Atmos. Chem. Phys. Discuss., 10, C3610–C3618, 2010 www.atmos-chem-phys-discuss.net/10/C3610/2010/

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Interactive comment on "Simulation of aerosol optical thickness during IMPACT (May 2008, The Netherlands) with ECHAM5-HAM" by G.-J. Roelofs et al.

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Received and published: 7 June 2010

Answers to the reviewers, manuscript acpd-10-5911-2010.

We thank both reviewers for their close reading of our manuscript and their comments and suggestions. We discuss a few general issues first. Other comments and suggestions from both reviewers will be addressed after that.

- General -
- 1- Both reviewers: "correlations between observed and simulated parameters"

In the original document we discussed the similarities between daily variabilities in AOT, C3610

wind direction, number concentration and integrated water vapor. Based on the shape of the different time series we suggested that the correlation between AOT and IWV is larger than between AOT and other parameters, but without presenting quantitative numbers. In the present version we removed this paragraph, partly because accurate calculation of correlation parameters is relatively cumbersome considering the different temporal scales in observations and measurements. More importantly, in section 4 of the manuscript the influence of humidity on AOT is discussed on a more physical level involving discussion of the relevant parameters, which in our opinion is more informative than a statistical relation.

In this study we compare simulated aerosol and meteorological parameters representative of a grid point with a horizontal resolution in the order of \sim 120 km and on a 2-hourly time resolution, with observations from a single location and a time resolution varying from seconds to minutes. As in many model-measurement comparison studies one can ask how model data and in-situ measurements can be matched if temporal and spatial resolutions are different. It must be determined if a point measurement is representative for a larger area, or that the sub-grid scale variability over the area of a model grid box is significant, in order to evaluate a comparison as match or mismatch. This requires knowledge of the spatial distribution of emissions and specific lifetimes of trace species for chemical destruction, transport and deposition. We estimate that the transport time through a grid box in our model is typically on the order of one to a few hours whereas the lifetime of aerosol is on the order of a few days. This leads us to conclude that sub-grid scale variabilities in aerosol composition and concentration due to synoptic advection are probably small. The spatial extent of clouds, however, is generally smaller than the grid size, so that sub-grid scale variabilities may hamper an accurate comparison to some extent. In our study discrepancies between model

²⁻ Both reviewers: "the effect of spatial and temporal differences in the comparison of simulated and observed parameters"

and observations are sometimes large. Based on the results we argue that these discrepancies indicate basic model inaccuracies, e.g., inaccurate vertical mixing in the boundary layer or chemical processes that are not considered in the study, rather than inconsistencies due to resolution differences. However, for sulfate (Fig. 4a) the averaging of point-source emissions over the gridpoint does affect the comparison, which is discussed in the manuscript.

3- Both reviewers: "comparison of simulated and observed particle concentration profiles from different areas"

In our study we compare simulated particle concentration profiles at Cabauw, averaged over May 2-14, and average particle concentration profiles constructed from ten observed profiles from the same period but at different places in NW Europe. During a large part of this period the lower atmosphere over NW Europe was relatively homogeneous with a steady westward advection and cloudless skies. Our comparison of modelled and observed particle profiles yields at the least a qualitative assessment of the representativity of the simulated vertical profile and the order of magnitude of particle concentrations. These were found to be affected by inaccuracies in the vertical mixing in the boundary layer, both near the surface and at the boundary layer top, while the comparison of simulated and observed relative humidity profiles gives a similar view. During May 2008 three aerosol concentration profiles upto10 km altitude were measured near Cabauw, on May 6, 8, and 21. We compared these with the corresponding simulated profiles, but this is not shown in the manuscript. Qualitatively this produced the same result as the comparison of the average profiles, indicating the same mixing problems. For May 6 the agreement was further worsened by excess dust in the simulation which is not expressed in the observed AOT, as mentioned in section 3. We have discussed this in the manuscript now, but given the small number of observed profiles for Cabauw we decided to keep the figure as it is in the original manuscript.

