Atmos. Chem. Phys. Discuss., 10, C3131–C3146, 2010
www.atmos-chem-phys-discuss.net/10/C3131/2010/

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.

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Received and published: 25 May 2010

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.

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.

[Author reply]: We would like to thank the reviewer for the useful comments on the manuscript. We have made changes in the revised version of the manuscript, taking into consideration the comments from the other 3 referees related to the issues raised in this review.

SPECIFIC COMMENTS:

a) Evaluation of the modeling system

1) 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.

[Author reply]: We are kindly referring Referee #4 to our detailed reply to Referee#3 about the comments for the comparison of the simulated number concentrations. We have also included in the manuscript a second table (Table 2b) with the statistics for

the number concentration from model and observations, according to the reviewer's suggestion. A discussion on the statistics for each station has been added to section 3.2 for each relevant station. From the statistics it is again obvious that the model did not reach a 1:1 relationship with the measured concentrations, as expected due to a number of uncertainties that follow the simulations with an air quality model (emissions, horizontal resolution, boundary values, calculation of the number distribution). The overall statistical evaluation indicates a reasonable model performance with respect to the PM10, PM2.5 comparison. The "shadow of doubt to the number concentration of different species" as the reviewer suggests, is a problem that all modelling systems face due to the absence of number concentration measurements for the speciated aerosols. On the other hand, the use of only four stations can not be considered adequate for a systematic model evaluation that can result into solid conclusions on the reliability of the model results, but rather provide an overall picture of the model's ability to describe the number concentration values in an acceptable way. In this work, we have made an attempt to assess the typical range of number concentrations, focusing on the higher values found entering the area of interest, and that within the limits of an agreeable model-data intercomparison. Again, the limited availability of data for the specific month of the year does not allow for a systematic evaluation of the model performance which is not the goal of the present work.

2) 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- 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.

[Author reply]: We do not fully comprehend what is the "contrary behaviour of the modelling system" that the reviewer suggests, since the underestimation of the measurements is evident in the entire range of the concentrations as expected. There are some values of the model that overestimate the measured concentrations but these are limited compared to the overall intercomparison. For clarifying the above behaviour we have provided a supplement with the time series plots of PM10 and PM2.5 daily concentrations for each station, as suggested by the reviewer. Due to the already numerous plots in the manuscript, we have chosen to include these plots in a supplement to avoid making the paper more difficult to read. For the grouping of the available stations we are kindly referring to our answer in Reviewer #3 (comment 4). We also believe that by providing the time series plots for each station we help the readers to identify the regional behaviour of the modelling system. One last point that we should clarify is that in this paper we do not attempt to validate the model results. We limit our work to a model-data intercomparison, which aids the evaluation of the overall performance of the air quality modelling system.

3) 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 ontinental 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).

[Author reply]: The use of the MODIS data was not meant for evaluation of the model results (validation was not our target in any part of this manuscript). As stated in the text, these products are used for qualitative and not a pixel-to-gridcell comparison, providing an overview of the spatial structures and relative magnitudes. In addition, the use of columnar mass loads gives an opportunity to assess the vertical dimension which is something missing from the point location values given by the station observations. Based on the above we believe that providing this additional information in the paper is worthwhile. The model did not match the MODIS data in all locations, as stated in the manuscript and commented by the reviewer, and there are many reasons to expect such outcome. One being that the model results are based on time averages (1h average meteorology and emissions, 2h average concentrations) and not instantaneous values, so the use of more simultaneous model output to the satellite passes would not be possible anyhow. And if we would choose a 2h average concentration close to a pass of the satellite, the result would still be in question because of the temporal mismatch of the compared data. We have chosen August 16 and 27, 2005 and the specific point locations (away from the Canary Islands) based on the triple criterion mentioned in the manuscript but also on the overview of the satellite vs model columnar mass loads. In the text, we do mention the spatial dislocations for the 28th of August (page 5199, lines 25-28: "The weaker match for the 14th along the coast and the spatial dislocation on the 28th may be related to temporal mismatches since the satellite observations occur in the middle of the day and the model output represents daily averages") and we have added the 20th of August as mentioned by the reviewer. The dust optical depth calculation in our previous publication is based on the SKIRON/Dust simulations and it is derived in a post-processing stage using a simple algorithm. In the paper mentioned by the reviewer, the dust optical depth was used for a 1-day episode of dust intrusion in Athens, where the contribution from other sources was negligible. In this work, the amounts of anthropogenic and other natural aerosols are important in the area and to include such calculation would be meaningful and possibly lead to misunderstandings instead of useful information. For the above reason we have stated in the manuscript that "the aerosol optical depth is not a standard model output" (page 5199, lines 15-17). Despite our decision not to include the dust optical depth in this paper, we have already performed the exercise for 16 and 27 August (the days selected for the number concentration analysis). For the area of Central Eastern Atlantic Ocean the only available AERONET stations were Izana and Santa Cruz Tenerife, whereas Capo Verde and Dakar did not give values for these days. The comparison is not conclusive as you can see from the plots in Figure 1 in this response and it was another reason not to proceed with this in the paper.

