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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 the anonymous second reviewer for his or her comments. These comments are of great value for our manuscript for publication in ACP. Below, we quote the review comments in italic and add our response in normal text.

In several places in the text, the references used are not the proper ones. Especially when speaking about aerosol microphysics in global models, the only ones referenced are M7-related works, and the GISS-MATRIX model (Bauer et al., 2008). Nevertheless, more models include aerosol microphysics, e.g. GISS-TOMAS, GLOMAP, CCCms and should be also referenced where appropriate.

We added two more references in the introduction: Lee and Adams (2010): Evaluation of aerosol distributions in the GISS-TOMAS global aerosol microphysics model with remote sensing observations (ACP), and Korhonen et al. (2008): A global model study of processes controlling aerosol size distributions in the Arctic spring and summer (JGR). We also added one reference in the methods section where log-normal distributions are introduced (2.2.1): Salzen (2006): Piecewise log-normal approximation of size distributions for aerosol modelling (ACP).

"TM5 uses operator splitting which means that each process (e.g. advection, chemistry) has a time step of 45 min": This is an incorrect statement. Operator splitting is the calculation of different processes independently, contrary to what happens in the real atmosphere; it has nothing to do with the timestep duration, at least not directly.

The reviewer is right. There is no direct causal relationship between operator splitting and the time step length. TM5 uses symmetric operator splitting. Within one time step, all processes are executed in one order and then repeated in reverse order, to cancel out most of the numerical errors. In this method, each process is done twice in a base time step and therefore has half the time step. This reasoning is a technical detail, which we will not include in our manuscript, but we will remove the causal term 'which means', because it is incorrect.

The parameterisation of dry and wet removal with respect to aerosol size is not very well explained. For example, in section 2.2.3, the authors mention that both the deposition and sedimentation velocities are aerosol-size dependent. They also mention the same for stratiform wet deposition. This would imply that during a single deposition step, not all aerosol sizes will be removed with the same efficiency, leading to a skewed distribution. Redistributing the aerosol to the same size distribution width and only changing the mode size will artificially unify the distribution, altering the removal's size-dependency applied at the bins. Assuming that this occurred on purpose for simplicity, which is fully acceptable, one would expect a discussion on how such an assumption affects results, especially because later in the manuscript wet removal is *discussed in such great detail.*

We discussed in the manuscript that fixing the standard deviation is necessary to have the destribution fully determined by the tracers: aerosol number and masses. Therefore, it simplifies a lot. As the reviewer expected, the simplification is done on purpose. We added a little discussion about the errors this simplification can introduce: The redistribution of aerosols moves aerosol mass from the middle sizes (with long lifetime) to the small and large sizes (with shorter lifetime). That can accelerate deposition. Quantifying the error is difficult, because it is computationally very expensive to do it correctly. And it is not known in which extent the parameterisation is tuned to compensate for these effects. Although it would be interesting to investigate this issue, it is beyond the scope of this manuscript.

In section 2.2.2 it is mentioned that SO4 from all modes is used to calculate NH4 and NO3, which are then being kept in the accumulation mode only. This is unrealistic, since Aitken and coarse SO4 should not contribute at all to the accumulation mode NH4 and NO3.

The reason is that we have EQSAM for ammonium nitrate, and EQSAM does not have size bins. Therefore, we convert sulphate to the bulk approach, with which EQSAM works. What we calculate with EQSAM is bulk ammonium nitrate. The rest of the model does not employ the bulk approach, so the ammonium nitrate must be distributed into our modes. We simplify by putting all ammonium nitrate in the accumulation mode, becuase the majority of the aerosol mass is there. Hereby, we incorrectly put ammonium nitrate from the Aitken and coarse modes to the accumulation mode. First, we think it is more incorrect to leave it as gas instead. Second, the mode of the ammonium nitrate is less important, because it could move to another mode whenever it wants by quick evaporation and recondensation. Third, a later ISORROPIA-GMXe simulation confirms that a vast marjoriy of the ammonium nitrate collects in the accumulation mode. In the manuscript, we improve the discussion why the ammonium nitrate is put

into the accumulation mode and we added a line about the ISORROPIA-GMXe run, with a reference to Nenes et al. (1998), about ISORROPIA and Pringle et al. (2010) about GMXe.

Section 2.2.4, Last line. How much wet removal is being slowed down? Which criteriorn is being used and how resolution plays a role. More quantitative discussion should be included here.

Wet deposition is a challenge in any global model, since the mixing time of the grid boxes can be slower than the timescale of wet deposition. The manuscript explains that this problem is treated 'pragmetically' so that the deposition is reduced in larger grid cells. The reviewer correctly remarks that just using the word 'pragmatically' is too pragmatic to explain what happens. It is a quite technical issue, which is explained in the appendix of Vignati et al (2010b). We will add a citation to this paper here and explain briefly what happens: A 'no-mix' parameter is introduced, that represents the time in which the in-cloud, below-cloud and cloud-free fractions of grid cells are treated quasi-independently.

