

Interactive comment on “The sensitivity of aerosol in Europe to two different emission inventories and temporal distribution of emissions” by A. de Meij et al.

A. de Meij et al.

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Detailed Responses to Reviewer 1.

General comments

1. Your model currently does not include information on the particle number concentration and size-distribution of the accumulation mode (SO₄, NH₄, NO₃, H₂O, BC, POM, MSA). To calculate AOD, you assume a size-distribution from Whitby for sulfate particles, although 60% of the mass emitted (EMEP) consists of BC and POM. Did you check the sensitivity of your results to the chosen size-distribution? This would be an interesting information when comparing absolute AOD values of the model to observations since AOD depends crucially on particle number concentration and aerosol

size-distribution. Please add some words on this issue.

The reviewer correctly remarks that there might be a sensitivity of our calculations to the assumed 'Whitby' distribution, which indeed was derived for a accumulation mode sulphate dominated aerosol ($r=0.034$ $\sigma=2.0$). While we think that the choice of this specific distribution is still reasonable we have now also evaluated the effects of assuming the 'water-soluble aerosol accumulation/Aitken mode' as presented in Table 4.2 in the d'Almeida climatology ($r=0.0285$ and $\sigma=2.239$). Putaud et al (2003) present a host of log-normal fits to observed size distributions at various locations in Europe. E.g. at the rural location Ispra Mode 2 parameters $r=0.024$ and $\sigma=1.91$. Using these parameters we calculate that the extinction coefficient would differ from the assumed Whitby distribution by 3% (higher) and 15% (lower), respectively.

This discussion has been included in revised paper p. 3294.

2. Most processes relevant for aerosols are size-dependent such as wet and dry deposition. However, for the most relevant aerosol components in Europe (SO_4 , NH_4 , NO_3 , H_2O , BC, POM) your model takes into account aerosol mass only. In contrast, mineral dust and sea salt particles are simulated with more detail including different aerosol modes and particle number concentration. Shouldn't this be the other way round? Please add some words on why you think this doesn't matter when investigating model results for Europe.

We do not claim that size-resolved aerosol dynamical processes do not matter when trying to understand aerosol concentrations and optics in Europe- however until now surprisingly little research has been performed evaluating the 'gains' of including aerosol microphysical models (such as the JRC M7 model) compared to bulk approaches. A systematic evaluation of this planned for future publications. However, we do think that for dust and seasalt it is unavoidable to include the highly size-dependent sedimentation and wet scavenging processes, leading to a significant change of the size distribution from the sources regions to other locations.

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3. BC and POM are known to be (at least partly) hydrophobic upon emission. Please give some information on how you treat the wet deposition of BC and POM. If the hydrophobic properties of these particles are ignored, wet deposition is expected to be overestimated.

In this particular set of model simulations we have assumed 100 % hydrophilic properties for BC/POM, and we hence assumed that BC/POM was removed by wet and dry depositional processes like soluble inorganic aerosol (SO₄). Analysis from AEROCOM indicates that indeed TM5 has relatively short residence times compared to other global models. We stress however that other assumptions on wet-removal (e.g. the fraction interstitial aerosol) may equally influence these model outcomes.

P4. 3269: The TM5 model is driven by meteorological data from ECMWF. Does this include prescribed data for clouds, precipitation, convection, etc.? Since clouds and precipitation are essential for the simulation of aerosols (in-cloud SO₄ production, wet deposition) this would be important to know.

As mentioned in section 2.1 TM5 utilized in this work information from the 6-hours IPS forecast on 3d cloud cover and cloud liquid water content, convective and stratiform rainfall rates at the surface, and surface heat fluxes to calculate convection.

5. p. 3276, Sect. 3: Please give some more information on the expected accuracy of the EMEP measurements. What does the NH₄NO₃ evaporation (T > 20_C) mean for the comparison of NH₄ in June? Temperatures exceeding 25 °C could lead to complete evaporation of the aerosol nitrate from the quartz filter. Between 20 and 25 °C the loss due to evaporation could be 50% (Schaap et al. 2003). Concluding from Schaap et al. 2003 is that quartz filters are suitable for aerosol nitrate measurements only when the temperature does not exceed 20 °C during sampling.

6. p. 3278, l.1/2: "...we selected those measurement stations able to represent the model spatial scale..." You excluded EMEP stations with a temporal correlation between model and measurement less than 0.5. But how did you determine which mea-

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surement station is representative for the large spatial scale of the model grid boxes. Please give some more information on this issue, which is essential when comparing the coarse model results to individual measurement sites, in particular to only few measurement sites.

First we should remark that our model is of relatively fine resolution compared to many previous global model studies, and that in general the topic of representativeness in those studies is avoided. We have spent considerable time to think about what could be a decisive parameter deciding whether a coarse resolution model can represent a point measurement. To our opinion the key lies in the ability of the model to represent synoptic scale variability; the correlation coefficient of daily observations and model calculations is taken into account. The final choice of a threshold of $r=0.5$ is arbitrary, and mainly a compromise of retaining a sufficient amount of data for comparison.

