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Atmospheric
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Interactive comment on “Global cloud and precipitation chemistry and wet deposition: tropospheric model simulations with ECHAM5/MESSy1” *by* H. Tost et al.

H. Tost

tost@mpch-mainz.mpg.de

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Comments to Referee #1

We thank the referee for his helpful comments, which we will try to answer in the following:
Point 1:
The detailed submodel description is given in Tost et al.(2006). The assumed droplet spectrum for precipitation follows Best (1950), and is required to determine the phase transfer coefficients, since the transfer coefficient of the mean radius is not identical to the mean transfer coefficient.

Point 2:
As stated in the manuscript the reaction mechanism including the labels can be found in the supplement. A sentence will be added stating, that SCM focusses on the uptake of the most soluble compounds, the acid-base equilibria and the SO_2 oxidation in the liquid phase by O_3 and H_2O_2. Both chemical reaction systems are coded with the kinetic preprocessor (KPP) and solved during runtime as a coupled differential equation system.
Point 3:
SCM and COM determine the pH from the dissolution of all relevant species. If aerosols are scavenged they influence the pH as well due to their composition (e.g. sulphate). However, the cloud microphysics is not linked to aerosol distributions in the current model setup. Initially, the pH start value for a chemical integration time step is neutral, but quickly adjusts due to the dissolution of the acidic compounds. With respect to precipitation the starting value is determined by the incoming precipitation flux from the layer above.
Point 4:
In the previous versions the precipitation LWC has not been determined by the amount of water falling through a specific layer, but by a parameterisation of Mason(1971).
Point 5:
These limitations are the more or less correct representation of the simulated precipitation fields, e.g. an overestimation of rainfall in the ITCZ, the fractionation into large-scale and convective rainfall, overestimated precipitation in Central Africa leading to very high wet deposition fluxes or an underestimation over the United States which partly causes too acidic rain water (see Section 4.2). This is already mentioned in the manuscript and will be added as supplementary arguments.
Point 6:
The aerosol submodel does not take aerosol nitrate into account. However, evaporating clouds and precipitation can lead to aerosol nitrate. This is considered by an aerosol nitrate tracer that does not influence the aerosol microphysics, but is trans-

ported and treated by the sink processes (also scavenging) as an aerosol compound.

Point 7:

Lightning $\mathrm{NO_x}$ contributes to the wet deposition, but is by a factor of ten lower than the local biomass burning emissions, and consequently not responsible for the enhanced nitrate deposition values. These are also seen in the simulated $\mathrm{NH_x}$ deposition values which originate also from a biomass burning source and are located in the same regions.

Point 8:

The data in Africa used in this study has been updated during this review process. The old version was announced to change (F. Dentener, personal communication), but was not received in time for the publication. Therefore the analysis will be repeated with more recent and representative data from the IDAF network, including the references for the updated data.

Point 9:

In case of a less efficient scavenging due to precipitation underestimations larger amounts of $\mathrm{NH_3}$ can be transported further downwind, since the gas phase chemistry of ammonia and aerosol processes (only the same as for nitrate) are not considered in this model setup. Consequently, deposition processes are the only sinks. Therefore, the deposition can occur further away from the sources if the precipitation distribution (temporal and spatial) is not captured accurately and this can have an impact also on accumulated values.

Point 10:

The correlation to the EMEP data is also worse in Dentener et al.(2006) than for the NDAP. To some extent this can be caused by a worse representation of the meteorology, fewer stations that are probably not representable for a larger region (e.g., Ispra in northern Italy is located at the bottom of mountains, but the model grid cell spans from the coast of the mediterranean into the Alpes. For the NDAP data in some gridboxes several stations are located. Therefore, one model box value has more chances to agree well with a station value. Additionally, capturing the meteorology in Europe is more difficult than in the US, due to the more complex European orography.

Point 11: adding vertical bars to a table:

The table layout is done by ACP/ACPD. In the original manuscript such vertical bars have been in the table, but were removed during the editing.

Point 12:

I think that both aspects are responsible for that. For instance the scavenging of HCHO is mainly independent of pH, but also for $\mathrm{HNO_3}$ which dissociates almost completely in any solution with a pH higher than 0 the pH limitation between 5 (prescribed) and 3 to 6 (explicitely simulated) cannot alone lead to the differences between the simulations. Nevertheless, this aspect will be added.