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4- Both reviewers:" possible errors in converting simulated aerosol mass to AOT"

Most of May 2008 the aerosol over Cabauw was (almost) purely scattering, except for the last week when large amounts of dust resided over Cabauw. The model-measurement comparison of surface PM10 strongly suggests that the amount of dust is considerably larger in the model than observed, and this is also true for the amounts of sulfate and organics on these days. The latter overestimation may cause too large aerosol water uptake, and this contributes to the overestimation of AOT. But dust is also slightly absorptive and absorption properties of dust are highly variable and uncertain. The refractive index for dust applied in ECHAM5-HAM is 1.52+i0.0011, derived from AERONET measurements. Stier et al. (2005) mention that the absorption efficiency is smaller than other estimates. The scattering efficiency of an absorbing particle is smaller than that of a purely scattering aerosol, and this may be partly responsible for the overestimation of AOT by the model, next to the overestimation in aerosol mass. We have added this to the discussion in section 4.2.

5- Reviewer 1: "AERONET L1.5 vs. L2"

In the previous manuscript we compared simulated AOT with data from AERONET level 1.5 (L1.5). In the present version we have changed Figure 2 (old Figure 1) to also include AERONET L2 data. These are better quality-assured compared to L1.5 but the number of data is much smaller, especially in the second, relatively moist half of the month. Figure 2 shows that at several instances cloud-filtered simulation data can be matched with the cloud-filtered AERONET L1.5 data but not with L2.0 data. This occurs on May 17 and, more important here, after May 24 when the atmosphere above Cabauw contains relatively large quantities of Sahara dust. Although high clouds may have been intermittently present in the dust period, it is possible that in the quality assurance procedure AERONET data with a large contribution from dust were erro-

neously associated with cloudy conditions and excluded. We have mentioned this now. Because of the higher consistency with the simulation results we decided to show both L1.5 and L2 data in the manuscript.

— Comments from Reviewer #1 —-

(Mandatory) See general (2) above.

(Highly recommended) We have added a new Figure 1 with the horizontal distribution of AOT over Europe for the dry, moist and dust periods, which illustrate the different episodes during the month. We have decided not to include satellite-retrievals of AOT. Interpretation of a comparison between model output and satellite retrieval, with one observation made each day (e.g., MODIS), is surrounded with much more uncertainty than a comparison with (almost continuous) AERONET observations, which are considered a benchmark for retrieval results. The goal of our manuscript is to analyse the contributions of different meteorological and aerosol physical and chemical characteristics to simulated AOT, and given the smaller amount and larger uncertainties we do not expect additional information from of satellite data for our study.

(Minor, p1, I31-p2, I2) We have added a remark on the uncertainties associated with satellite retrieval of aerosol properties in the Introduction.

(Minor, p3, I9-10) The relevance of this sentence is indeed not clear, and we have removed it from the text.

(Mandatory, page 4) We added more information on the model spatial and temporal resolution.

(Highly recommended, p5, I30). See general (5) above.

(Mandatory, p5, I20-26) See general (4) above.

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(Minor, p7, I24-30) See general (1) above.

(Mandatory, p8, I3) We have included the proper web address.

(Mandatory) We explained acronyms used in the text.

(Minor, p9, I20-21) The dates are indeed not consistent, it should be May 2-14.

(Highly recommended, p9) See general (3) above.

(Mandatory, p10, I7-20) As we mention in the text now, we used all available model data and measurements for calculation of average values. We agree with the reviewer that this procedure is not fully consistent quantitatively but the result is sufficiently clear to obtain a qualitatively consistent picture of the model performance. We have rewritten the paragraph with less emphasis on the quantitative aspects.

(Highly recommended, p11, l1-6) We have added a few sentences and references for more information on the simulation method. A new study is in progress that focuses on the last part of the month.

(Highly recommended, p11, I18-20 & p16, I4-6) The spurious upward mixing from the boundary layer may lead to excess moisture in the free troposphere, as also seen in Figure 7e. However, the overestimation of IWV is attributed mostly to the BL where the specific water vapor concentration is larger.