7. p. 3304/3308, Tab.2/6: The standard deviation of the AODs is very large (almost as large as the mean value), in case of S_EMEP (Tab. 2, June) even larger than the mean value. Thus, it is probably worth looking at median and e.g. 10%- and 90%-percentiles, too.

We present a detailed analysis of this at question 1 of General comments of reviewer 2.

Specific comments

8. p. 3267, l. 22: "Two major uncertainties of the current regional and global scale emission inventories..." Which major uncertainties? Please be more specific.

a) the accurate estimation of the quantity of the aerosols and precursor emissions b) the role of the temporal distribution of the emissions in the inventories.

9. p. 3268, l. 13: The AeroCom emission inventory for the year 2000 should be referenced (Dentener et al., 2006). Done.

10. p. 3270, l. 2: Please explain the abbreviation "EBI". Eulerian Backward Iterative.

11. p. 3270, l. 18: Aerosol components of nitric acid should be NO₃ (not HNO₃), of sulfuric acid SO₄ (not H₂SO₄). Corrected.

12. p. 3270, l. 21: EQSAM v03d is used to calculate gas-/aerosol-partitioning and the aerosol liquid water content of the SO₄-NH₄-NO₃-H₂O system. Why do you not apply EQSAM to calculate the water uptake of sea salt particles? Indeed EQSAMv03d contains a parameterization for the uptake of water by sea-salt. The older current water uptake parameterization by Gerber was implemented along with an older EQSAM version; and unfortunately the two parameterizations have not been made internally consistent. We note however, that we do not have indications that there would be problems with the Gerber parameterization so that we think that the seasalt results can be used for this work. Note also that we assume externally mixed aerosols, which means that water uptake of sea salt is independent of the water uptake of inorganic aerosol.

13. p. 3271, l. 3: What do you mean by "...in relation to the model grid size."? We mean that there is a grid-resolution dependency which is accounted for: i.e. in general the effective removal on finer grids is faster than on coarser grid.

Rephrased: "Removal by stratiform clouds considers precipitation formation and evaporation, and cloud cover, and takes into account a grid-dependency. Effectively rain-out on smaller grids works more effectively than on larger grids."

14. p. 3270: I guess all aerosol components in the accumulation mode (SO₄, NH₄, NO₃, H₂O, BC, POM, MSA) are internally mixed. If so, please say so. We consider all three accumulation mode aerosol classes (inorganic (NO₃, SO₄, NH₄), BC, and POM) externally mixed. We made this in the text more clear.

15. p. 3273, l. 14/20: Reference year of Dentener et al. should be 2006 instead of 2005. Corrected

16. p. 3273, l. 17: Please specify whether SO₂ emissions are given in Tg(S) or in Tg(SO₂). p. 3289, l. 3289: "NH₃ and NO_x emissions by..." Do you mean "concentra-

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tions" instead of "emissions"? Concentrations. This is corrected in the text.

17. p. 3292, l. 16: Please add some information on how you convert between POM and OC (in the model data). We used a constant factor of 1.4 in the conversion from POM to OC. While this factor is fairly uncertain, the value for this factor was chosen for consistency with the assumptions made in the AEROCOM database.

18. p. 3298, l. 14: Replace "2006b" with "2006". Done.

19. p. 3318, Fig. 5: "Brown presents AOD by dust, green AOD by inorganic aerosol and the associated aerosol water." This statement does not match the color legend of the individual plots (a-e). Corrected in the figure caption.

Detailed Responses to Reviewer 2.

Introductory remarks 1a Concerns - EMEP emission and EMEP station data seem linked

The EMEP synthesizing centre West, responsible for the EMEP emission inventories, and EMEP measurement network are really two independent bodies, with different people involved. Our subjective judgement is that the emission inventory is made without "tuning" too much to match EMEP model data with the measurements. E.g. EMEP-W currently has an underestimate of modeled BC by a factor two, nevertheless the bottom-up inventory was not adapted.

1b simulated aots on June 11 of 1.4 or 1.6 over Europe seem awfully high (why are simulated (background) aots so low if emissions are relatively high (model bias?))

The AOTs of 1.4-1.6 are due to high RH situations (uptake of water on the aerosol); we explain in the text that the cloud-screening algorithms of MODIS and AERONET attributed this situation to clouds. We want to demonstrate however, that in many cases in the vicinity of frontal systems, there might be 'hazy' conditions, which are difficult to categorize as cloud or aerosol. See also point 2.

