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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Using beryllium-7 to assess cross-tropopause transport in global models" *by* H. Liu et al.

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We thank Referee #1 for detailed and constructive comments. Our responses are itemized below.

"GENERAL COMMENTS: Liu et al. study the impact of various meteorological data sets used in the Global Modeling Initiative (GMI) chemical transport model (CTM). In particular, they use a beryllium like aerosol tracer to assess stratosphere-troposphere exchange (STE) in the different meteorological fields. Since it is well known that all data fields have issues in realistically representing this process, constraints on STE in the model were made based on surface concentration measurements as well as on observed deposition fluxes of beryllium 7 (7Be). Moreover, the model results were

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compared with airborne measurements of 7Be and ozone in the upper troposphere and lower stratosphere. The observational constraints were further applied to other model simulations with the GFDL AM2 and GEOS-Chem, finally leading to the conclusion that these constraints result in a more reliable STE flux assessment."

"Although research is conducted for a long time, STE is still a topic of intense debate. To assess the strength and location of STE properly is important for both air pollution studies as well as climate projections. Comparing available meteorological fields and assessing the strength of the errors of STE as well as the physical reason for these errors is a valuable contribution to further improve the quality of meteorological data sets. The paper is generally written in an understandable way, however, sometimes with the tendency to be a bit too descriptive. This finally results in a relatively long paper. The figures and tables are generally of good quality and presented along the discussion in the text. The applied methods are sound and the conclusions follow the discussion in the sections before. Overall, I support the publication of this study in ACP, after the following comments are addressed."

Reply – Thanks for the comments. Following Referee #2's suggestion, this paper has been shortened.

"DETAILED COMMENTS: 1) The introduction could benefit from some updated references. For instance, the paper from Riese et al. (2012), JGR, which shows the importance of assessing the concentrations of certain trace species in the UTLS (page 26134, line 8). The study of Skerlak et al (2014) presents climatology of STE and numbers on STE mass fluxes over the entire ERA-INTERIM time period (see paragraph on STE starting on page 26136, line 21). Maybe recent model comparison (from CCMI, ACCMIP, CMIP5) could also give further numbers."

Reply – We have included in the text the following references: Riese et al. (2012), Skerlak et al. (2014), and Young et al. (2013). We now state in the Introduction section that "Though correct representation of STE is essential for simulating 7Be, ozone and

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other trace species in the troposphere (e.g., Riese et al., 2012), large variations exist among models." and "Observation-based estimates of STE fluxes of ozone into the troposphere are typically in the range of 400-600 Tg/year (Murphy and Fahey, 1994). Some global models are able to produce STE fluxes of ozone in this range (e.g., Olsen et al., 2004; Hsu et al., 2005; Hsu and Prather, 2009; Lin et al., 2012; Young et al., 2013; Skerlak et al., 2014)." Reference: Young, P. J., et al.: Pre-industrial to end 21st century projections of tropospheric ozone from the Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP), Atmos. Chem. Phys., 13, 2063-2090, doi:10.5194/acp-13-2063-2013, 2013."

"2) Regarding the model (section 2.1), what does "nearly full chemical treatment" mean? Could you specify the chemical mechanism a bit more in detail (NOx-VOC-O3, sulfur?). Furthermore, why are the aerosol and chemistry simulations conducted independently? "

Reply – We have revised the text to "The GMI (http://gmi.gsfc.nasa.gov) CTM is a global 3-D composition model that includes a full treatment of both stratospheric and tropospheric photochemical and physical processes. It uses a 114-species chemical mechanism that combines the stratospheric mechanism of Douglass et al. (2004) with the tropospheric mechanism of Bey et al. (2001). The chemical mechanism includes both stratospheric and tropospheric heterogeneous reactions. Tropospheric aerosol (sulfate, dust, sea salt, organic carbon, and black carbon) fields are taken from the Goddard Chemistry, Aerosol, Radiation and Transport model (GOCART). Details of the model are described in Duncan et al. (2007, 2008), Strahan et al. (2007), and Considine et al. (2008)." Note that for the GMI full-chemistry simulation, tropospheric heterogeneous reactions and aerosol photochemical effects use monthly tropospheric aerosol fields from the GOCART model. For the radionuclide aerosol tracer (7Be) simulation, it does not involve interactions with chemistry and can therefore be conducted independent of full-chemistry simulations.

"3) The horizontal resolution of the model simulation with 4°x5° is very coarse (section

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2.1), especially since it is known that STE is very sensitive to the model resolution both in the vertical and horizontal. Is it possible to include at least one further simulation with one meteorological data set with an increased resolution and to discuss the differences? Is STE still overestimated in a high-resolution simulation? And consequently, is there a "threshold" resolution at which the observations are met satisfactory without applying further constraints. Also in section 6, wouldn't it be more sensible to use a higher vertical resolution in the tropopause region in the AM2 simulations? Moreover, did I understand it right that always only full levels are either stratospheric or tropospheric or is there also an interpolation applied between two model (interface) layers?"

