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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.

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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.

Anonymous Referee #3 Received and published: 7 April 2010

This paper presents the results of a chemical aerosol transport model and it compares these results with satellite and in situ measurements. The study focuses on August 2005, a period when there was intense hurricane and tropical storm activity over the Atlantic Ocean.

I am not qualified to comment on the technical aspects of the model. I restrict my comments to the generalized nature of the results in the context of my research in this general region. I refer readers to another reviewer, Zev Levin, who presents a more comprehensive review that touches on the issues of the model and the cloud-process related aspects of the results. In general, the study presents some interesting results. However certain aspects of the paper could be improved by a more focused analysis.

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

1) My major comment is that the title of the paper does not capture the coverage of the paper. The results focus on the eastern Tropical North Atlantic Ocean.

[Author Reply]: We agree in changing the title to: "Chemically aged and mixed aerosols over the Eastern Central Atlantic Ocean – Potential impacts".

2) The model focuses on three aerosol mass fractions: Total PM2.5, Total PM10 and Sulfate PM10. Also three size ranges: 0.03–0.1 μm (Aitken mode) and 0.1–2.5 μm ("accumulation mode") and PM10. I would argue that the particle size range used for the "accumulation" mode, 0.1 to 2.5 μm diameter is too broad. The extension to 2.5μm includes a great deal of the dust mass. The mass median diameter of dust over the eastern Atlantic to the Caribbean is about 3 μm. I assume that this was done because of the desire to compare with the PM 2.5 measurements made at air quality stations. However many of the European and Mediterranean basin sites are impacted by pollution sources (which do, indeed, contain substantial mass in the accumulation mode as usually defined, 0.1 – 1.0 μm diameter) whereas, in contrast, the stations along the west coast of North Africa will be heavily impacted by dust much of which will fall into the "accumulation" size mode as defined in this paper. Thus the testing of the model against the European sites may not be ideal for the intended purpose of the model.

[Author Reply]: In principal we agree with the reviewer's critical comments on the usage of the 3 size modes to describe such a broad range of pollutants. We would ideally prefer to use up to 10 different sizes (or more) but at this stage we were restricted by two constraints: one was the online sea salt emissions production which is formulated in these 3 size sections and second was the need to compare with available measurements in the area. Also, using only European sites for the model evaluation was a necessary and not a preferred step, as we could not have access to data from other parts of the domain (namely sites in the African continent or elsewhere). In principle it is not a problem that some of the desert dust is represented in the size range up to 2.5 μm, as this is also observed. The question is more whether this size range satisfies the definition of "accumulation mode", which was done for the practical reasons outlined above.

3) As acknowledged by the authors, the model tends to substantially underestimate the measured concentrations as shown in Figure 2 where the regression slopes range from 0.37 to 0.6 for the three classes of aerosols. The authors state that this result "is expected, since the model simulation did not include organic aerosols. . .." While this is a reasonable assumption for some stations, it is not necessarily generally applicable. The underestimation of the model is particularly evident in panel Figure 2a, Total PM10; there is a heavy clustering of points where the model yields consistently low values compared to the measured values. One would not think that organic aerosols would contributed in a major way to PM10 concentrations. These data are obtained from very different environments, some dominated by pollution, some by dust, and some dust-marine.

[Author Reply]: The underestimation of the measured concentrations is more evident in the PM2.5 diagram (Fig.2b) than in the PM10 (Fig.2a). The clustering in the low modelled PM10 values is partially a result from the PM2.5 mass concentration as PM10 includes also PM2.5. We agree with the reviewer about the organics. In the paper we have mentioned the organics in the PM2.5 comparison and about the PM10 we state

in page 5195, lines 2-3: "The linear regression of the PM10 scatter diagram shows a relatively good model performance for the coarse aerosol, possibly because the organic aerosol mass is only minor." Most of the aerosols used in this study have their maximum mass concentration in the 2nd size (0.1-2.5μm), meaning sulfates, nitrates, ammonium, whereas mass is larger in the coarse size (2.5-10μm) for desert dust, sodium and chloride (from sea salt). Moreover, the coarse resolution of the model does not allow for a detailed representation of urban or suburban locations leading to the underestimation of the measured concentrations in these locations.

