**Geoscientific
Model Development
Discussions**

Interactive comment on "Quantifying atmospheric transport, chemistry, and mixing using a new trajectory-box model and a global atmospheric-chemistry GCM" by H. Riede et al.

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Dear referees, dear Editor,

we address the various comments, suggestions, and questions in one single answer to establish the necessary links among them where necessary.

There are five sections of answers, METHOD (M), MODEL SETUP (S), EXTENSION/MODIFICATION OF PARTS (E), TERMINOLOGY/TECHNICAL (T), and FIGURES (F). R1, R2, R3 refers to a question or comment from the respective referee. A is the co-authors' answer.

1 METHOD (M)

M01

R2: "simultaneous implementation of a Lagrangian chemical transport model along with with an Eulerian chemical transport model and an atmospheric chemistry GCM"

A: As described in Abstract and Introduction, the models involved in the presented quantification method consist of 1) a trajectory-box model (CAABA/MJT), i.e., an atmospheric-chemistry box model in which initial and boundary conditions from a trajectory can be prescribed, 2) an atmospheric trajectory model (LAGRANTO), and 3) an atmospheric-chemistry GCM (EMAC).

M02

R3: "justification for using the trajectory model needed"

R1: "What is better in this method compared to applying atmospheric trajectories along model output of mass diagnostics from a 3D model?"

A: With our model approach, an offline analysis of processes in a global GCM is possible through a consistent trajectory-box model. The model hierarchy and its models are described in the Abstract and in the Introduction. The method presented was developed to quantify contributions of chemistry, transport, and mixing to an observed mixing ratio, e. g., measured on-board an aircraft. Without the trajectory-box model we could not as easily separate these contributions along the trajectories leading to the measurement site because we would have to run the full GCM again and write out concentration tendencies for each species for each process and reaction at each time step at each grid point closest to the trajectory paths. With the GCM-based trajectories and the trajectory-box model, in contrast, we focus on the relevant portions of space and time, i. e. the air masses that will eventually reach the measurement sites. With its help, we make information about the GCM accessible a posteriori and thus save data

volume and time (see M03).

M03

R2: "What is meant by time-efficient?", page 469, line 13

A: Time-efficiency here refers to the advantage of not having to run the complete GCM again for the quantification (see also M02). A trajectory-box model calculation takes about half a minute to a minute for five days. To run the full GCM for the same period of time on one CPU takes in a very rough estimate about 7 days (1 year of simulation on 256 processors takes about 2 days), not accounting for additional diagnostic rate constant output for each time step. Using the GCM data offline instead of re-simulating with operator-splitting diagnostics thus saves - by this rough estimate - more than 99.99% of time.

M04

R3: "Undisturbed transport", "the quantification", "theoretical contribution of transport", Section 2

R3: "quantification result", Section 3

A: The simplest approach for campaign data analysis is trying to relate, for instance, high ozone levels in the upper troposphere with backward trajectories from the stratosphere. This qualitative approach, however, does not reveal what happened along the way. With our quantitative method, we supply additional information about contributions of various processes and where they take effect.

With "the quantification", we mean our method as described throughout the manuscript, which yields quantitative influences of transport, chemistry, and mixing as its result.

"Undisturbed transport" means transport alone without chemistry or mixing as if all species were inert and contained in isolated air parcels. Quantitatively, this is defined by the mixing ratio at the starting point of the trajectory.

"Theoretical" in this context refers to the fact that in reality there usually is no undisturbed transport, but a mixture of contributions from other processes as well. The term "theoretical" will be omitted, since all contributions within our method are "theoretical" in that sense, not just the transport.

M05

R2: "tone down novelty claims, see Manonom, Xiaoen, Wongwises, A new Lagrangian-Eulerian coupling model system, Advances in Atmospheric Sciences, 2007."

R3: "on-top analysis, what does that mean?", Section 7

A: In contrast to Manonom et al. (2007), we couple a Lagrangian to a Eulerian model system offline for process analysis and sensitivity studies. All of the three models we use for our method presented here are stand-alone models that do not interact during runtime. We use the trajectory-box model CAABA/MJT with the help of the trajectory model LAGRANTO to interpret the results of the GCM EMAC.

M06

R2: "How about integrated process analysis in CMAQ?"

A: In the CMAQ integrated process analysis, the process tendencies at a stationary observatory are analysed in the Eulerian framework revealing the regional contributions (from neighbouring grid boxes).

In our approach, we follow the air-flow in the Lagrangian frame of reference towards an observational point and quantify contributions of transport, chemistry, and mixing along this trajectory revealing the complete history of an air mass - time step by time step or integrated over time.