4) 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 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.

[Author reply]: We do not entirely understand the reviewer's comment "the meteorology simulated by Skiron/Dust has not been compared with evaluation when the concentrations fields have been adequately estimated (which is not the case)" and we assume that the comments that follow this sentence are related to that and we will reply to the next ones, hoping to meet the reviewer's suggestions. This is work being done for almost 30 years by some of the co-authors of this manuscript. Testing the meteorology before proceeding to the air quality simulations is a standard method that has been performed by the authors. We will recall the general problem for the analysis fields prepared from large meteorological centres such as ECMWF and NCEP. It is well known that the surface and upper air observations of the entire Sub-Sahel region are of bad quality or they even suffer from lack of data. Having a regional model run for one month it is obvious that for some days the model-data intercomparison is better and for some others is worse. Checking the meteorological model performance was the first step we have done and we find no useful information to provide in this paper. In general we can say that this intercomparison gave us the go-ahead to use the meteorological fields for the air quality simulations. Adding this information is out of the scope of this paper. The atmospheric model SKIRON/Dust did not produce rain at the specific chosen locations during 16 and 27 August 2005 and that has been compared with the 3h TRMM rainfall archives for its validity. In the text we mention that there is absence of rain so the species did not undergo wet deposition (page 5203, lines 20-21) and we have added a sentence mentioning the use of TRMM as a verification tool. For the cloud cover we agree with the reviewer on the importance of including the verification of the appearance of clouds in the area for 16 and 27 August, although it is difficult to directly verify the calculated cloud water content (g/m3, 3-dimensional array) with a satellite product (except for the TRMM/TOVAS cloud liquid water which gives monthly estimates and thus unusable for our purpose). Also the satellite products usually give daily or monthly profiles for the cloud cover and cloud optical depth but we have included them in the manuscript (in Figure 6) to justify that our analysis is not based on a "model dream". We should mention again here that the chosen locations P1 to P6 are in areas with shallow cloud formations. For the reviewer's convenience we are presenting the relevant plots from the satellites in Figures 2a and 2b in this response, where it is evident that the locations P1 to P6 are in shallow cloud formation regimes.

b) Presentation-interpretation of the main results

1) 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.

[Author reply]: The reviewer has possibly misunderstood the accumulated precipitation fields in Figure 1. The accumulated field shown in the target area spans a range of 0.1-2mm for 12h accumulated precipitation. These low values are not indicative of actual precipitation, but they are rather common when the relative humidity is high in the area. Furthermore, the wet deposition process in the air quality model CAMx which is directly associated with the rainfall rate from SKIRON/Dust model, showed zero wet deposited fields for the species for those specific days in the target area.

2) 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).

[Author reply]: The confirmation of the presence of clouds in the area is addressed in our response in the previous comment with the use of the MODIS cloud fraction fields (Figures 1 and 2). About the absence of rain we have explained our methodology in the same previous comment that neither the SKIRON/Dust model nor the TRMM satellite product (3h) gave rain amounts at the specific locations. Also the wet deposited amounts of species in the CAMx model were zero for those days and locations. Based on the above we see no reason in changing the arguments in our discussion, as proposed by the reviewer.

3) 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.

[Author reply]: We completely agree with the reviewer on the structure and the physics of the Saharan air layer in the area under investigation. The depth of the dust layers over Sahara vary significantly over day and night. The transport of dust plumes injected during different hours of the day can be found later on over the sea at different layers also. These features are easily explainable and are already known from a number of publications in the past and we did not expand our discussion towards that direction knowing it is not novel work. Drawing the wind and temperature fields in such complex figures (Figures 9 to 14) will only make them unreadable and very difficult to analyze. The issue of different vertical structure of the different aerosol species is important and interesting to analyze but it is a secondary issue in the present work. Our emphasis is given on the range of number concentration values found entering the area from a mixture of natural and anthropogenic aerosols. About the differences in the vertical structure of the dust plume in point P4 compared to the other locations, one should keep in mind that the P4 is closer to land, as P2 and P3 are, but most important it is closer to the outflow region of the dust plume. As P4 is located further south is receives the largest amount of dust from the Mauritania and Western Sahara compared to the locations P2 and P3 which are close to land but in a higher latitude.

4) 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.

[Author reply]: We fully agree with the above comment but discussing and analyzing nitrates is an entire paper by itself. We have already a lengthy paper that we do not

wish to make it more difficult to read.

