Atmos. Chem. Phys. Discuss., 10, C9930–C9934, 2010
www.atmos-chem-phys-discuss.net/10/C9930/2010/

Atmospheric Chemistry and Physics Discussions

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

Anonymous Referee #2

Received and published: 18 November 2010

The manuscript presents a modeling study with the two-way nested global model TM5 over Europe, studying the total aerosol distribution during 2006. The authors identify wet removal and emissions as two major sources of the discrepancies between modeled and measured aerosols at several locations throughout Europe. The manuscript is well written, with only minor improvements in English required (for a few examples, see technical corrections). The study presents a very comprehensive budget analysis, which the authors claim should be the standard way in presenting modeling results. Although the budget analysis is well presented, the rest of the paper requires some improvements in both presenting the modeling work and the presentation of results prior publication.

General comments

1) In several places in the text, the references used are not the proper ones. Especially

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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, CCCma, and should be also referenced where appropriate.

2) "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.

3) The parameterization 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 aerosols 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.

4) 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.

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

6) "The aerosol mass emissions have an assumed lognormal distribution" means that

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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?

7) Measurements 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.

8) 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 resemblance in the fields presented.

9) 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 modeled 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, they mention that total oxidized nitrate is ok, same for nitrate, but not HNO3. Since the total oxidized nitrate is the sum of the other two, how can this be possible? Positive and negative errors canceling 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 measurements 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?

10) Page 21406, line 20: I do not understand how I can see such a thing in Fig. 4.

11) Aerosol budget: Very comprehensive in general, with a lot of useful information,

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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.

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

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

14) 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 factor 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 is needed.

Technical corrections

1) Some improvement in English usage is required. These include, but are not limited to: "we observe" should be used for measurements, in case of modeling 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".

2) The last two paragraphs of section 1 say more or less the same things, the authors

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might want to consider merging them into one.

3) Page 21399, line 20: IPCC is not a model intercomparison study.

4) 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?

5) Page 21406, line 18: 15% of the air mass or volume?

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