

Interactive comment on “Uncertainties in atmospheric chemistry modelling due to convection and scavenging parameterisations – Part 1: Implications for global modelling” by H. Tost et al.

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Author comment to Uncertainties in atmospheric chemistry modelling due to convection and scavenging parameterisations – Part 1: Implications for global modelling

by H. Tost, M. G. Lawrence and P. Jöckel

We thank all the referees for their helpful comments, efforts in reviewing the manuscript and the discussion.

While conducting this study, we have mainly aimed at making a comparison with two
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field campaigns. Analysing the data, we found that it would be interesting to show the results that we obtained on the global scale, in particular the implications of the use of different convection schemes. Because the combined work would have become rather extensive we decided to separate the general global analysis from the field campaign analysis. As only one of the referees has a similar opinion that the global results are interesting on their own, we now plan to merge the anticipated Part1 and Part2 (discussing the observations of the two field campaigns) into one paper. Consequently, this merged manuscript will contain more information than the current version, which is already considered slightly long. Therefore, a complete revision of the manuscript is required. Furthermore, several additional colleagues, i.e., those who contributed to the measurements have to be directly involved in the analysis, implying that we need more time to finish it. Therefore, we ask for a little more patience from the editor and the referees, before a revised version can be submitted. We hope the referees will still be willing to review the revised manuscript in a similar detailed way, since it will differ significantly from the current version published in ACPD.

Comments to Referee#1:

We hope, that with the planned revision the comments of referee#1 will be satisfied. However, as we already mentioned in the submitted paper, a “judgement” of the convection schemes, as desired by the referee is rather complicated, and is likely not going to be a definitive one, rather it will depend on the compound, and the location and meteorological conditions.

Comments to Referee#2:

1) Our formulation was indeed poorly chosen . Even though the measurement data for radiation and precipitation has - of course - uncertainties (e.g., comparing NOAA and ERBE radiation or CMAP and GPCP precipitation data), our intention was to show that all model simulations more or less accurately reproduce the observed precipitation and radiation fields. This will be reformulated. The detailed differences between the

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schemes implemented in this model applied without re-tuning have been published previously. (Tost et al., ACP, 2006, Influence of different convection parameterisations in a GCM).

2) We completely agree that the convective activity and the averaged mass fluxes have contributions from both, strength and frequency. This will be analysed in more detail (with anticipated figures in the supplement) in the revised manuscript.

3) The reference should point to Eq. 2. And the overturning time is, of course, shorter in simulations with stronger convective activity; we thank the referee for pointing out this mistake.

4) We agree, that this cannot be concluded from Figure 1. However, the way most of the parameterisations work is that they “adjust” the atmosphere to a more stable state by a redistribution of the moist static energy. These schemes mostly contain a certain relaxation time, before a stabilised state is reached. The stronger the convective activity the more likely it is also that the scheme is active several subsequent time steps, implying a higher frequency of convective activity.

5) We will reformulate this.

6) We will reformulate this.

7) This is a typo; the half-life time is indeed 3.8 days, and the e-folding time 5.5 days.

8) The values in Fig. 3d are slightly higher near the surface, but show lower values (greenish colours) up to an altitude of 700 hPa, i.e., the outflow regime of shallow convection. The formulation will be improved to: “The less intense shallow convection in EMA leads to lower values in the upper part of the tropical boundary layer.” Since the mixing by shallow convection is not as efficient as in T1, the surface concentrations (near the emission source) show higher values, whereas the upper part of the boundary layer is characterised by lower R_n mixing ratios. The higher values in the stratosphere are not significant, even though 100% differences are exceeded, since there is hardly

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any R_n in the stratosphere, and thus the small absolute differences can produce large relative ones. Slightly enhanced values in the lower stratosphere are found since the Ema scheme tends to simulate slightly deeper convection, i.e. the outflow height is higher and the slow ascent above the convective towers due to radiative cooling into the stratosphere has to cover less distance, i.e. a slightly higher fraction of R_n can reach the stratosphere before it decays.