"The aerosol mass emissions have an assumed lognormal distribution" means that the inventories have an assumed distribution which is being modified to fit that of the model's, or they are introduced in the model with the model's assumed distribution?

The emission inventories prescribe the emitted mass and median radius of the emitted aerosols. From this, the number emissions are calculated using a fixed standard deviation. Thus, the assumed lognormal distribution is not used for anything but calculating the emitted aerosol number.

Measurement section: Dust, OC and BC are not even mentioned. Since they are discussed later on, and more importantly the title of the paper mentions aerosols in *general, some discussion about their measurements should be included here.*

The discussion about BC and POM measurements was not included, because hardly any measurements of BC and POM were present for 2006. We will add a comparison to the measurements from the EMEP EC-OC campaign of 2002 and 2003. Though this is a different year, it makes sense to compare the annual means. We will also add references of earlier model studies that are compared to the EC-OC campaign. Dust is very hard to measure as a fraction of the aerosol mass. There are observations of trace metals (e.g. potassium, magnesium), but it is very uncertain to derive the total dust mass from these tracer metals. We will add a short discussion about dust in the measurement section.

Figure 1 should be re-ordered, in the same way its discussion flows. In its discussion, the authors claim that carbonaceous aerosols are similar with SOx and NOy, but all three are very different between them, I hardly see any resemblence in the fields presented.

In our revised manuscript, we want to fix the order of tracers as much as possible in a continuous order flowing from gas to aerosols (first precursors, then gases in equilibrium, then ions, then bulk) and within one group from reduced to more and more oxidised. Then, figure 1 and the legend of figure 4 must be read in columns. Tables 3 and 4 also have that order. We are reconstructing the model evaluation part and will also use this order. To have the discussion parallel to the figures, we will re-order the discussion. The resemblence between the distributions of carbonaceous compounds, SOx and NOy is that they all exhibit a hot-spot structure, which is typical for primary components with a limited lifetime, though the hot spots are at different places. We will clarify this in our revised manuscript. However, the difference in the hot-spot locations for NOy and BC is not so different.

The model evaluation is poor. The locations of stations should be shown, ideally per aerosol component, if not a plot that shows in a graphic way both the model's results and the measurements. This is particularly important, since later on the spatial agreement of the aerosol measurement patterns with the modelled results is mentioned (also in the optics section), but not shown. In addition, the comparison of model results with measurements for NH3 and HNO3 should be shown, even if it is not very good. The authors claim that parts of the model's overestimation of NH3 (contrary to other models that underestimate NH3) is the fact that TM5 does not have a diurnal variability of NH3 emissions. To my knowledge, no model has. Further, the mention that total oxidised nitrate is ok, same for nitrate, but not HNO3. Since the total oxidised nitrate is the sum of the other two, how can this be possible? Positive and negative errors cancelling each other? For SO2, a discussion about possible errors, but no mention is made for its removal. If the removal of SO2 is underestimated in the model, then everything should move to the correct direction. Same as in the measurement section, no discussion is made about the model's performance concerning dust, OC and BC. Last for the model evaluation part, what is the "unaccounted mass" beyond water.

The number of graphs on model evaluation is kept limited to keep the paper concise. We will add a comparison for BC and POM with the EC-OC-campaign observations. We will also add topographic maps with model results and observations. To keep the number of points limited, we will use land-averages. For example, we have 84 stations for total PM. Showing them all individually would result in an unreadable image. To represent the number of stations, we will use larger signs for more stations in a country. We will do the same for the AERONET stations, but as we only have eighteen stations, we do not use the land-averages, but rather put the points on the correct topographic locations.

In the revised manuscript, we decided to leave out the graphs of NH3 and HNO3, because they are considered less important than the ones shown. The reason that the model has difficulties with these compounds are explained in the paper. Furthermore, the measurements of total ammonium and total nitrate are more reliable than those of only gas or only aerosol (explained in methods section). This also explains why total nitrate and aerosol are represented well, and nitric acid (the difference) is not represented well. Many EMEP stations report two of the three quantities (total NO3, aerosol NO3 or HNO3) and not the third, though it could be deduced by the sum-relationship. However, using this sum-relationship is unreliable as different measurements methods are used for the two values. We trust the EMEP stations in assessing their data's reliability. Moreover, the nitric acid concentrations can be very small compared to aerosol nitrate, so that the relative uncertainties grow very high.

Concerning our overestimation of ammonia, we addressed the difficulties with modelling the stable boundary layer. TM5 has unclear stable boundary layers, because it hardly can calculate a gradient at such a small height scale (50 m). This unclear boundary layer amplifies the error due to the lack of diurnal cycle in the modelled emissions (which is a problem especially for ammonia as explained in the manuscript).

Concerning the sulphur dioxide removal, we did address the removal process of SO2 "... underestimated loss of sulphur dioxide that does not lead to sulphate production, e.g. dry deposition (Chin et al., 2000)", page 21405, lines 18-20. That is what would 'move everything in the right direction'.

About OC and BC: We are going to add a comparison with the measurements from the EC-OC campaign.