(Mandatory, p13, I25-28) See general (4) above.

(Highly recommended, Fig 3) This is now Figure 4. Until now we have not been able to obtain hourly PM10 measurements, therefore we have used the daily averaged values from the "Landelijk meetnet" in the Netherlands. For consistency we have adapted the model output to daily averaged values as well.

(Minor Fig 4) Yes all observations were performed at Cabauw, we have added this information to the figure caption.

(Mandatory, Fig 5) The dashed lines represent the standard deviation, this is now noted in the figure caption.

- Comments from Reviewer #2 ---

Title: we have changed the title according to the reviewers suggestion.

P5912 L5: we changed the text to improve clarity and consistency. L6: yes, by nudging the model is made to follow the ECMWF meteorology. Only surface pressure, vorticity and divergence are nudged. All other parameters, including humidity which is of utmost relevance to our study, are calculated explicitly. L23: We have changed the text according to the reviewers suggestion.

P5913: We replaced the clear-sky with the all-sky estimate.

P5915 L8: The altitudes listed are altitudes above the surface. In regions with variable orography the altitudes above sea level may actually vary significantly from gridpoint to gridpoint. Over the Netherlands, however, where the surface altitude is close to the mean sea level the values are representative. L9: These values are from ECMWF analysis, not from a particular dataset. L14: we have changed the text according to the reviewers suggestion.

P5916: we have changed the text according to the reviewers suggestion.

P5917: The value listed as the accuracy of AERONET follows from the combined effects of uncertainties associated with calibration, atmospheric pressure, and total ozone amount (Eck et al., 2005).

P5919 L6: See general (1) above. L22: PM10 is the aerosol mass for particles smaller than 10 μ m diameter. We have explained this in the text. L23: the webaddress is correct now. L25: in this study we focused on AOT and how aerosol and meteorological

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characteristics contribute to AOT in general. In a follow-up paper we focus specifically on the dusty period, and this question will be answered there. On these particular days, the model places about 60% of the total column dust mass at Cabauw below 2.5 km.

P5921: This is the time used by the model, i.e., UTC, which is now mentioned explicitly in the text.

P5923: L4: the variability here was estimated visually. We now present the approximate range, instead, to prevent confusion. L29: No, because the actual sonde measurements were carried out consistently and regardless of clear or cloudy conditions. Note that for the dry period this is not relevant.

P5924 L3: We changed the text accordingly. L15: Cloudsat is another instrument in the A-Train, but should not be mentioned here, so we removed it from the text.

P5925 L23: We changed the text accordingly. L24: We emphasize that during the dry period the air over a large part of NW Europe was characterized by subsidence and therefore cloudless. We infer that the only other possibility of the enhanced RH in the upper BL is advection.

P5926. We added a reference within the manuscript text since the study is not under review yet.

P5931: we corrected this typo.

P5936: As mentioned earlier we have removed the remarks on correlation between the different parameters from the text.

P5937: We added this information in the figure caption.

P5938: For consistency we have adapted the model output to daily averaged values, consistent with the observed PM10. As mentioned in the manuscript, the coarse mode does not significantly influence the aerosol optical thickness apart from the two dust episodes between May 5-9 and after May 26. Sea salt concentrations were rather low

throughout the month, as seen in Fig. 4, so the dust mass can be taken as representative for the coarse mode mass. Theoretically it is possible to calculate the mass fraction in a mode above a certain threshold size. However, the 56 μm threshold is indicative only: the particle capture efficiency shows a decreasing trend around 56 μm . Further, ECHAM5-HAM considers a fixed aerosol modal width which may not be realistic. This means that a comparison of sub-modal mass is implicitly inaccurate. We are still waiting, however, for detailed size-resolved particle concentration measurements, which would enable a closer examination of the representativity of the simulated aerosol size distribution.

P5941: Yes, we clarified that in the caption now. We also changed the computed standard deviation with one computed in log space.

P5945: we corrected this.

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