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Atmospheric Chemistry and Physics Discussions

Interactive comment on "The European aerosol budget in 2006" *by* J. M. J. Aan de Brugh et al.

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We thank A. De Meij, the reviewer, for his comments. We will use his comments to improve the manuscript for publication in ACP. Below, we quote the review comments in italic and add our response in normal text.

One should not draw strong conclusions on the model performance by evaluating annual means. This does not give any information how good the model calculates gas and aerosol concentrations during specific episodes (e.g. dust events in spring, summer) or during cold periods (low PBL heights and the sensitivity of NO3 aerosol formation). I suggest including a comparison of calculated aerosol concentrations with observations for a winter month. This allows the author to compare the implementation of M7 in TM5 with an earlier study by De Meij et al. (2006).

It is true that the model's performance is not adequately evaluated with just annual means. The number of graphs on model evaluation was kept limited because our research focusses on the budget rather than the model evaluation. To improve the model evaluation, we decided to add time series of January 2006 of the four EMEP stations that have hourly data of PM10 for that month (figure will be added in the manuscript). This way, we evaluate the ability of TM5 to simulate events at the right time with the right magnitude. We added the following interpretation in the manuscript:

"We clearly calculate less noise than observed. TM5, as a global model, is unable to simulate local effects of short timescales. This is mostly visible in Narbeth (GB) where TM5 cannot follow the rapid changes in PM10 that are observed. Vredepeel (NL) and Vavihill (SE) are represented quite well. The timing of the peaks is roughly correct. Only the magnitudes of the two peaks are significantly underestimated, namely the Vredepeel (NL) peak around day 27 and the Vavihill (SE) peak around day 15. TM5 is clearly unable to simulate aerosol concentrations on Ayia Marina (CY). Here, resolution plays a big role, because Cyprus is an island as small as a grid box. The big peaks in the model results of Ayia Marina (CY) are dust stroms, that are not observed at the station. Only the broad peak at the beginning is, though much smaller, visible in the observations."

The manuscript shows the budgets of the inorganic components, natural dust, BC and POM. However, the model performance regarding BC and POM is not evaluated. I believe that adding the evaluations of calculated POM and BC concentrations would improve the quality of the model evaluation.

The reason that the BC and POM evaluation was not included was the very low amount of data of BC and POM at the EMEP stations in 2006. We will add a comparison between our yearly averaged model results of 2006 with yearly averaged observations of the 2002-2003 EC/OC-campaign. The results indicate that black carbon is represented well and that organic matter is severely underestimated. Both for BC and POM, there

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is a quite good correlation between model results and observations.

Specific comment: Page 21393, Line 5, add reference Kaufman (2002): Kaufman, Y.,J., Tanré, D. and Boucher, O., 12 September 2002. A satellite view of aerosols in the climate system, Nature, Vol. 419.

This reference contains useful information about the effect of aerosols on the climate, so it is a useful addition to the introduction. We will add the reference at the location proposed by the reviewer.

Page 21394, Line 14, also water uptake on aerosols, chemical formation of aerosols, model resolution dependency on aerosol calculations, horizontal and vertical distribution of the emissions and meteorology.

We cited Textor et al. (2006) at this location to highlight the poor agreement among models concerning aerosol processes, which we use to conclude that a budget analysis is important. We agree that there is much more interesting information in Textor et al. (2006), but we do not think it is necessary to mention it in this paragraph.

Page 21396, Line 9, wet and dry deposition. Later you deal with wet deposition and the related uncertainties in the calculated AODs. How is the cloud formation done in TM5? Perhaps you can write a few ilnes about this.

Cloud formation is not done in TM5. The information about the clouds is taken from the ECMWF meteorological data. We will clarify this by mentioning the data fields in section 2.1 where we introduce the ECMWF data.

Page 21396, Line 13. Please explain the abbreviation SVN.

SVN stands for subversion. That is the version control system used for TM5. The

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reason to mention it is only for reproducibility. The exact model code used in this study can be re-obtained with this number.

Page 21397, Line 1, 5 and 11. Median radius, instead of median size. In the same section: you should say that the aerosols in M7 are internally mixed for the water soluble components, which is later important for interpreting the calculated AODs. I assume that you optical properties are based on externally mixed aerosols?

We corrected the 'median size' to 'median radius'. In M7, the aerosols are considered internally mixed. For the AOD, we calculate the refractive index of the aerosols (which are mixtures of components). We do not use a volume-weighted mean, but we use the formulas designed for this as in the references cited (Maxwell-Garnett (1904) and Bruggeman (1935)).

Page 21397, section 2.2.2. Ammonium and nitrate, what are the uncertainties in EQSAM at high relative humidities (>95%) Do you use a cut-off value for the RH? Which version of EQSAM are you using, Version 2?