M07

R2: "Why is the chemical analysis only applicable to slow species?", page 466, line 23

R2: "'more sophisticated approaches, such as presented in Lehmann (2004) are required' too vague", page 466, line 25

A: In principle, the contributions of various chemical equations in the chemical mechanism to mixing ratios of species are straightforward to obtain. This is done in CMAQ and in CAABA/MJT. The reaction rates and stoichiometric coefficients written out at each time step provide just that information. This is true for both, species involved (e. g., ozone) or not involved (e.g., carbon monoxide) in catalytic cycles. However, chemical source and sink terms of species taking part in catalytic cycling present huge contributions for production and destruction with respect to just these cycling reactions with maybe only a small net change in mixing ratio. The reactions causing the small net change "drown" in comparatively huge numbers of continuous creation and destruction. But those are the ones especially important for further investigation regarding lifetimes on a larger scale (ensemble lifetime or chemical residence time), in contrast to the single-molecule lifetime. The method presented by Lehmann 2004 provides the possibility of analysing chemical reaction pathways including chemical reaction cycles and neglecting these fast cycles if desired, for example for calculating the ensemble lifetime time of ozone, for which burden divided by loss would just give the immensely shorter molecular lifetime. We will expand the text of the revised manuscript accordingly.

M08

R1: "How about the secondary effect of mixing on chemistry?", page 480, line 10

A: We consider this secondary effect as mixing influence, since that part of chemistry could not have happened without prior mixing. That implies that we might obtain mixing contributions for such short-lived species as OH, which might seem unreasonable. But since we know how short-lived OH is, we instantly know that OH mixing really is OH chemistry taking place as a consequence of mixing. Therefore, OH and other very short-lived species can serve as an indicator of mixing induced chemistry. This can be confirmed and extended to longer-lived species by reducing the backward time period

of the trajectory to which our method is applied. When the method is started at a later point in time after mixing has occurred, the chemistry contribution that once was secondary can now be observed as primary chemistry effect for short-lived and long-lived species.

M09

R1: "What are external J-rates, how are J-rates derived in the model?" "role of cloud fraction and cloud water content set to zero", page 461, line 29

A: We stress that the box model CAABA/MJT and the GCM EMAC share the same submodel for photolysis rate calculations, namely the submodel JVAL which is based on the approach of Landgraf and Crutzen (J. Landgraf and P. J. Crutzen, An Efficient Method for Online Calculations of Photolysis and Heating Rates, J. Atmos. Sci., 55 (5), 863-878, 1998.)

In our approach, photolysis rates are sampled from the GCM EMAC in order to increase the consistency between the GCM and the trajectory-box model. Cloud effects, for instance, cannot be resolved by the trajectory-box model. For simplicity, only the photolysis rate for nitrogen dioxide (NO2) is sampled in the present setup. By comparison between this "external" photolysis rate and the JVAL photolysis rate, a correction factor is calculated, with which all JVAL photolysis rates are scaled.

We had decided not to focus too much on this procedure since it represents no method-inherent limitation. It is chosen for simplicity of current calculations. This procedure using sampled photolysis rates for NO2 can be extended to all other species for which GCM photolysis data is available. These can then be decoupled from the above correction based on NO2.

M10

R2: "mixing is only defined within the model hierarchy", page 466, line 14

A: The contribution of mixing is only defined within the model hierarchy since it is de-

fined as a difference term between two very consistent models. Thus, an assessment as done for chemistry in Fig. 10 cannot be conducted for mixing.

M11

R2: "transferability to observations"/"transfer of results"

A: As pointed out in the first paragraph of Section 6 (Discussion), there are two types of consistency which are discussed. The first type refers to the consistency between the models in the model hierarchy (trajectory box model CAABA/MJT and the atmospheric-chemistry GCM EMAC). Within this hierarchy, the quantification into contributions of transport, chemistry, and mixing is always defined (within the limitations presented in the publication). The second type of consistency refers to "the transferability of the model-based analysis to observations".

Whereas the offline analysis of processes in the atmospheric-chemistry GCM by means of the trajectory-box model is always applicable, the assumption that these results hold true as explanation for observational data is not straightforward. Therefore, it is stressed that the transfer of these findings to observational data needs at least the comparison of data from both the GCM and observations at the point of observation, and comparison of trajectories based on wind fields from both EMAC and ECMWF.

M12

R2: "How about adjoint or DDM sensitivity analysis?" (A. Sandu, D. N. Daescu, and G. R. Carmichael, Direct and adjoint sensitivity analysis of chemical kinetic systems with KPP: Part I - theory and software tools, 2003; A. Sandu, D. N. Daescu, G. R. Carmichael, and T. Chai, Adjoint sensitivity analysis of regional air quality models, 2005.)

A: These methods can certainly be combined with our method presented here in a similar way as the method of Lehmann (2004). This is, however, beyond the scope of this publication.

