Atmos. Chem. Phys. Discuss., 7, S9345–S9352, 2008
www.atmos-chem-phys-discuss.net/7/S9345/2008/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Aerosol distribution over Europe: a model evaluation study withdetailed aerosol microphysics" *by* B. Langmann et al.

B. Langmann et al.

Received and published: 21 February 2008

Answers to Reviewer 1:

General comments:

The following sentences have been added to the manuscript on page 17896, section 2:

'Nitrate and ammonium are not yet considered in the M7 version applied here, however, M7 is flexible to be extended to more components. Nitrate aerosols are expected to become more important in the future atmosphere due to increase in nitrate precursor emissions and the decline of (NH4)2SO4 aerosols in wide regions of the Earth (Bauer et al., 2007)'

Specific comments:

1) Thanks for the advice. This information should indeed appear in the model set-up section. The following sentences have been modified in / added to the manuscript on page 17898, section 2:

'REMOTE is applied with a time step of 5 minutes in 0.5° horizontal resolution and 20 vertical layers of increasing thickness between the Earth's surface and the 10 hPa pressure level using terrain following hybrid pressure-sigma coordinates. The prognostic equations are written on an Arakawa-C-grid (Mesinger and Arakawa, 1976). The height of the lowest layer with prognostic trace species concentration is approximately 40 m, dependent on surface pressure.'

2) Considering the eastern European sites of Czech Republic, Latvia, Lithuania, Poland, Russia, Slovakia and Slovenia only for the scatter diagrams, shows such a tendency only for June 2003, not for the winter time, when a much bigger effect due to domestic coal heating would be expected. Therefore, it seems that EMEP has introduced a correction to an overestimation of SO2 emissions in Eastern Europe.

3) Ship emissions are held constant throughout the year. A pronounced difference in SOx emissions between January and July occurs over Eastern Europe due to domestic burning.

4) Thanks for this advice. This information should have been included in the model set-up section. Biogenic VOC emissions are determined based on Guenther et al. (1991, 1993) in REMOTE. The photochemical model is the RADM II model as described in Stockwell et al. (1990). It is a widely used module in regional air pollution modelling, e.g in the EURAD (Hass et al., Atmos. Environ. 27, 867-887, 1993) and the WRF (Grell et al., Atmos. Environ. 39, 6957- 6975, 2005) model. Various evaluation studies have demonstrated that RADM II is a suitable tool to determine tropospheric photochemistry.

The following sentences have been modified in / added to the manuscript on page 17896 and 17897, section 2:

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'After being released in the atmosphere, gas phase and aerosol phase species undergo transport processes (horizontal and vertical advection (Smolarkiewitz, 1983), transport in convective clouds (Tiedtke, 1989), vertical turbulent diffusion (Mellor and Yamada, 1974)) and are removed from the atmosphere by sedimentation, dry and wet deposition.'

'Photochemical production and loss in REMOTE is determined by the RADM II chemical scheme (Stockwell et al., 1990) by 163 chemical reactions in the gas phase including a wide range of hydrocarbon degradation reactions. Photolysis rates are calculated as described by Madronich (1986) and Chang et al. (1987). Aqueous phase chemistry processes is implemented according to Walcek and Taylor (1986).'

'In addition to anthropogenic emissions, terrestrial biogenic terpene and isoprene emissions from forests are considered based on Guenther et al. (1991, 1993).'

5) The line of agreement in Fig. 7 left is missing by purpose to emphasise that the scale of the x and y axis are different as noted in the figure caption. This is in contrast to all other scatter plots included in the manuscript.

Answers to Reviewer 2:

1) Information about the model characteristics have been improved (see also answer to reviewer 1, specific comment 1).

