time coverage of the simulation, this is an important question to be addressed in the manuscript.

b) Emissions from ships have been included using the EMEP database. Considering that its coverage does not include the southern region of the simulation domain, the authors should comment their possible impact (nitrates and sulphates) in MBL of the target region.

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