Reply – (1). We discussed the differences in STE of 7Be between a coarse resolution run ($4^\circ \times 5^\circ$) and an increased resolution run ($2^\circ \times 2.5^\circ$) in a previous study (Liu et al., 2001). As we stated in the text "The simulations presented here use a degraded horizontal resolution ($4^\circ \times 5^\circ$) for computational expediency. Degraded horizontal resolution slightly increases cross-tropopause transport (Liu et al., 2001). Nevertheless, our objective is to assess cross-tropopause transport in meteorological data sets at the resolution used to drive the model, not necessarily at the original or finer resolution." (2). We feel that finding a "threshold" resolution is beyond the scope of this paper. (3). Following Referee #2's suggestion, section 6 is now eliminated. (4). Yes, always only full levels are either stratospheric or tropospheric. There is no interpolation applied between two model layers.

"4) Could you please mention which advection scheme you are using and provide a reference? (page 26140, line 5)"

Reply – The reference is now added: "The model uses the flux-form semi-Lagrangian advection scheme of Lin and Rood (1996). . .. . .". Reference: Lin, S. J. and Rood, R. B.: Multidimensional flux-form semi-Lagrangian transport schemes, Mon. Weather Rev., 124(9), 2046-2070, 1996.

"5) Beryllium attaches to the ambient aerosol and then it is treated as the aerosol. In

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the model the bulk mass is tracked. Is there an aerosol climatology provided to the model to assess the ambient aerosol. Or is the beryllium treated as a quasi-passive tracer with a fixed modal representation? If so, then it would be sensitive to give some additional meta information, such as molar mass (important for the dry deposition) and radius and sigma of the aerosol (usually important for the scavenging efficiency). Submicron particles can still have quite different scavenging efficiencies."

Reply – We state in the Introduction section that "Beryllium-7 ..... After production, it attaches immediately to ubiquitous submicron aerosols in the ambient air. The fate of 7Be then becomes that of those aerosols, which move with the air until scavenged by precipitation or deposited to the surface." 7Be is treated as a quasi-passive tracer, and the mass of 7Be (not the mass of the ambiennt aerosol) is tracked. The molecular weight of 7Be is 7 kg / kmole. Our scavenging scheme (Liu et al., 2001) does not take into account the submicron aerosol size-dependency of scavenging efficiencies.

"6) In section 2.2, it is mentioned that clouds, precipitation as well as convective transport are mainly responsible to obtain a good distribution of 7Be. However, for STE often the strength and location of the subtropical jet is also crucial where quasi-isentropic exchange between tropospheric and stratospheric air masses can occur. Maybe it is worth thinking about to include a comparison of the location and strength of the jets between the different meteorological fields. The comparison in section 4 would also benefit from such a discussion. Is it possible to assess the error in the model deposition fluxes resulting from the over- and underestimation of precipitation? (page 26141, line 17 - 20)"

Reply – This is a good point. We will include a comparison of the location and strength of the subtropical jet and discuss it in section 2.2 and section 4. It is difficult to assess the error in the model deposition fluxes due to the bias in the model precipitation because such assessment would require information on the observed vertical distribution of precipitation.

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"7) In section 2.3: Following comment 5), how is the beryllium introduced into the model? Is it distributed evenly over the globe? Are there hot spots? Does the stratospheric aerosol have different parameters (size, sigma, etc.) than the aerosols in the upper troposphere? Moreover, and please correct me if I am wrong, but do I get it right that you use a constant 7Be production rate, which is, however, not so constant in reality (since it depends on the solar cycle). Thus, the constraints from the observations are based on a variable source strength, however, the model results are based on a constant source strength. Is it somehow possible to discuss this potential error between observations and model results a bit more in detail?"

Reply – See our reply to comment 5) above. Now we state in section 2.3 that "The LP67 source is represented as a function of latitude and altitude (pressure) and does not vary with season (see Figure 1 of Koch et al., 1996)". We also state at the beginning of section 2.5 that "We estimate an average solar year value simply by averaging the long-term records of 7Be observations multiplied by 0.72 to correct to the 1958 solar maximum source (Koch et al., 1996)."

"8) Section 2.4 is difficult to read. However, the main points are not that difficult to understand. Maybe think about reducing the current content slightly and highlight the main points a bit more."

Reply – This section has been shortened, as also suggested by Referee #2.