Point 13:

In highly polluted regions a high content of organics can increase the $\mathrm{NO_2}$ recycling for ozone production, but as well for the formation of nitric acid. Due to the high effective solubility, this effect is expected to be dominant over the pH dependent uptake (see topic above).

Point 14:

The vertical resolution is different in the EVAL setup (Jöckel et al, 2006) and the simulations using the different aqueous phase chemistry. Consequently, the meteorology is different, including parameters in the model formulation (microphysical constants, etc.). Furthermore, the EVAL values are averaged over several years whereas in Table 4 only the year 2000 data from the simulations is chosen (compare the Taylor diagrams for the interannual variability of the EVAL simulation).

Point 15:

A discussion of the lower part of Table 4 will be added in a revised manuscript comparing the values with Table 2.

Point 16:

These comparisons are given in the text including the observations, their location and references, which can be compared with Figure 5.

Point 17:

Some of the formulations must be changed. However, if the deposition of e.g. $\mathrm{HNO_3}$ (value independent of water amount) is too high, the too high acidity can result from both too high deposition and too low amount of water.
Point 18: Convective scavenging:
The simplification in using only the cloud liquid water that is converted to precipitation does not necessarily imply a too high rain-out efficiency. For large scale clouds the total amount of dissolved material in the cloud is multiplied with the fraction of how much water rains out. In the convective part, the dissolved material is completely scavenged, but the amount of water available for the scavenging is lower than the actual water content. For very soluble species, e.g. $\mathrm{HNO_3}$ this indeed can overestimate the wet deposition, whereas for other compounds it hardly has any effect. However, if the $\mathrm{HNO_3}$ is taken up by the cloud it will dissolve and form aerosol nitrate after cloud evaporation, that will sedimentate and finally be deposited as well (dry or wet deposition), but hardly take part in gas phase chemistry. In general this is a weakness of the scheme, that will be removed in the next model versions.
Point 19: Section 4.3:
Currently we think about a revision of this section, especially the part of ozone. We would like to keep the Figures since they offer a direct comparison of near surface mixing ratios between the different simulations and will probably also like to keep the species separated as much as possible. However, in argumentation chains involving more species these links will be added more clearly.
Point 20: the vertical profiles:
These figures will be revised in a way to obtain more clarity, but without the loss of too much information.
Point 21:
Even though these terms are often used in combination with aerosols, direct and indirect effects are not restricted to this topic. Since scavenging directly influences some species whereas others are only affected by the result of the scavenging (what is exactly the meaning of direct and indirect) we like to stay with the terms. We will add a

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note, that these may not be mistaken for any aerosol effects.

Point 22: Table 5:

It is not the square root that is negative. The square root shows how the geometric average in units of standard deviation is determined, but the bias itself can be positive or negative.

Point 23: volcanic $\mathrm{SO_2}$:

The volcanic emissions were neglected in all simulations. We agree to a certain degree to the arguments of referee #1. However, some of the volcanic emissions are located in regions far from the observation stations and can therefore have an impact on vertical profiles in remote regions. In the global budget they might not appear significant, especially regarding some of emission uncertainties in East Asia, but to some degree they can have impacts away from the pollution centers. The third argument might as well be true, and the improved model will show in upcoming simulations, even though the bias for $\mathrm{HNO_3}$ and HCOOH is much lower than for $\mathrm{SO_2}$.

Point 24:

We agree with the referee, that the studies of Lelieveld and Crutzen(1991) and Liang and Jacob(1997) analyse two different aspects, the further the effect of scavenging and removal of species from the gas phase, whereas the latter the detailed aqueous phase chemistry. Therefore, it is not surprising that completely different results were obtained, since the first effect is much more important as the second, just the same way this is stated in our manuscript.

Point 25:

Improved resolution hopefully will result in a better representation of the hydrological cycle as a prerequisite for scavenging and aqueous phase chemistry. The interaction with the ice phase will make the removal of nitric acid, PAN, and other compounds from the upper troposphere more realistic (if not completely removing a fraction from the species, at least a vertical downward transport process into the middle and lower troposphere where later on scavenging by liquid clouds will finally remove these compounds). Further improvements are the correct treatment of convective cloud water,

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helping to overcome the issues discussed above and a more realistic aerosol nucleation scavenging.

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