4) Rather than grouping all these data together as was done in this paper, it might be more meaningful to use data grouped by environment or region based on the very brief in Figure 2. The separation of the station data on this basis would allow us to better assess the model results. It is notable, for example that the regression in Fig. 2c, PM10 sulfate daily average values, is driven by a few very high values which I would suspect come from the Mediterranean stations. I suspect that if these were removed, the regressions would change substantially with a slope closer to 1. Also I suspect that the intercept would be closer to zero. As it now stands there is a very substantial Y intercept in the model estimate (1.87).

[Author Reply]: Although the reviewer gives an interesting idea about the statistical approach, grouping the data per region is not possible due to the limited number of stations available. We have chosen these stations based on the fact that they were influenced by dust and sulphate aerosols, on their types (the majority of the stations are rural background) and the availability of data for August 2005 (information on the selected stations appears in section 3.2, 1st paragraph). If we group the data per region we will limit the statistical significance of the results. Following the reviewer's advice we have investigated the regression for sulphates and it does not change significantly if we remove the 4-5 higher values from the diagram and the intercept is higher than before (2.018). The scatter diagram after removing the high values is shown below (see Fig. 1).

5) a) Figure 3 is a multi-panel graphic presentation of the comparison between modelled and measured number concentrations for four stations, all European. The data from two stations, Jungfraujoch and Puy de Dom are plotted in one panel, 3a; a single regression line is calculated. (What is the particle size range for Fig. 3a?) It is my impression that while the two distributions are similar, there may be some significant differences. Perhaps the regression lines should be plotted separately.

[Author Reply]: The data from Jungfraujoch and Puy de Dome are plotted in a single scatter diagram (Fig3a) because of their similarities; they use the same instrument for the measurements "... a condensation particle counter instrument (CPC TSI 3010)" and the particle size range is "... aerosol sizes larger than 0.01μm for FR0030 and in the range 0.005-10μm for CH0001 (diameter)". The regression lines are almost identical, making it hard to be distinguished if plotted separately. To be more precise we have plotted two scatter diagrams for the two stations as the reviewer suggested (see Fig. 2) to indicate their similarities. The idea of plotting them in one diagram was to save as much space as possible, since the result was the same.

b) In the cases of panel 3a and 3b, the regressions are largely driven by relatively few high values. The large mass of the data at relatively low concentrations (which are nonetheless rather high which suggests pollution impacts) are essentially uncorrelated, especially in 3b. Furthermore the model results in all three panels (i.e., all four stations) show that the model estimates are biased high at lower measured concentrations. All three panels yield Y intercepts that range from 616 to 845 cm-3. These intercept values are comparable in magnitude to some of the highest values estimated for the eastern Atlantic sites later in the paper.

[Author Reply]: As we state in the manuscript, the comparison of the modelled number concentrations with the available measurements is not a 1:1 relationship, given the coarse resolution of the simulation, the calculation of the modelled number concentration and the exclusion of organic particles. Trying to exclude high values from the statistical analysis is not always feasible, especially for stations like Melpitz (DE44, Fig.

3b) which has approximately 15% of the data values above 3000cm-3. Nevertheless, we performed the exercise suggested by the reviewer and in the diagrams below it can be seen that the changes are not significant for FR030 and CH01 (see Fig. 3). For the Melpitz and the Zugspitze station the lower values exhibit a weak correlation and this can be explained by the fact that the comparison is based on the 1st size section of the model (Aitken mode) due to incompatibilities between model and measured size sections. Our main goal was to present the comparison with all available data and the statistical analysis beyond the scatter diagrams gave us a reasonable confidence on the conclusions drawn by this work.

c) The use of data from mountain stations complicates the interpretation of the model performance. Mountains, especially these which are located on massive ranges (and contrasted, for example, to Izaña or Mauna Loa which have problems of their own) generate complex meteorology and circulation patterns that are difficult for models to resolve. There are also relatively local effects (upslope/downslope winds driven by local heating and cooling) which the model could not resolve.