"9) In section 5 a comparison with other modeling studies is presented. It would also be helpful to provide some further numbers from studies of STE. Potentially, studies analyzing reanalysis data sets could be included, such as Skerlak et al. (2014). However, this could also be part of the introduction (see first comment)."

Reply – See our reply to comment 1) above.

"10) At the end of section 6 changes between results from model simulation are discussed. The differences are attributed to the various GEOS-DAS versions. Could you

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potentially list the major changes between the various data sets. Is this related to finer native resolution, different physical parameterizations, etc.? This might further help to understand the changes in the beryllium results. Why is cross tropopause transport slower in GEOS3-DAS? Is the jet structure different? Does the location of the tropopause vary strongly? Have there been significantly changes in the assimilation cycle?"

Reply – Following Referee #2's suggestion, we have eliminated section 6.

"11) Why is the thermal tropopause used to calculate the tropospheric TOC in section 7? Would it not be more sensible to use the dynamical tropopause (based on a potential vorticity threshold, eg. 2 pvu). The temperature lapse rate tropopause is often not assumed to be the best marker between stratosphere and troposphere, since there are situations where the thermal tropopause is not so well defined."

Reply – Point is well taken. On the other hand, we discuss in section 4 about the effect of different definitions of the tropopause by citing the work of Stajner et al. (2008): "Stajner et al. (2008) used four different definitions of the tropopause on the basis of temperature lapse rate (World Meteorological Organization or WMO definition), potential vorticity (PV), and isentropic surfaces or ozone surfaces. They found that the WMO tropopause was about 0.7-1 km (in the northern mid-latitude) or 0.5-1 km (in the tropics) higher than the ozone or PV determined tropopause." The temperature lapse rate tropopause used here should not affect the conclusion of this study.

"12) The discussion has often a quite descriptive character. An example is the second and third paragraph in section 4. STE is compared based on surface measurements of 7Be. It is mainly stated that there are differences between the model simulations but not what causes the differences, which are the main driving processes that cause the differences. I think the manuscript would benefit significantly, if such additional information could be provided directly."

Reply – See our reply to comment 6) above.

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“13) What is meant with satisfactory representation in line 7 on page 26157? Is this based on a certain (small) deviation from the expectation?”

Reply – We have revised the sentence to “This suggests that models which utilize either of these fields could use the “Linoz” ozone scheme and expect reasonable representation of the stratospheric influence on tropospheric ozone on a global scale (i.e., STE fluxes of ozone within the range of observation-based estimates).”

“TECHNICAL COMMENTS:

1) page 26142, line 6 : LP67 is not specified explicitly; 2) page 26146, line 5 : tropopheric –> tropospheric; 3) page 26147, line 16: please define once SCM, either in the text or in a figure caption”

Reply – Done.

“4) page 26150, line 19: what is meant with standard simulation?”

Reply – It means the reference simulation where no adjustment of 7Be cross-tropopause fluxes was made. To avoid confusion, we have removed “standard”.

“5) page 26161, line 16: "to" is missing –> helping "to" reduce ?”

Reply – Both are OK.

“6) page 26136, line 19: verus –> versus”

Reply – Corrected.

“7) page 26150, line 10: middle latitudes –> mid-latitudes”

Reply – Changed.

“8) page 26158, line 4 : greatest –> largest”

Reply – Changed.

“9) Figure 2: maybe the convective mass fluxes could be compared to a high resolution

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convective mass flux of one reanalysis data set to see how large the difference are caused by the reduced resolution."

Reply – We believe that this is beyond the main scope of this study.

"10) All colored latitude-height plots could benefit from a color bar (Figures 3, 6a, 7a, 9a, 10a, 11, 12)"

Reply – Thanks for this suggestion, which we will consider in the revision.

"11) Figures 11 and 12 could potentially be merged to one Figure 11 with two panels a,b, since they show the same content."

Reply – Following Referee #2's suggestion, we have removed Figures 11 and 12 and relevant discussions.

"12) In all line plots: at least in the printed version, the blue and purple lines are difficult to distinguish." Reply – We have replotted all figures to improve their quality.

"13) Caption of Figure 7 says 'Same as Fig. 4....', I think this should read 'Same as Fig. 6...'."

Reply – Corrected.

"14) Figure 15: a) and b) are mentioned in the caption but are not assigned to the two panels. Moreover, is it possible to add the zonal variability to ΔTOC and Δ7Be in panel a), maybe by using gray contours. And is it further possible to add error bars to the best fit line in panel b)?"

Reply – "a) and b)" are now added to the two panels. The zonal variabilities of ΔTOC and Δ7Be are added in panel a). We choose to show in the legend the errors of the best-fitting parameters.

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 26131, 2015.