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Comment on acp-2021-865

Anonymous Referee #1

Referee comment on "Age spectra and other transport diagnostics in the North American monsoon UTLS from SEAC^{4}RS in situ trace gas measurements" by Eric A. Ray et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-865-RC1, 2022

This paper presents a new method to deduce age spectra and related transport diagnostics from in situ observations. The method is based on recently published studies, but also includes several new modifications and improvements. Transport diagnostics, like age spectra with respect to tropical and North American surface source regions, path integrated trace gas lifetimes, and source region fractions are calculated from aircraft in situ tracer measurements from the SEAC4RS campaign in 2013 in North America. In particular, a high impact from local sources related to rapid convective transport is found up to about 380K potential temperature.

The calculation of these transport diagnostics directly from measurements in the UTLS is novel and very valuable for gaining insight into the short transport time scales of that region and for constraining model transport. I think this method has a high potential to be applied to other datasets and the paper will be of great interest to scientists focussing on both measurements and modelling and on troposphere and stratosphere. Hence, I highly recommend publication in ACP. The paper is overall well written and organized, and the results are clearly presented. A few minor and specific comments are given below. I leave it to the authors to decide whether these help further improving the paper.

Minor comment:

I sometimes had a somewhat hard time to follow the reasoning regarding the method. I see that there is a lot of theory behind and it is a balance to not overload the paper with equations and details but still present enough information to enable readers to follow. To make the method available and applicable to more than just a few experts, I suggest to spend some more work on improving the motivation and description of the method details (mainly Sect. 3 and 2). For example, after Eq. 1, the difficulties of applying this relation to measurements could be described, and that certain approximations are indeed necessary. Then, following Eq 3, the approximations used here (which are decribed then in the following) could be summarized.

One problem I had when reading was that it remained sometimes unclear to me what were assumptions to simplify/enable the calculation and what followed from the general theory. A few exemplary related specific comments are given below. As far as I can see, approximations used here include stationarity of age spectrum (e.g., no seasonality), the inverse Gaussian shaped 1D spectrum, usage of altitude z instead of tracer-based equivalent height, restriction to / choice of 2 surface source regions. It is clear to me that such approximations need to be done to make the method applicable to real data. I'd only suggest to clarify at several places what results from general theory (as e.g. provided by Holzer and Hall, 2000) and what are additional assumptions/approximations. As said above, a summary of these in Sect. 3 and maybe also an enhanced discussion (e.g., L720ff) could help the reader.

Specific comments:

L24: "... provide a range of transport diagnostics ...". I find this formulation somehow vague and suggest to be more specific - e.g. state which diagnostics explicitly.

L107: Perhaps better say "CO2 surface mixing ratios ..."

L126: As far as I can see, figure 3 is the first figure mentioned in the text. Perhaps change the order of figures - if this makes sense...

L194: If I understand the theory correctly, including time-dependence into the age spectrum via letting K be a function of t is not equivalent to considering a time-depending transport operator in the continuity equation. (The specific form of G in Eq. (2) is the Greens function only for stationary 1D diffusive transport). If I'm correct, I suggest to clarify this here and say that this is another assumption.

L200ff: Isn't this compact relationship equivalent to the relationship between scale height and lifetime found by Ehhalt et al. (2007, their Fig. 7)?

L212: "... do not assume steady state conditions ..." Is this true in general? I understand that the surface mixing ratio is allowed to depend on time, but that for the age spectrum still steady state conditions are assumed. (See also my comment above regarding L194). Please clarify.

L251: But this assumption neglects the pathway described in L230, that air from NA surface can be transported into the lower stratosphere via the tropical upper troposphere. Is there a sound reasoning why this pathway can be neglected, or is it just to make the computation feasible? In the same spirit, are there good reasons why transport from other

extratropical surface regions beside North America to the sampling region can be neglected?

L253: Related to the above comment, I'd suggest to write here something like "We assume that the age spectra can be partitioned as ...".

Eqns. 4/5: I don't understand the separation here into f and G functions. First, the partitioning into scaling factors and G I see as an empirical ansatz - is this correct? (If yes, I'd suggest to state that). And why are transport parameters from tropical and NH surface patches the same? Shouldn't the G's on the rhs of the equations actually be age spectra for the specific surface patches (e.g. as in Hauck et al., 2020, Eq. 12)? This would then also affect the rearrangement leading to Eq. 12. Maybe a few more words for explanation could be helpful, in the sense whether this ansatz follows from the general theory or is an assumption.

Eqns. 7/8: I don't fully get the meaning of these equations here? E.g., why is there a y_TR dependence on the lhs if the latitude dependence is integrated out? Or should this be y_pTR as on the rhs?

L275ff: Also here, regarding the steps leading to Eq. 11, it is not clear to me whether these follow from theory, or are further approximations to construct a surface mixing ratio time series indepedent of transit time which can be pulled out of the integral.

L480-L515: I find the description of the various scalings applied rather complicated, technical and confusing. Perhaps, these paragraphs could be moved to an appendix and just very briefly summarized here, to not distract the reader's focus?

L645: I think two other reasons which likely contribute to the differences between the here observed and recently published surface source region fractions are: 1) SEAC4RS focussed on sampling strong convection where the local source impact is likely much stronger than in the zonal mean; 2) The models used in previous studies likely underestimate the convective impact.

L737: "... primarily applies to the tropical source region age spectra ..." I'm not sure whether this can be stated. The studies by Hauck et al. (2020) and Yan et al. (2021) showed also very clear seasonality for extratropical source region age spectra, which in relative terms could be even clearer than for tropical spectra. If I'm correct, I'd suggest to just delete this part of the sentence.

L768: "... most significant source of air ..." In view of the presented results (e.g., Fig.

10c) I'd rather say "... a significant source ...". The NA fraction is indeed substantial, but on average below 50% (as far as I can see).

Figure S1: What is the meaning of the intensity/darkness of orange shading? Would be good to specify that in the caption.

Technical corrections:

L144: Isn't "extrapolated" instead of "interpolated" what you mean here?

L466: Isn't it "Eq. 3" which relates mu to G and should be referred to here?