C175

M13

R2: "How about using inert tracers?"

A: Due to different spatial gradients for every tracer, which depend for instance on the the source distribution, the derived transport and mixing contributions of an inert tracer would be very specific for that inert tracer and could not be generalised to all tracers. For the transport contribution, each tracer is assumed to be its own inert and isolated tracer (see answer to M04).

2   MODEL SETUP (S)

S01

R3: "How many reactions? Was OH solved or kept constant?"

A: The mechanism used for the present example includes 385 gas phase species, 180 gas phase reactions, and 60 photolysis reactions. OH was calculated prognostically.

S02

R2: "Why is sedimentation and deposition switched off?", page 461

A: Sedimentation and deposition are implemented and available for the box model CAABA. We decided to switch off sedimentation and deposition for this approach in order to separate the kinetics from other processes. In that way we can ask the questions: "What would we obtain at a measurement site if tracers were just advected for the last five days?" (trajectory model), "What if we also allow chemistry to take place?" (trajectory-box model), and finally "What if we additionally allow physical processes other than advection?" (GCM).

S03

R2: "Why is aerosol chemistry switched off?"

A: In contrast to sedimentation and deposition, aerosol chemistry is not switched off for the benefit of the method but in this case for simplicity of the example. It is not necessary for presenting the method. We mentioned it to correctly describe the model setup. In the revised version, we will phrase that in a clearer way. Please note that in the trajectory-box model setup being consistent with the GCM is of highest priority. Sensitivity studies with an altered (more comprehensive) chemistry including aerosol chemistry can be conducted, but then - analogous to simulations along ECMWF trajectories - mixing is undefined.

S04

R2: "Why is heterogeneous chemistry switched off?"

A: Here, the same as for aerosol chemistry is true. Heterogenous chemistry is not crucial for showing the principle in an application. It would have required to sample more trajectory information from EMAC grid datasets for the calculation of heterogeneous reaction rates without illustrating an important additional point in the current context. In further application studies it will be taken into careful consideration.

S05

R2: Why is cloud fraction and cloud water content set to zero?

A: The answer is similar to the one about heterogeneous chemistry. The additional sampling of these trajectory boundary conditions from EMAC 3-D data fields is possible, but not necessary for the illustration of the presented method.

3   EXTENSION/MODIFICATION OF PARTS (E)

E01

R3: "Paragraph 3 of Section 5.2 needs a citation"

A: The following citation will be inserted: P. M. Barbosa, D. Stroppiana, and J.-M. Gr'egoir, An assessment of vegetation fire in Africa (1981-1991): Burned areas, burned biomass, and atmospheric emissions, Global Biogeochemical Cycles, 13 (4), 933-950, 1999.

E02

R2: "'all of these models have been designed for their own special purpose' too vague", page 457, line 10

A: We will remove this sentence.

E03

R2: "move introductory discussion of the statistical approach currently presented in Section 3 to Introduction"

A: The introductory part will be shifted to the Introduction in order to provide the full overview at the beginning.

E04

R3: "Heading of section 3 too complicated"

A: There are two separate things mentioned in the title, firstly the statistical basis for the assessment of results (picking representative trajectories) and secondly the transferability issue ("We quantified in the model world - Can we apply it to real observations?"). We will split the Section into two Subsections for clarity.

E05

R3: "You don't need to mention N_2 and O_2", Section 4

A: We will remove that for clarity.

E06

R3: "biomass burning activities in boreal winter → DJF"

A: "Boreal winter" is a well-defined, widely-used term. We prefer that to explaining the acronym.

E07

R3: "only meteorology compared to observations, how about species' concentrations? There is so much CO data, please include some comparison"

A: The focus of the present publication is the method, not the results. Since we are aware that examples help to understand processes, we decided to include a very basic example of what can be obtained with the method. The pictures we could provide about this one example in terms of different species, process analysis, and various periods of analysis from single time steps to whole trajectories etc. would fill several pages, so we chose a small story to show what the method can show without claiming a full scientific application.

A thorough comparison between observational data along flight tracks and GCM data as well as the application of the method described here is in preparation for a journal not focusing on model development. A single point-to-point comparison between the model and observational data at the end of the sample trajectory is not very relevant and to find observations exactly along one or more Lagrangian trajectories proves impossible. If those data were available, we would not need our method.

4 TERMINOLOGY/TECHNICAL (T)

T01

R3: "getting lost in acronyms"

A: In the revised version, we will include a table with models and their acronyms to complement Fig. 1.