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General comments

2 s1048 Also the apparently good comparison between AERONET monthly statistics seems to be helped by the few high humidity / high aot events. Otherwise the simulated AOD would be low. Maybe for such a comparison it would be more honest to do a comparison by rel.hum ranges given the sensitivity of the model to ambient relative humidity (- at least pick a lower threshold than 90%). Some sentences regarding potential biases of the TM5 simulated aots might be useful, based on comparisons to simulations of other models with the same AeroCom emissions.

On request of the reviewer (see also reviewer 1, General comments point 7) we made this analysis based on the 5 station data shown in Figure 1a-e. This analysis reveals that AOD at lower RHs is underestimated due to too low inorganic aerosol concentrations. However at higher RHs (70-80% and 80-90%) we see that high AOD values are calculated and large standard deviations are calculated for the model AOD due to the non-linear effect of RH on aerosol water concentrations, which impact the total model AOD.

We included this in the discussions part (section 5) as well as the figures which are presented in this document, Figure1.

3. However, it remains unclear, if internal mixing, which is not considered by the model, would necessitate an even higher temporal resolution.

We did not understand this statement of the reviewer with regard to temporal resolution.

4. This possibly suggests that overall processing of emissions in models are more important than emission themselves indicating that each model has a mind of its own largely independent of emission input (a result which was also observed when harmonizing aerosol emissions in AeroCom Exp. B (see Textor et al. 2005). This is an overall nice contribution and appropriate for ACP.

We partly agree with this statement to the extend that when we compare the AERO-

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COM and EMEP inventory for Europe, a perhaps larger uncertainty is due to the uncertain aerosol production and removal processes. This is indeed inline with Textor et al., 2005. We include a brief discussion in section 5.

Minor comments P3272 BC mass fraction seems high at 25%

European wide PM_{2.5} as well as BC emissions are highly uncertain. Unfortunately in the Eurodelta/EMEP emission inventory used for this study, only PM_{2.5} emissions were provided, so that we had to make assumptions on the fraction BC in PM_{2.5}. We followed a methodology similar to Schaap et al (JGR, 2003); who estimated that, taken all sectors together, BC constituted 25% of all emissions PM_{2.5} emissions (his Table 2). Given the dominating importance of road-transport and small combustion sources we correlated these numbers with the measurements of Putaud (2003), who give for the average of 'kerbside' measurements a fraction of 17 % EC in PM_{2.5}. The expert judgement of 25 %, corrects for the fact that there must be some secondary aerosol in these kerbside measurements.

P3277 MODIS data have a positive bias over land (see Remer et al, 2005) AOD by MODIS is recalculated with the bias given by Remer (2005), and the reference to this paper is made.

P3278 EMEP emission data validating with EMEP station data ? (are they not dependent?) See answer 1a.

P3279 Any recommendation (for the AeroCom next generation data-set) for SO₂ emission heights?

There are marked differences in the recommended emission heights for SO₂ between AEROCOM and EMEP inventories. Both recommendations are not backed-up by detailed data-base work. Roughly the differences in emission heights can be summarized as:

0-100m 100-300m 300-1000m 0-100m 100-300m 300-1000m AERO AERO AERO

EMEP EMEP EMEP Power plants 0% 100% 0% 0% 8% 92% Res. Combustion 100%
0% 0% 50% 50% 0%

Based on our model results we tentatively judge that the EMEP emission heights give more realistic SO₂ surface concentrations- although in those simulations also the emissions their self were varied. We recommend a detailed analysis of stack height + effective plume rise from various sectors for new recommendations.

P3283 Is the difference in POM between EMEP and AEROCOM related to SOA (if so - say so) Yes, this is mentioned in section 2.3.3.

P3284 MODIS Angstrom parameter are biased high over land (qualitative use recommended) We are aware that Angstrom by MODIS is biased high over land, therefore the comparison made with the Angstrom coefficients and the type of aerosol in calculated by the model (section 4.2) is done on a qualitative base.

P3285 I have a hard time accepting these large (simulated) aots -also given the MODIS biases I prefer a discussion on patterns rather the numbers (where there any AERONET data to substantiate large aots?)

P3288 monthly average comparison seems often fortunate (generally low simulations seem balanced by high rel.hum. / aot event). What if we only compare aot at rel.hum <70%? See answer 2 of reviewer 2.

P3293 N?? -species are NOT part of the AeroCom but of the (your) 'extended AeroCom' dataset (say so) We used the IASA 2000 emissions, as described in Dentener et al. [2004]

3276/22 calculate 3276/25 are 3288/12 presence of inorganic

Typos have been corrected.

(a) (b)

(c) (d)

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(e) Fig. 1. to the General comments point 7 of reviewer 1 and point 2 of reviewer 2. AOD calculated by the model (blue) and observed by AERONET (red) at different relative humidity ranges (40-50%, 50-60%, 60-70%, 70-80%, 80-90%), for El Arenosillo (a), IMC Oristano (b), Ispra (c), Moldova (d) and Avignon (e) for June 2000. The black line presents the standard deviation.

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