9) Depending on the form of the rewritten manuscript the references to the respective figures (supplement or manuscript) will be checked carefully.

10) In contrast to CO , which only reacts with OH , $HCHO$ undergoes in addition reactions with NO_3 (nighttime oxidation), Cl and Br , and it photolyses in two different pathways. Consequently, more compounds can directly influence the local $HCHO$ mixing ratio as well as the photolysis rate, which depends on the actinic flux. This will be added in the manuscript/supplement.

11) Correct notation will be applied carefully through the revised manuscript.

12) The chemistry of HNO_3 itself is relatively simple, i.e. reaction with OH and photolysis, but the chemistry of its precursors is more complex. As for $HCHO$ this will be mentioned in the revised manuscript.

13) The B1 scheme tends to produce slightly more ice in the upper troposphere, therefore the uptake on the ice (which is parameterised based on the IWC) is more efficient, i.e. slightly depleting the gas phase compared to T1. The lower mid-troposphere values can be explained by the more efficient/undiluted transport into the upper troposphere. Since HNO_3 itself is efficiently washed out, the local production from insoluble precursors and horizontal advection dominates in regions with convective activity.

Technical: Captions and units as well as labels will be checked carefully before submission of a revised version.

Comments to Referee#3:

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Main comments: 1) We are very much aware of the work of Mahowald et al. (1995, JGR) and we are not sure when or how the reference to this work ended up getting dropped out of the current manuscript. In any case we are sorry for this omission. It definitively belongs prominently in the discussion of previous work concerning the topic of parameterised convection and its uncertainties and will be included in the introduction of the revised manuscript. But we do not completely agree, that our approach is not unique, since to our knowledge no study has been published that applies several convection schemes in a 3D general circulation model including comprehensive chemistry in both, gas and aqueous phase, including ice uptake of HNO₃. Therefore, our study goes well beyond the work of Mahowald et al., who applied convection and boundary layer schemes in a single column model applying several meteorological conditions and used representative tracers with various lifetimes, which did not interact with each other.

It is to a large extent these interactions of various compounds with different lifetimes, which lead to the effects described for some compounds in this paper. Even though the findings for Rn and CO are not substantially new (or at least comparable with previous studies), the modified OH mixing ratios leading to differences in the CO values have not been analysed in detail before.

The other criticism, namely that it is not a new finding that short-lived compounds are more affected by convection should also be seen positively: even though complex (and sometimes very fast) chemistry can take part in the conversion of precursors, convection is still a dominating process for short-lived compounds. This can now be concluded with the help of our study. Consequently, we cannot (and actually don't want to) show that previous assumptions have been wrong, but rather confirm those using a more complex and comprehensive model.

A comparison with observations was planned for Part2, but now it will become part of a combined, revised manuscript, without the necessity to assume horizontal inflow or background conditions.

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2) The sensitivity of many compounds is indeed one of the main findings and objectives of this study. Even though this is not in general completely new, it is new with respect to the complex parameter space that has been investigated in our work. The tuning has been applied to rule out additional deficiencies in tracer mixing ratios, which might result from a climate state that has been perturbed by applying the different parameterisations. Since that aspect can now be ruled out (due to the re-tuning and the nudging to the observed meteorology), it is indeed mainly the formulation of the parameterised convection that leads to the differences in tracer mixing ratios. Even though the convection schemes are frequently undergoing improvement, and new more physical or more realistic schemes are also being developed, the parameterisations will always lead to an uncertainty. One major goal of this study is to make the scientific community aware of this large degree of uncertainty in global chemistry-climate models, and to better bring this to the attention of the broader atmospheric chemistry community, especially those who are less familiar with the details of chemistry-climate models and the differences between various models. Furthermore, our study allows a quantification of these uncertainties (in our model system). All the parameterisations we used in our study are valid schemes and are applied in global and/or regional modelling. Even though some new approaches have been developed recently, a complete neglect of one or the other scheme should not be pursued. Even more, as already stated by Mahowald et al. (1995) each scheme might have some advantages and disadvantages, that atmospheric chemistry modellers simply have to live with. An analysis of the meteorological impact of the individual schemes in a “non-tuned and non-nudged” environment has already been shown extensively by us (Tost et al., ACP, 2006, Influence of different convection parameterisations in a GCM), including a comparison with observations and respective correlations, biases, etc., whereas the current paper really focuses on the impact on atmospheric chemistry.