Unaccounted mass is frequently mentioned in measurements if the total aerosol mass exceeds the sum of all accounted masses. It can also contain dust.

Page 21406 Line 20: I do not understand how I can see such a thing in Fig 4.

Figure 4 shows the horizontal fluxes of the boundary layer and the free atmosphere. We want to show that the absolute fluxes are higher in the free atmosphere, which is clearly visible in figure 4 by the longer bars. However, this is not true for the net fluxes (tables 3 and 4). We discussed on page 21407, lines 1-5 that the fluxes in the free

atmosphere are acually quite similar to those of the boundary layer, but the jet stream import and export is added in the free atmospheric fluxes.

Aerosol budget. Very comprehensive in general, with a lot of useful information, gives value to the paper. Few questions/comments: How much of the emissions are injected directly in the free troposphere? For sea-salt, the large oceanic contribution to the European domain drives the conclusion that Europe is a source of sea-salt to the free troposphere. This is an awkward conclusion at a first glance and should be phrased carefully. My guess is that if one will take the continental part of the domain only, then they might see sea-salt descending from the free troposphere to the boundary layer, same as dust does.

The emissions that are injected directly into the free atmosphere are shown in table 4. An emission (E) flux in the free atmosphere is an emission that is directly injected in the free atmosphere. The reviewer is correct with the remark about sea salt. The positive vertical flux of sea salt is probably only due to the emissions within the European domain, so it is not so special as it is described in the manuscript. We will add more nuance in this part.

Optical analysis. The discussion of seasonality should be supported by figures.

This is again a point where it is hard to be both complete and concise. We will add a figure with average observed and modelled AOD per month.

Page 21410, Line 15: Kahnert (2010) mentions a factor of 2 difference for the radiative forcing of aerosols, not the optical depth.

Intuitively, one would suggest that the radiative forcing of aerosols is very tightly related with the optical depth, since both are caused by the scattering and absorption of light. We discussed this point with Kahnert and it appears that there a highly nonlinear relationship between the aerosols' radiative forcing and their contribution to the optical depth. The text will be corrected and clarfied with an additional reference added.

Appendix: I do not understand why this appendix is present. The authors claim that they've reduced the number of parameters in order to reduce their lookup table by using dimensional analysis, and provide two equations that are currently being used for aerosol extinction, but can be applied on single scattering albedo and asymmetry faqctor as well. To me it looks like they re-invented the size parameter, widely used in aerosol optics calculations for several decades. In case I've misunderstood something here, this means that a better explanation.

The reviewer is right that this appendix does not contain any new science, so it will be left out.

Some improvements in English usage is required. These include, but are not limited to: "we observe" should be used for measurements, in case of modelling one should use "we calculate", "free atmosphere" is not defined (25 layers are mentioned in section 2.1, but not up to where), my guess is that "free troposphere" was meant to be used, with the stratosphere excluded; "marginal" is frequently used, where "limited", "negligible", "small", etc should; "Descending flux" should be either "negative flux" or descending air mass".

We will change the terms "we observe" for model results. We use "free atmosphere", because the model domains includes the majority of the stratosphere. The part that is not modelled (e.g. upper stratosphere, mesosphere) are chemically quite unimportant. Only for nitric acid and ozone, a boundary condition at the top of the model domain is applied. Using "free troposphere" would be wrong, because it does not stop at the tropopause. We will clarify this in the methods section where we introduce the vertical layers of TM5. We agree that we used the word "marginal" way too often. We will change that to the more proper words like "negligible" or "small". We also agree that

'descending flux' is wrong. We will use 'negative vertical flux' instead.

The last two paragraphs of section 1 say more or less the same things, the authors might want to consider merging them into one.

The last paragraph in the introduction was the obligatory 'here we do this, and here we do that'-paragraph, while we actually already introduced the structure of the paper in the paragraph before. We will 'merge' these two paragraphs by removing the last paragraph.

Page 21399, Line 20. IPCC is not a model intercomparison study.

This is a misformulation. We know that IPCC is an organisation, not a study.

In the aerosol optics discussion, r is used for wet radius, while the same symbol was used for dry radius in Eq. 1 and 2. In addition, the prefactor beta has to be explained. What is it?

We will use subscript 'w' for wet radius. The prefactor beta is quite unimportant and is not meant to raise quations. By calculating the Angstrom parameter, a power-law relationship between wavelength and AOD is assumed. A power-law has a power (minus the Angstrom parameter) and a prefactor (beta). The unit of beta is a disaster (nm^{alpha}), so quantitative interpretation is next to impossible. What one can consider is that when beta is doubled and alpha stays the same, the AOD is doubled at every wavelength, which is the case if all aerosol concentrations are doubled (with the same size distribution). Beta is a measure for the overall optical depth and alpha determines the wavelength-dependence. But without knowing alpha, beta cannot be interpreted, because the unit would be unknown.

Page 214706, Line 18. 15% of the air mass or volume.

We mean 15% of the air mass. We defined the boundary layer from 1000 hPa to 850 hPa (=150 hPa) and the free atmosphere from 850 hPa to the top of the atmosphere (=850 hPa).

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