We are using EQSAM version 3. We will add this version number in the manuscript. EQSAM 3 uses a cut-off RH of 99%. We do expect that the error of this cut-off will be small compared to other errors.

Page 21400, a description of NH3 emissions is missing.

The description of ammonia emissions was forgotten. We will add it in the revised manuscript. The ammonia emissions are postprocessed data that originate from the inventories of Bouwman et al. (1997).

Page 21401, 2.2.6. Maybe you can add the real and imaginary refractive indices in a table. What is the reason to compare the AODs at 440nm? Common practice is

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550nm (many optical instruments deliver products around this wavelength). No need to change to 550nm of course.

While observing the AERONET website, we noticed that 440 nm is a wavelength of which many stations have data in 2006. There is no scientific reason to take this wavelength. As we also evaluate the Angstrom parameter, the AODs at other wavelenghts could be estimated. We do not think it is valuable to add a table of refractive indices. The refractive indices are based on references, that are cited in the manuscript. Analysing the uncertainties of the refractive indices is beyond the scope of this manuscript.

Page 21402. In Table 5 you describe the method how you compare modelled AOD with observations. I think that you should move that description to section 2.3. Secondly, say a few words on how the sun photometers of AERONET operate.

This is a good point. The data sampling issue in the caption of table 5 should be moved to the methods section. We also added some text about the sun photometers from the AERONET website.

Page 21403, section 3.1 Line 13 SOx (SO2, H2SO4), do the same for NOy (NO2, NO, PAN, NO3, HNO4, N2O5).

We already have this explanation in the captions. We will add the same explanation in the text. The order of this section may change a bit. We will make sure the explanation is at the first occurence 'SOx' and 'NOy' in the text.

Page 21404, Line 1, The calculated annual mean AOD

That is true. We will correct this.

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Page 21406, Line 24 you mention that the numbers in the Tables 3 and 4 are raw model results. Could you please explain the difference of 45 Gg/year for dust sinks and sources in Table 3? All the other numbers are within 2-3 Gg/yr difference.

Actually, the sources and sinks do not close by definition. The difference between them is the accumulation or depletion in the atmosphere during the modelled year 2006. This accumulation or depletion is less than one percent of the total budget. For all tracers except for dust, this value is in the order of a gigagram, which is comparable to rounding errors. But for dust, this value is higher.

Page 21409, Line 28. Part of the underestimation...the model. The study of De Meij et al. (2006) shows that calculated AODs at low RH ranges are underestimated, indicating that the AEROCOM emissions are too low. Another reason for the underestimation of the AODs is that vertical distribution is not well represented, as mentioned earlier.

This is an interesting addition with an another reference to De Meij et al. (2006). The paper was already cited in the manuscript in a different section.

Page 21411, section 2.5: for how long remains BC and POM insoluble before it can be washed out.

The aging (becoming soluble) of BC and POM is calculated online in M7. When enough soluble matter is condensed, an aerosol becomes soluble. M7 keeps the budget of all processes (including aging), so we could look up the aging time from the result data. It appears that both BC and POM age within one day in the boundary layer and in two days in the free atmosphere. The results are about the same for BC and POM, which is logical, because they are on the same aerosol mode of M7.

Page 21412, section 3.5.2; you mention that in spring 2006 smoke was transported from Russia because of easterly winds. Analysing ECMWF monthly mean winds over

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the western part of Russia (at 850 hPa, 700hPh and 500hPa) for March-June 2005 and 2006, I find a dominant westerly monthly mean wind direction. However, for some days in e.g. 14-15 June 2006 the dominant wind direction is easterly. Analysing back trajectories for the period you mention in the figure caption, I find that for the Lille station (70km from Dunkerque) the air mass originates only from the east between 9th May – 14 May. Therefore I think you should specify the dates when the easterly winds are occuring as you did in the figure caption of Fig 6.

The easterly winds are not evaluated first. We marked a period with red triangles to catch the seperate population of points in Minsk. First of all, we tried to improve the result by implementing GFED 3 monthly mean emission data for the specific year 2006. This did not work out, since the emissions in GFED 3 were spread out over the entireties of April and May, while the real emissions were not. We analysed the wind with TM5 itself, since it writes out the wind data at the EMEP stations. When averaging all (54) EMEP stations that are located between 20 and 30 degrees east, we obtain easterly winds from April 22nd until May 8th (roughly the last week of April and the first week of May). The rest of April and May, the winds were westerly, resulting in westerly monthly mean winds for both April and May as mentioned by the reviewer. We will add a notification about this in the revised manuscript.

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 21391, 2010.