

Interactive comment on “Technical Note: simulation of detailed aerosol chemistry on the global scale using MECCA-AERO” by A. Kerkweg et al.

A. Kerkweg et al.

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We thank the three anonymous referees for their valuable and constructive comments, to which we reply in the following:

Comments to Referee No. 1:

- 1.) We changed this.
- 2.) M7 is an aerosol dynamical model, developed by Vignati et al. at ISPRA (as cited in the paper). This aerosol model describes the aerosol distribution by 7 lognormal modes (4 modes regarded as soluble modes and 3 as insoluble

modes). We added the latter information to the revised manuscript.

- 3.) Unfortunately, the suggested statement is partly wrong. The mixing ratios themselves are not determined by the ratio to Cl^- . Although the tracer mixing ratio tendencies (which are added to the tracer mixing ratio due to the emission process) are scaled by the ratios of the assumed abundance in sea water, the tracer mixing ratios of Cl^- , Br^- , I^- , IO_3^- and HCO_3^- are independently changed by other processes, e.g. by chemistry. Thus the actual tracer mixing ratios of these ions are not simply related to each other by their ratios in sea water.
- 4.) The emission flux of Na^+ is the sum of the emission fluxes of Cl^- , Br^- , I^- , IO_3^- and HCO_3^- . This is required, since otherwise the electro-neutrality of the sea salt aerosol would be violated. This leads to a difference in the emission of Na^+ compared to the calculated sea salt mass flux, however this difference is small, as the emission fluxes of Br^- and HCO_3^- (the second most abundant species) are three orders of magnitude smaller.
- 5.) These are results of the same simulation as shown in Section 3. We add this information to the revised manuscript.

Regarding the arctic aerosol chemistry you are right. We can not calculate Arctic halogen chemistry with the current model restrictions. We hope to be able to lower the threshold with further improvements in our model.

Anyway, most of the halogen chemistry in the arctic is likely to happen on the deposited snow, ice and especially in the brine layer. These domains are not yet explicitly included in our model. Thus calculation of halogen chemistry in the Arctic would be useless at this stage of the model development anyway.

- *What aging value is used to define a particle to be 'fresh' or 'old' ? Why do you choose this value ?*

Each particle, that was already in the air before the emission calculation, is assumed to be an old particle, as – due to the process splitting in the model – these particles have undergone all aging processes (e.g., chemistry, sedimentation, scavenging and wet deposition). Therefore only those particles freshly emitted in each emission calculation are assumed to be 'fresh'. Or – the other way round – aged particles are at least one model timestep old.

This is likely to be the most reasonable assumption we can make as the model provides no further information about particle aging.

- *Page 3313: Does 'the sum of all particles after emission' mean the total particles that are still staying in the air but not the total particles emitted into the air?*

'The sum of all particles after emission' is the number of all particles airborne after emission i.e., the sum of particles airborne before the emission and those particles freshly emitted. Or in other words, it is the total number of airborne particles after sea salt emission but before deposition calculations. We added some additional clarifying words in the revised manuscript.

- *Comment Page 3319, line 1-2: the correction termbecomes most important in regions with highest wind speed. Could you explain this phenomenon to the reader? In equation 10, the change of ratio with wind speed is not clearly seen.*

At high wind speeds the emission flux is highest and the size of the aerosol particles is largest (this is inherent in the number of new particles in equation 10). Thus a high amount of sea salt mass is emitted. BUT most of this mass is instantaneously deposited because the particles at high wind speed are larger and thus heavier. Thus, in reality the net emission flux (i.e. emission - deposition) is approximately the same as at low wind speeds. In the model without the correc-

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Comments to Referee No. 3:

- *p. 3305, line 20 - p. 3306, line 9; as well as section 2.2:*

We describe only sulfuric acid, as this is the species for which we actually included this procedure. Of course it can be applied for other species as well. But we wanted to avoid the impression that the treatment of other species than sulphate is already implemented. For this reason we refrain from a generalised description. This statement will be included in the revised manuscript.

To answer to the second part of your statement is difficult, as we seem to talk about different models. MECCA-AERO and M7 are independent submodels within the MESSy framework. They themselves do not include advection, dry deposition or scavenging. The tracers defined by the ADM or ACM submodels are treated independently by the ECHAM5 advection and the MESSy submodels DRYDEP and SCAV.