[Author Reply]: In principle we agree with the reviewer's opinion on selecting the stations for the comparison with the model results. Nevertheless, in several cases as in our study, it was not feasible to acquire data from other locations since the observations were not freely available and/or the desired period of the simulation was not covered. The model resolution cannot resolve local effects as pointed out by the reviewer, and this is a well known problem for all modelling systems. Depending on the resolution one can lose and/or gain different types of information (boundary values, transport effects, local effects, missing emissions among others), which should be accounted for when interpreting the model results. In this work we are primarily interested on the long-range transport of pollutants and not on resolving local and complex circulation patterns. The results are concentrated on the maximum modelled number concentrations of the different aerosol species found in the Eastern-Central Atlantic Ocean. The model resolution is able to represent most of the mountainous areas, for example

Melpitz station is located in 87m asl and the terrain height representing the appropriate grid cell is 110m asl. The rest of the stations are in higher altitudes that the model represents in a satisfactory way. We believe that using stations at higher altitudes in such model studies has certain benefits such as less impact from local sources or boundary layer plumes that are horizontally transported.

6) In testing the model product the authors compare the model AOD against satellite AOD. While this is interesting, a more stringent test would be to compare against AERONET data which would also provide an estimate of fine/coarse aerosol fractions. A quick check of the AERONET site shows some data for the days of interest at a number of sites in the modeling domain including some island stations in the Cape Verde Islands and Canary Islands.

[Author Reply]: The satellite product we used in this work is not the AOD because with the model we do not calculate AOD. Instead, we used the columnar aerosol mass loads (g/cm2) from the MODIS instrument (which is calculated from AOD) and compared them qualitatively with the modelled aerosol mass loads (page 5198, last paragraph). Unfortunately AERONET does not provide such product to add it in the relevant analysis.

Fig. 9 to 14: These figures have some interesting aspects to them. But it is difficult to extract much information from them. There are so many. The paper reports (ca. 5201/13) that the desert dust maximum is 255 μg cm-2 on the 16th and 301 μg/cm2 on the 27 August. These column loadings are reasonable. If one assumes that the dust is carried in a layer several km thick, these loadings convert to dust concentrations of a few hundred μg m-3, values which are commonly measured on Izaña, Tenerife, during dust outbreaks.

[Author Reply]: We are happy to see that the reviewer finds our numbers within reasonable scales.

I am not qualified to comment on the portions of the paper dealing with cloud-aerosol

processes.

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

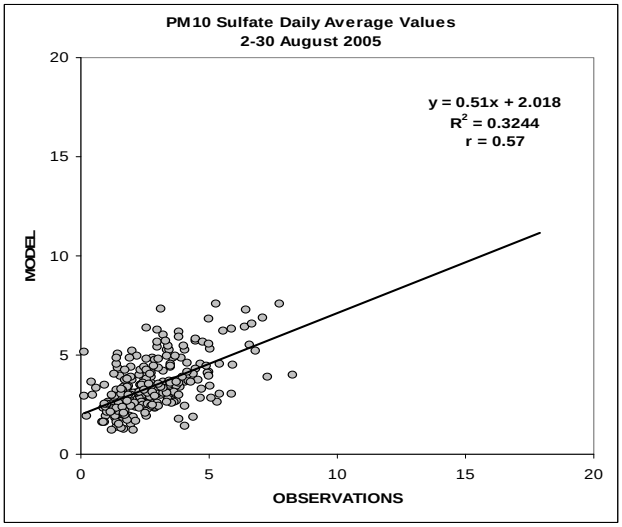

PM10 Sulfate Daily Average Values
2-30 August 2005
y = 0.51x + 2.018
R2 = 0.3244
r = 0.57
MODEL
OBSERVATIONS
0
5
10
15
20