The model REMOTE has been successfully applied to investigate several scientific questions in different regions of the Earth. Several of these studies carefully evaluate the meteorological model simulation results against available measurements (T2m, wind speed and direction, precipitation). The diurnal variability of the meteorological and chemical processes in the planetary boundary layer as well as the planetary boundary layer height and spatial variability could be reproduced by REMOTE pretty well over Europe (Langmann, 2000; Langmann and Bauer, 2002; Langmann et al., 2003). Two recent papers (Marmer and Langmann, 2007; Marmer et al., 2007) in-

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vestigate the inter-annual variability of aerosol distributions over Europe by using a bulk mass approach. In Indonesia, for example, REMOTE overestimates precipitation (Langmann and Heil, Atmos. Chem. Phys. 4, 2145-2160, 2004; Heil et al., Mitig. Adapt. Strat. Glob. Change 12, 113-133, doi:10.1007/s11027-006-9045-6, 2007) and thereby wet deposition, the main removal process of aerosols from the atmosphere. However, compared to Indonesia and its huge convective activity, European climate is much more moderate and easier to reproduce by numerical modelling (see also answer to comment 2)

2) p. 17897: Thanks for the advise. This information should indeed appear in the model set-up section (see also answer to reviewer 1, specific comment 1). The following sentences have been modified in / added to the manuscript on page 17898, section 2:

'REMOTE is applied with a time step of 5 minutes in 0.5° horizontal resolution and 20 vertical layers of increasing thickness between the Earth's surface and the 10 hPa pressure level using terrain following hybrid pressure-sigma coordinates. The prognostic equations are written on an Arakawa-C-grid (Mesinger and Arakawa, 1976), where the height of the lowest layer with prognostic trace species concentration is approximately 40 m, dependent on surface pressure.'

The following sentences have been added to the manuscript on page 17899, section 3.1:

'Previous studies with the REMOTE model have evaluated the meteorological model simulation results against available measurements (T2m, wind speed and direction, precipitation). The diurnal variability of the meteorological and chemical processes in the planetary boundary layer as well as the planetary boundary layer height and spatial variability could be reproduced by REMOTE pretty well over Europe (Langmann, 2000; Langmann and Bauer, 2002; Langmann et al., 2003). Two recent papers (Marmer and Langmann, 2007; Marmer et al., 2007) investigate the inter-annual variability of aerosol distributions over Europe by using a bulk mass approach.'

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3) p. 17897: In the current model set-up, primary organic carbon aerosol particles are released only into the soluble accumulation and aitken mode. Secondary organic carbon aerosol (SOC) formation is not yet considered. To analyse if SOA is formed by condensation of low vapour pressure organic gases on pre-existing particles, or by forming new particles in the atmosphere by nucleation will be subject of a future study.

We added on page 17897, section 2:

'Secondary organic aerosol formation is not considered in the current study.'

Fig. 5 of O'Dowd et al., Geophys. Res. Lett., L01801, doi:10.1029/2007GL030331 (2008) shows near surface sea-spray mass concentration (top) in the accumulation mode as determined by the REMOTE model and percentage organic contribution to sea spray (bottom), where sea-spray is assumed to consist of sea salt and organic carbon only. For the model simulations described in the current manuscript we didn’t store the monthly natural emissions fluxes which are determined in the model dependent on the meteorological conditions. In addition, we do not take DMS emissions from the ocean into account directly.

4) p. 17898, lines 6-8: The impact of aerosol particles on clouds and precipitation formation has not been taken into account in the present study, so the cloud microphysics is not affected, neither by accumulation and coarse mode sulfate particles nor by smaller particles.

5) p. 17898, line 22: corrected

6) p. 17900 and Figs 2 and 3: As already mentioned on page 17899 of the manuscript, the model grid box where the Mace Head site is located, is covered approximately half by water and half by land. As surface friction over land is higher than over water, the modelled wind speed is slightly lower than the measured ones. Looking at the modelled wind speed one grid box to the west (open Atlantic), the temporal evolution of wind speed during the two months remains similar, but slightly higher wind speeds as ob-

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served are modelled. Even though no observations over the open ocean are available, there is no indication from the comparison with the Mace Head data that wind speed should be underestimated by REMOTE. At Mace Head, sea salt mass concentration in TSP has been measured and REMOTE model results agree reasonably well (see Fig. 4 from O'Dowd et al., Geophys. Res. Lett., L01801, doi:10.1029/2007GL030331, 2008). For the Danish and Norwegian sites in Europe only monthly mean data are available for sea salt, but not wind speed so that we could not analyse the reason for the overestimation at these sites. Wet deposition of chloride cannot be said to be overestimated in general by REMOTE (Fig. 3b of the manuscript), but it is indeed overestimated in June at the Danish sites.