The independence of the models (i.e., the operator splitting approach) is the reason for the overlap, as both models require sulphate condensation. The ADM calculates condensation onto all modes, whereas the ACM calculates the explicit chemistry only for a subset of the ADM modes. To treat the condensation of different modes by different submodels (with different approaches) would lead to undesired inconsistencies.

To call the ADM from the ACM would not solve the problem, as the ADM does not handle all the species resolved in the ACM.

- *Section 2.3*

The effect of dry deposition is largest, as it is calculated directly after the emissions in the operator splitting sequence of our model. Thus the dry deposition accounts for a part of substance removal, which would alternatively happen through sedimentation, if the order in the operator splitting sequence would be the other way round. We add this information to the revised manuscript. Additionally, sedimentation is largely driven by the ambient radius of the aerosol. In our model only particle numbers and dry sea salt mass are emitted. Thus the new, to the emissions adapted radius is only available after the calculations of the aerosol dynamics. But the aerosol dynamics itself establishes the internal mixture, i.e., the differentiation between “old” and “fresh” particles is not longer possible.

- *p. 3302*

We removed the Amundsen et al. reference in the revised manuscript.

- We changed the next three items according to the referees suggestion.

- *p. 3303, lines 18-20*

We removed the references to Amundsen and Toppings in the revised manuscript. But as Metzger and al. applied their approach in a global model by now, we changed the reference to a more recent one including a global study. (Metzger and Lelieveld, ACPD, 2007)

- *p. 3303, lines 20-21*

We do not think the capabilities to be similar. The most important point in MECCA-AERO is the explicit calculation of non-equilibrium aerosol chemistry. As is stated in Jacobson et al., 2002, they are using EQUISOLVII to resolve the aerosol chemistry. Thus they only included equilibrium aerosol chemistry. That is a big difference, e.g. for bromine chemistry.

- We changed the next three items according to the referees suggestion.

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- *Why is $F_{\text{dep}}(X)$ so different from $F_{\text{dep}}(SS) \times f(X)$?*

The difference arises from of the different composition of aged and fresh sea salt. For chlorine the difference is large for highly acidified aerosol particles only. For bromine it is large, because bromine is very efficiently depleted in the aerosol phase. Thus, underestimating the removal of fresh sea salt leads to an overestimation of the source of gaseous bromine. The second large effect is on the aerosol pH. Emission of HCO_3^- adds alkalinity to the aerosol. If its emission is overestimated the aerosol would be to alkaline, hence the aerosol chemistry differs. We are going to add this information to the revised manuscript.

- *Eq. 11*

$F_{\text{realdep}}(X)$ is the most realistic flux, whereas F_{corr} is the flux we have to apply to correct the calculations in the model. We changed the text in the revised manuscript to clarify this.

- *p.3315, l.9-10*

In this part of the section we are talking about different setups w.r.t. the chosen reaction mechanism. In all cases we used the Rosenbrock solver of 3rd order with automatic timestepping.

The stability problems obviously occur by application on the global scale and the usage of a varying aerosol distribution. In box and column models the boundary conditions are mostly relatively smooth, e.g., no large temperature or humidity gradients occur, and the aerosol physical properties, such as the liquid water content are not changing so rapidly, especially as the time steps are usually much shorter as in global models. In a global model effects of e.g., clouds, advection etc. combined with a much larger time step, can lead to much higher differences in each grid box yielding conditions far from equilibrium and an increased stiffness of the chemical PDE system.

- *p. 3316, lines 11-14*

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The restriction to a liquid water content of $10^{-12} \text{ m}^3/\text{m}^3$ for simulations including bromine chemistry is a requirement for the numerical stability. We were not yet able (without solver crashes) to integrate stiffer systems (i.e. systems with a lower LWC) under all atmospheric conditions, if bromine chemistry was included in the system.

The error introduced by neglecting the chemistry for aerosol phases with a liquid water content smaller than $10^{-12} \text{ m}^3/\text{m}^3$, is certainly smaller than the error of neglecting the process completely. W.r.t. bromine chemistry in the marine boundary layer, the chemistry of MECCA-AERO enables us to simulate the source of gaseous bromine and the recycling within the marine boundary layer. This is the most important part, which was so far parameterised in all global model studies. Indeed it would be better to be able to simulate it in the whole troposphere, but for this we need further improvements (as discussed in the paper). We are still working on these improvements.

- We are going to apply the required technical corrections.

Interactive comment on Atmos. Chem. Phys. Discuss., 7, 3301, 2007.

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