3) The “tuning” of the model setups are definitively specific to the current setups, i.e., depending on the large-scale condensation and the boundary layer scheme. We here completely agree with the referee. Consequently, the findings for individual compounds

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from this study cannot be expected one-by-one from a different model, even if it uses the same convection scheme. The combination of all physical components in a modelling system determines to which degree a model has to be tuned and to achieve similar convective activity. The same scheme might need a different set of tuning parameters in another model setup. That is one of the reasons why we re-tuned the model setups by modifying convective parameters only. The general statement that the choice of the convection scheme can impact the simulated results will nevertheless be true for all model simulations which use a parameterisation. This has already been touched in the discussion section of the ACPD version of this paper. As already stated above, we do not believe that a general judgement of one scheme being superior to the others will be possible from this or any other study, thus it was more important for us to characterise and quantify the differences resulting from the convection schemes. It is questionable anyhow, if such a judgement is possible at all. The limitation to four months simulation time is indeed an issue. As stated at the top of our reply, the primary intention was an investigation of the two campaigns. The findings are not necessarily representative for a full year or even a decade, but most of the compounds analysed have a lifetime substantially shorter than the simulation period. Initialisation has been chosen from a long transient simulation using the T1 scheme (that is why we chose this as our reference). In other seasons the effects of the convection parameterisations might indeed be different, but it is very unlikely that all the simulations with the different schemes will suddenly re-converge to the same state after diverging from the initial state. The strong dependence of the tracer mixing ratios on the convection parameterisations is likely to remain. A test simulation which we performed in lower horizontal and vertical resolution, but over an extended period supports this statement; however, we think that it is better to focus on the high-resolution simulations rather than extending the paper further to include any details of the less reliable low-resolution results.

4) The interaction of chemically reactive compounds is not straightforward to analyse. In case of the non-reactive R_n the analysis is much more related to the dynamic quan-

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tities that can be extracted from the model. But with chemical reactions involved, the effective turn-over rates must be taken into account as well. Consequently, in a complex chemical setup such information is not readily accessible anymore, even though attempts are undertaken to gather such information. As the convective dynamics and the impacts on inert tracers with constant lifetimes have been investigated in previous studies with single column models (e.g., Mahowald et al, 1995), it has been more important for us to look at the overall picture, even though in that case not all of the reasons could be revealed any more.

5) Section 3 will be reformulated completely for the revised manuscript. Some of the details are likely to be moved into the supplement, whereas we attempt to provide more precise information in the main manuscript .

Minor comments:

1) The lightning distribution is only shortly mentioned, since the impact of the chosen convection scheme on lightning has been described earlier (Tost et al., ACP, 2007, Lightning and convection parameterisations – uncertainties in global modelling). As stated in the manuscript, the scheme following Price and Rind (i.e., a scheme that is based on the convective cloud top height) has been chosen (and adjusted to achieve globally the observed flash frequency), since it turned out to be mostly robust against an exchange of the convection scheme. Nevertheless, there are differences in the lightning NO_x emission distribution and to a small degree also in the total amount. This has some impact on upper tropospheric NO, NO₂ and consequently HNO₃, but only a minor impact on the other compounds.

2) The large-scale condensation (=cloud) and radiation schemes are listed and referenced for further detail in Table1. The boundary layer scheme is the one that is native to ECHAM5, and therefore a description of that can be found in the ECHAM5 documentation, which is also referenced in the manuscript. All applied convection parameterisation schemes take both, shallow and deep convection into account.