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.

[Author reply]: This has been done accordingly.

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.

[Author reply]: This has been done accordingly.

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.

[Author reply]: This has been done accordingly.

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

[Author reply]: As already commented by Reviewer #3, the Figures 9 to 14 are complicated enough for the reader to follow. Adding meteorological fields in them will only make them unreadable.

OTHER SPECIFIC COMMENTS: The title of the manuscript should be changed to include the word modeling, otherwise the read er 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.

[Author reply]: We have agreed with referee#3 to change the title of the manuscript to "Chemically aged and mixed aerosols over the Eastern Central Atlantic Ocean – Potential impacts". After the new suggestion we have added the word modelling to our title: “Modelling the chemically aged and mixed aerosols over the Eastern Central Atlantic Ocean – Potential impacts”

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.

[Author reply]: The way the systems runs is by using the ECMWF 3D fields for initialization and nudging of the lateral boundaries. This information has been clarified in the manuscript in section 2 (Models and data used).

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.

[Author reply]: The reviewer is correct about the missing shipping emissions from the Central and Southern Atlantic Ocean, as neither EMEP nor GEIA inventories include this area in their database. We have added a comment on that in the text as suggested by the reviewer (section 2: Models and data used, paragraph 8).

Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/10/C3131/2010/acpd-10-C3131-2010-supplement.pdf

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

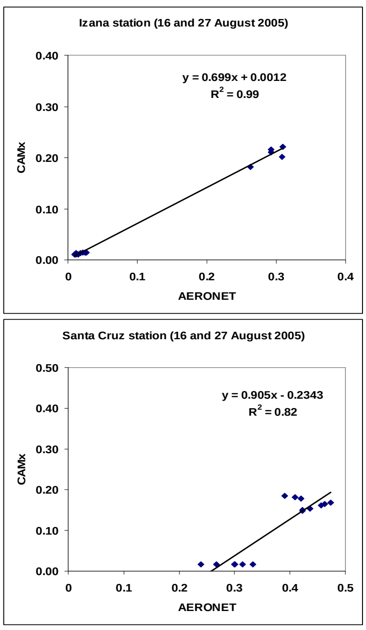

Figure 1: Comparison between the dust optical depth calculated by the CAMx model and Aerosol Optical Depth from AERONET for 16 and 27 August 2005.

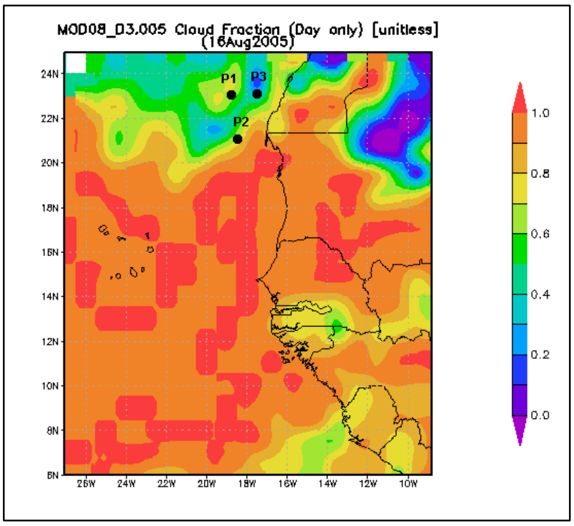

Fig.2a: Daily cloud fraction from MODIS Terra v.5 for August 16, 2005. The black dots denote the approximate position of the first 3 locations (P1 to P3) used in this work.

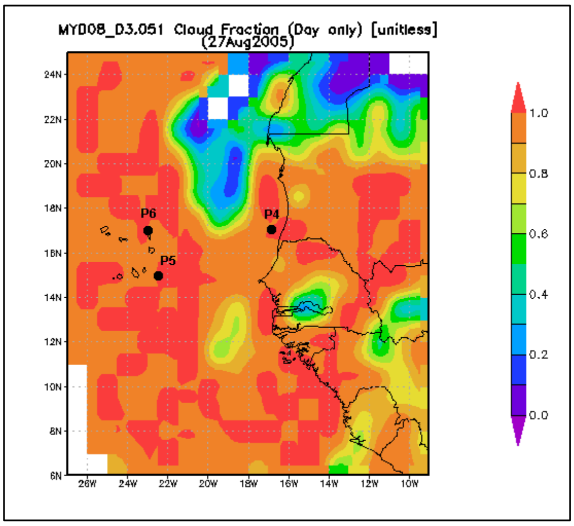

Fig.2b: Daily cloud fraction from MODIS Aqua v5.1 for August 27, 2005. The black dots denote the approximate position of the last 3 locations (P4 to P6) used in this work.