T02

R2: "explain submodels and base models", page 457, line 24

A: These are terminology of the MESSy standard (Jöckel et al., 2005). The base model is the pace maker of the whole model system. It ideally consists of just a runtime clock, but at the moment of a GCM (ECHAM5). Submodels represent different Earth system processes and they can be switched on or off independently. In one application, for example, several convection schemes were implemented (submodel CONVECT) and were switched on and off without changing the rest of the model setup (Tost et al. 2007, ACP).

T03

R3: "What is a waypoint?", Section 4, page 462, line 12

R2: "individual waypoints", "requirement for equidistance", page 462, line 12

A: A waypoint is one of the points along the trajectory. The waypoints along with the accordingly discretised boundary conditions are passed to the trajectory-box model as a part of the input. A trajectory consists of a number of individual waypoints for each of which boundary conditions are discretised. These waypoints do not need to occur in regular time steps or in regular space intervals.

T04

R2: "quantification analysis"

A: On page 457, line 1 after "model hierarchy", ", hereafter referred to as quantification analysis." will be inserted.

T05

R2: "pure chemistry", page 458, line 24

A: Meant is "undisturbed chemistry" (= kinetics), neglecting all secondary effects on chemistry by physical processes such as mixing.

T06

R2: "uncertainty concerning chemistry: errors in rate constants, missing reactions, numerical errors, resolution artifacts?", page 459, line 10

A: This section is referring to uncertainties contained in the contribution of chemistry arising from possibly different transport pathways, which in turn cause different tracer initialisations.

T07

R2: "frayed-out start", page 465, line 1

A: Wrong term, will be replaced by "non-coherent", "spread-out" or similar.

T08

R2: "dynamic situation -> local meteorology?"

A: Since it affects a whole region, we will change it to "regional atmospheric dynamics" or "regional wind patterns described by the trajectories"

T09

R2: "keeping its statistical weight"

A: The statistical weight is used later to analyse how representative our results are for certain transport pathways.

T10

R2: "38, 15, 9, 40: Why the reversal of trend?", page 464, line 27

A: The data series refers to relative percent change. In the example, the convergence of trajectories on their five-day travel leads to fewer and fewer trajectories needed to represent the overall movement of air masses reaching the site of observation.

The respective numbers of representative trajectories for the considered time interval of observations are: 21 (5 days back), 13 (4 days back), 11 (3 days back), 10 (2 days back), 6 (1 day back).

Thus, (13-21)/21 = -38%, (11-13)/13 = -15%, (10-11)/11 = -9%, (6-10)/10 = -40%.

To prevent misunderstanding, we will mention both relative percent change and absolute percent change. The relative percent change is especially useful for analysing the temporal development of atmospheric dynamics in a selected region (see text immediately after the numbers in the manuscript).

After a re-assessment of the numbers, there are two minor changes as follows: 10 (3 days back), 9 (2 days back). Thus, the updated relative and absolute percent changes reducing the backward travel time by full days starting from five days are:

Relative percent change: (13-21)/21 = -38%, (10-13)/13 = -23%, (9-10)/10 = -10%, (6-9)/9 = -33%.

Absolute percent change: -38% (62% left for the last 4 days of travel), -52% (48% left for the last 3 days), -57% (43% left for the last 2 days), -71% (29% left for the last day).

T11

R3: "transfer of quantification results to observations: you mean model-data agreement?" Section 7

A: Model-data agreement is a prerequisite for the transferability of model results to observations.

5 FIGURES (F)

F01

R3: "Are CAABA-MJT and CAABA/MJT the same thing?", Figure 1

A: Yes, will be corrected.

F02

R3: "Why no overlap between integration points and trajectory points?", Figure 4

A: Since we wanted to keep the figure general and since they do not necessarily coincide, we chose not to let them coincide. The trajectory points come from an external source (trajectory model) as input to the trajectory-box model, we simply don't expect them to have the same time steps as the trajectory-box model. Because of the diurnal cycle and the linear interpolation between trajectory waypoints, we cannot trust that the waypoints are close enough in time, they could be for example 2 hours apart due to the trajectory model. Therefore, the trajectory-box model time step is not changed, except if a trajectory waypoint is closer than the next integration point, in which case shorter time steps are inserted around the trajectory waypoint.

F03

R3: "confusing, what are trajectory points, what are waypoints?", Figure 5

A: There are only 2 types of points: 1) integration points, the ones generated by the steady time stepping of the trajectory-box model (shown in blue), and 2) the ones provided by the external trajectory input, shown in orange. These are called trajectory waypoints, trajectory points, or waypoints. They stand in contrast with the integration points. In order to be consistent "way" will be inserted wherever the point is missing it.

Thank you very much for asking the right questions in order to help me present my work in a clearer way. We will consider all of the points hindering a clear comprehension

discussed above and make changes accordingly.

Sincerely, Hella Riede on behalf of all co-authors

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