

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

### **Anonymous Referee #1**

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Review of Tost et al.: “Global cloud and precipitation chemistry and wet deposition...”

Wet removal of trace species is a highly uncertain process in the atmosphere and therefore difficult to represent accurately in large scale chemistry transport models or chemistry climate models. It requires a good representation of the hydrological cycle, involving evaporation and cloud and precipitation formation, the horizontal and vertical distribution of atmospheric trace species, and chemical transformations in the gas and aqueous phases. The balance between aqueous phase and gasphase concentrations of trace species is delicately governed by their solubility, but may further be affected by other species through their influence on the acidity. It is extremely worthwhile to

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investigate how a model represents wet deposition of species, since it does not only reflect a validation of the model but also contains information on the specific processes that lead to model errors and may need improvement. The work of Tost et al. is a thorough investigation of the wet deposition characteristics simulated by the coupled chemistry-climate model ECHAM5/MESy1, covering a multitude of species and different approaches of the chemical system and the scavenging chemistry and providing insight in associated relations and feedbacks.

The paper is well suited for publication in ACP. The paper is constructed in a logical way, but more information is needed in some places as indicated below. Some arguments are not solid and can be improved. Section 4.3 is rather lengthy and I suggest to shorten and rewrite this for clarity.

Specific comments:

page 3, column 1, line 8, "... even though ....spectra are assumed": what assumption is made and how does this influence the dissolution?

p3, c1, bottom: Can you briefly describe in more detail how the SCM and COM chemistry representations differ?

In EASY2 a pH is prescribed. How is the initial cloud water pH (i.e., before dissolution and dissociation of trace gases) established in the other experiments, is there a contribution by aerosol on which the clouds grow?

p3, c2, l14: what is the correction of the precipitation LWC?

p4, c1, bottom: "the precipitation distribution ... limitations ... have to be considered..." what is the nature of these limitations? As far as I could see the limitations are not explicitly considered in the evaluation.

p5, c1, top: Does the model consider aerosol nitrate or does the nitrate deposition originate from gaseous HNO<sub>3</sub> alone?

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p6, c1, line 15: “biomass burning...”, does lightning NO<sub>x</sub> significantly contribute to the nitrate wet dep in Africa?

p6, c1, line ~25: “the observations partly ... specific year ... not representative” Later a simulation for 2000 is mentioned. Is the model-observation agreement with this simulation better?

p6, c2, l11 “Furthermore, the effects ... precipitation rates ... such as NH<sub>3</sub>”. I can imagine this is important for individual precipitation events, but to what extent does it influence monthly averages or cumulative amounts?

p7, c2, Table 2. I’m surprised by the small correlations between model and observations for the EMEP nitrate and sulfate, especially when compared with the good correlation for NADP and given the fact that the emissions in North America and Europe are relatively well known (as stated 7 lines higher). What is the cause of this, does it pertain to a specific season, a bad representation of meteorology?

p8, Table 8. Insert vertical bars before 2nd, 6th and 10th columns for clarity.

p8, c1, middle “This is partly caused...” I think that the prescribed pH=5 is a more likely cause.

p8, c1, bottom “.. altered oxidation capacity.”: Why is that, and how much is it affected?

p9, Table 4. Why are the model values in Table 4 different from those in Table 3? Is that caused by a different vertical resolution? If so, how does the resolution affect the scavenging?

p9, Table4: the lower part of Table 4 is not discussed. Either discuss or remove.

p10. Given the fact that pH is extensively discussed, why not include a table with modeled and observed pH? Also the abstract says: “... prognostic calculations of the pH ... results are also in accordance with observations.” That should be demonstrated.

p10, c1, lines 12-17: This may partly .... underestimation of total precipitation .... over-

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estimation of  $\text{H}_3\text{O}^+$  concentration...” As written here, both reasons come down to the same conclusion: when the water amount is too low, the concentration is automatically too high. You can not directly compare the sulfate etc. deposition (in  $\text{mg}/\text{m}^2$  therefore independent of water amount) and the acidity (mole/l water, so directly depending on water amount). The same goes for the statement a few lines below on pH and ammonium in Europe.

p10, c2,l 13: The text states that for scavenging by convective clouds the cloud liquid water is not considered but only the precipitating fraction. This implies that the simulated rain-out efficiency will be overestimated and the lifetime of soluble species will be too short, especially in the tropical regions. Can you comment?

Section 4.3. In the different sensitivity studies the behaviour of each of the species is discussed separately. This makes this section rather lengthy and it is difficult to get a good idea how the chemical system behaves. I would suggest to rewrite this in terms of the system as a whole. In my opinion an important factor is the scavenging of  $\text{HNO}_3$  and  $\text{N}_2\text{O}_5$ . A change in scavenging efficiency influences atmospheric  $\text{NO}_x$ , and therefore  $\text{O}_3$  and  $\text{OH}$ . This affects production of  $\text{HO}_2$  and therefore of  $\text{H}_2\text{O}_2$ . The latter is also affected by scavenging, whereas  $\text{SO}_2$  depends on scavenging and the oxidant concentrations. Less scavenging  $\rightarrow$  higher  $\text{NO}_x$   $\rightarrow$  more  $\text{O}_3$ ,  $\text{OH}$  and  $\text{H}_2\text{O}_2$  etc. Remove figures 6, 8, 10, 12 and 14.

Regarding figures 7, 9, 11, 13 and 15: I find the latter plots a little “messy” (pardon the pun), it is hard to distinguish the modeled average profile from the spread. Maybe the spread can be indicated by horizontal bars at different altitudes.

General: the authors often use “direct effects” and indirect effects” for chemical influences. Since these terms are often associated with aerosol climate effects, which are not meant here, I suggest a different phrasing.

Table 5. I do not understand how a square root (table caption) can yield negative numbers.

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p20, c2, below. “The underestimation in COM and SCM ... volcanic SO<sub>2</sub>” I find this strange for several reasons. 1) Were these emissions considered in the other experiments, why is there no similar underestimation in the other simulations? 2) Apart from that, I doubt that the neglect of volcanic SO<sub>2</sub> would have such a large effect on a global scale. At least, it should be expressed in the sulfate deposition which would be consistently smaller than observed, but this is not the case. 3) A too low concentration does not necessarily mean too low emissions. In steady state:  $[SO_2] = Em/Loss$ , so Emission may be too small, but also Loss may be too large. Since HNO<sub>3</sub> and HCOOH concentrations are also underestimated it is more likely that wet removal is too efficient (maybe associated with the way convective precipitation scavenging is treated - see earlier comment).

p21, c1, lower half: Can you say that the study of Lelieveld and Crutzen reflects for a large part the importance of the effect of HNO<sub>3</sub> scavenging and subsequent NO<sub>x</sub> effects on O<sub>3</sub>, whereas Liang and Jacob focus on the effect of aqueous phase radical chemistry?

p21, c2, lower half: “Improved model simulations ... improvements of the aq. phase chemistry ... ice clouds ... frozen precipitation” Can you explain what specific improvements you expect from this?

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Interactive comment on Atmos. Chem. Phys. Discuss., 7, 785, 2007.

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