7) p. 17901, line 15: Why should oxidant limited conditions not occur during summer in the model atmosphere? The diurnal cycle and spatial variability of the planetary boundary layer height as determined by REMOTE has been evaluated in former studies (e. g. Langmann, 2000). Langmann and Bauer (2002) and Langmann et al. (2003) investigated the impact of trace species concentrations at the boundaries of the limited area model REMOTE and found an impact of up to 10 % on ozone concentrations in the PBL. For the current study, the boundary trace species concentrations, e.g. for ozone have been fixed, which offers another explanation for the oxidant limitation. Convective transport into and out of the PBL is another source of uncertainty. Some investigations are described in Langmann (2000). As mentioned on page17897 of the current manuscript, a size dependent dry deposition parameterisation is already in use in REMOTE. Overestimated SO2 emissions by EMEP in Eastern Europe as mentioned by reviewer 1 could also contribute in summer to the overestimation of SO2 atmospheric concentration.

On page 17901, section 3.5 the following sentences have been added:

'Another possible explanation for oxidant limited conditions is that lateral boundary concentrations are held constant throughout the simulation. By applying a global to mesoscale model chain, Langmann et al. (2003) showed for ozone concentration that

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the global model dominates the nested higher resolution model results increasingly with height. In addition, convective events couple free troposphere and PBL air masses so that ozone from above can be injected into the PBL contributing an amount of 5-10 ppbv to near-surface ozone in the afternoon hours.'

8) p. 17901, line 20: A good agreement is stated in the manuscript on page 17901, line 20 explicitly for January, not for June. For sure, there is agreement that H2O2 is the main oxidising agent for SO2 in the aqueous phase, besides O3 in the aqueous phase, both pH dependent, and OH in the gas phase, in particular during summer. The manuscript focuses on O3 as an indicator for the oxidising capacity of the atmosphere, as neither H2O2 nor OH concentrations are measured routinely.

To clarify this issue, the following sentences have been added to the manuscript on page 17896, section 2 (see also reviewers 1 specific comment no 4): 'Sulfate production in the aqueous phase is determined dependent on pH via oxidation by H2O2, O3, methylhydrogenperoxide, peroxyacetic acid and catalysed by Fe3+ and Mn2+. Cloud water pH is determined solving iteratively an ion balance which is continuously maintained (Walcek and Taylor, 1986)'

9) p. 17903, lines 4-5: Thanks for the comment, but the current manuscript under review does not take into account any impact of aerosol particles on clouds.

10) p. 17903, lines 11-12: Yttri et al. (2007) analysed OC and BC concentrations at coastal and inland European locations. As already mentioned on page 17903, line 8, transport of marine POC from the ocean to continental areas is found to be relatively small. For the model simulations described in the current manuscript we didn't store the monthly natural emissions fluxes which are determined in the model dependent on the meteorological conditions. We explicitly included in the model set-up section that SOC is not taken into account in the current study (see also answer to comment 3). POC is not added to the insoluble mode. In a sensitivity test, we attributed POC emissions to an insoluble mode only. The results were only slightly different, as hygroscopic growth

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of OC aerosols is currently neglected in REMOTE/M7 as well as in ECHAM5-HAM (Stier et al., 2005).

11) p. 17904, line 11: corrected

12) p. 17906, Fig. 9: The differences (REMOTE/M7 minus bulk approach) between the two versions for June 2003 in the first model layer are comparable to the differences shown in Fig. 10 of the manuscript. We decided to present the column difference in the manuscript because it is more suitable to point to transport processes across the lateral boundaries. In cloud sulfate formation is very similar in both model versions (see also answer to comment 8).

13) p. 17908, lines 3-4: By providing more information about previous studies on the simulated meteorological situation (see answers to comment 1 and 2) we hope that the conclusion is now more convincing.

Interactive comment on Atmos. Chem. Phys. Discuss., 7, 17893, 2007.