Fig. 1

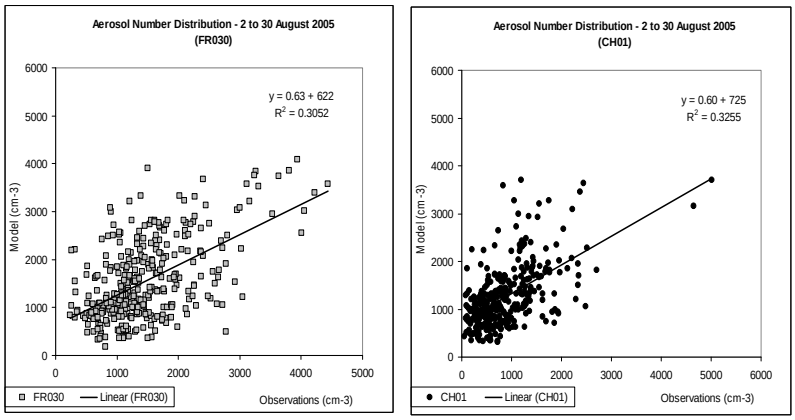
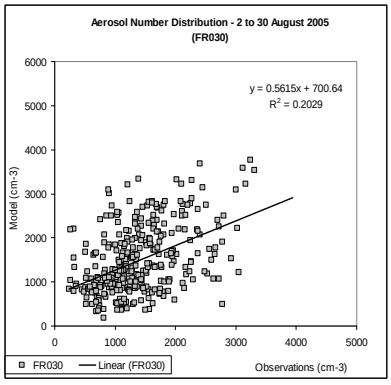
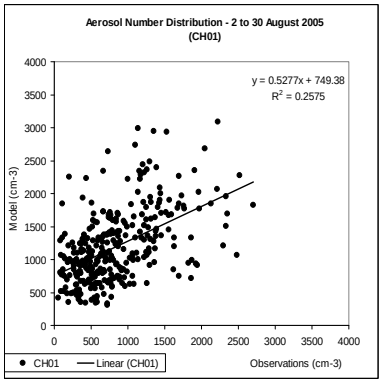
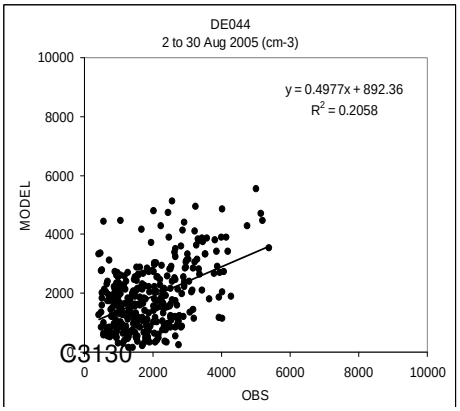

Aerosol Number Distribution - 2 to 30 August 2005
(FR030)
y = 0.63 + 622
R2 = 0.3052
Model (cm-3)
Observations (cm-3)
FR030
Linear (FR030)
Aerosol Number Distribution - 2 to 30 August 2005
(CH01)
y = 0.60 + 725
R2 = 0.3255
Model (cm-3)
Observations (cm-3)
CH01
Linear (CH01)

Fig. 2


Aerosol Number Distribution - 2 to 30 August 2005
(FR030)
y = 0.5615x + 700.64
R2 = 0.2029
Model (cm-3)
Observations (cm-3)
FR030
Linear (FR030)


Aerosol Number Distribution - 2 to 30 August 2005
(CH01)
y = 0.5277x + 749.38
R2 = 0.2575
Model (cm-3)
Observations (cm-3)
CH01
Linear (CH01)


DE044
2 to 30 Aug 2005 (cm-3)
y = 0.4977x + 892.36
R2 = 0.2058
MODEL
OBS

C3130