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3) We meant: “global modelling of atmospheric chemistry” which has a slightly different focus, and other resource requirements than numerical weather prediction. This will be reformulated.

4) The entrainment is hidden in the terms C_{kud} and C_{kdd} , respectively. Since the mass fluxes are determined by entrainment and detrainment according to Eq. 1, the updraft and downdraft mixing ratios in Eq.2 are also influenced by entrainment.

5) The first term describes the rest of the air mass, which has neither been impacted directly by the updraft, nor by the downdraft nor by the subsidence. Consequently, this has been named “as not affected by convection” but is better rephrased to “not directly affected by convection”.

6) The tuning has been applied by several simulations (without chemistry) changing the parameters and analysing the response of precipitation and OLR. Since no mathematical or detailed statistical analysis technique has been applied, it was a “tuning by hand”. Per convection scheme around 10 simulations have been performed, until a satisfactory agreement with the observed data has been achieved.

7) The consequent use of the abbreviations will be checked carefully for the revised version. In the ACP format the table will appear in the middle of the text (in the model description section), to make it easier to see them right at the beginning of the manuscript.

8) The Hack scheme is only run in conjunction with the Zhang-McFarlane scheme, as for instance applied in the MATCH model. The additions of Wilcox refer only to the Zhang-McFarlane part; consequently the abbreviation should indicate this modification. This has been explained in detail in the Tost et al. (2006) paper, but will be repeated here for better understanding of the current manuscript.

9) As also mentioned above (answer the comments of Referee#2), this has been a mistake and we are glad that we have been pointed to it before the finalisation of the manuscript for ACP.

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10) Our study in 2006 indicated that with this scheme a drier atmosphere (compared to integrated water vapour columns from satellite observations) has been simulated, and the tuning did not help much with that. The sentence will be reformulated.

11) We disagree that it is worthwhile to repeat this information in the main paper, but are willing to provide a table for the precipitation distributions in the supplement.

12) We checked the availability of OH reaction partners. Concentrations of several compounds used for OH reactivity measurements/calculations have been added (weighted on their reaction rates). Even though this is not a proof, it supports the stated hypothesis, however.

13) The term “exemplary species” was chosen, since instead of HCHO another compound with similar gas phase reactivity and solubility could have been shown. Since for HCHO more observational is available from at least one of the campaigns, this compound has been selected. HCHO is more reactive than CO (for chemical reactions of HCHO considered, see above in the replies to Referee#2) and has medium solubility.

14) All convection schemes create feedbacks on the large-scale circulation via the hydrological cycle. That has been mentioned in the model setup section. The nudging towards ECMWF analysis data “pushes” all simulations to the same observed state of the atmosphere on the synoptic and larger scales, but nevertheless there are some differences in meteorology between the simulations.

15) This will be altered into “southern hemisphere”.

16) Since the Section3 will be rewritten, this paragraph will also be reformulated.

17) The overall lifetime for nitrate and sulphate will be difficult to estimate, since the determination of the total production rate for both compounds is not straight-forward, because they are chemically produced, and furthermore for nitric acid a fraction is recycled with NO_x. If “lifetime = burden / deposition” is meant, this information can be provided in the revised version/supplement.

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18) For highly soluble compounds the scavenging completely counteracts the vertical transport, i.e., in case of a convective event almost all nitrate and sulphate will be wet deposited from the atmosphere. If a certain scheme does not trigger convection, often the large-scale cloud scheme produces a little amount of precipitation instead, which can also efficiently remove the nitrate and sulphate. This will be reformulated.

19) We agree. Decreasing condensed water by evaporation increases the specific humidity and the moist static energy. In this case, we referred to a decrease in condensed water by formation of precipitation and subsequent removal of condensed water from the atmosphere. Consequently this water cannot re-evaporate in the atmosphere any more and does not contribute to the moist static energy any longer.

20) We will reformulate this sentence. Since it is specific to the discussed topic of tuning, we want to keep it in this section. The motivation for this study has of course not been based on that statement, but it provides an extra aspect.

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