Review of the paper:

"Age Spectra and Other Transport Diagnostics in the North American Monsoon UTLS from SEACRS In Situ Trace Gas Measurements"

written by Ray et al.

General:

This is a very interesting and important paper which is worth to be published. It presents a new method how to interpret in situ data observed in the vicinity of the UTLS during one campaign (SEACRS in 2013). By using the age-spectrum technique, the authors quantify: (1) the origin of the sources of the observed species in terms of the boundary layer fractions (tropics or North America), (2) the (path-integrated) lifetimes for the variety of the observed tracer gases, (3) the "outliers", i.e. observations which deviate from a "mean behavior" and are very likely signatures of "polluted" air. In addition, the K-parameter of the age spectrum, assumed here as a 1D spectrum (Hall and Plumb, 1994), was derived from the experimental data and, consequently the age of air of the observed air masses could be calculated. The presented paper can be considered as a pioneering work how to include 20 different species observed at two aircraft during few weeks of a campaign to quantify the transport pathways, timescales of transport, chemical composition of air on its way from the polluted boundary layer to the UTLS region. Although this a is well-written manuscript with almost perfectly designed figures, the formulation of the method is the weakest point of the manuscript and is my major critical point of this review.

Major point (divided into 3 sub-points):

- I read your paper 3 times to understand what you are doing. The major difficulty for me was to recognize that you iteratively determine the age spectrum G, starting at the Earth surface and than moving up, level by level. So if you write in line 126 "The surface measurements are convolved with the UTLS age spectra to derive integrated surface boundary conditions" you just killed the reader. You should first introduce equation (1) and (2), boundary conditions, etc, and then introduce your iteration procedure. "To derive integrated surface boundary condition" makes sense for me only within the iteration loop. Maybe a schematic figure would help (like Fig 3 for your assumptions (4) and (5)). Fig 5 is a very good example for the procedure at one level but not for the iteration connecting level n with level $n + 1$.

- I would reserve the word "convolution" only for the equation (1). Equations (7) and (8) are much more "smoother" of the lower boundary and more technical if compared with the main convolution (1)

- The 1d spectrum (Hall and Plumb, 1994) is a very strong simplification. I would state it more clearly at few places like by introducing eq (4) and (5) which are, from my point of view, one of the "smartest ideas" of the paper.

Minor points:

- L21: "path-integrated lifetimes" - in my opinion, the partitioning between the NMA and tropical origin is also very important

- L25: “can be compared with chemistry-transport model” - the value of K being a combination of vertical advection and diffusivity is rather difficult to compare...here a better approximation of the 1d age spectrum function would be better but I know that this is not so easy to get an appropriate analytical solution.
- L47: I would not introduce “global lifetimes” which do not play any role for this paper
- general: path-integrated lifetimes - it is not clear what we can win from this concept...better approximation for a ”true” lifetime of chemically active species?...would be nice to get more motivation for this concept in the introduction.
- L125: “The surface measurements are convolved with the UTLS age spectra to derive integrated surface boundary conditions for each trace gas”

  ...to derive source latitude distributions (like in Fig 6c), I would concentrate in this first subsection only on the boundary conditions and separate their introduction from the age-spectrum dependent quantities. For the “integrated surface boundary condition” you need something like eq (1). At this stage this equation is not known to the reader, see may major point
- L129: “...so the inclusion of older spectra times does not significantly change the derived surface boundary conditions.”

  ...once again, boundary conditions are independent on the age spectrum...however, to calculate age spectrum, boundary conditions in the past (up to 30 years) have to be known

  I think, you mix here two concepts: boundary condition and age spectrum. For this, you need equation (1) that is not present for the reader at this stage of explanation. This is also little bit related to you iteration procedure to derive the age spectrum. I would recommend to introduce these concepts step-by-step: (a) boundary conditions, (b) age spectrum (c) convolution of the boundary condition and age spectrum, i.e. eq.(1) (d) iteration procedure
- L205-210: I would prefer to use $\mu(\tau)$ instead of $\mu - \tau$. Same for μ^*
- L275-280: Eq. (7) and (8): I would not call it “convolution” but much more “smoothing” with a Gaussian smoother. Eqs (9) and (10) should be much more convolutions. The dependence on y_{TR} in (7) and (8) is not present on the right hand side. This part, i.e. eq. (7)-(10), is the weakest part of the description. I think, I know what you want but this is not correctly formulated.
- Eq. (12) and (13) should explain the iteration procedure. I would recommend to do it in more detailed way, using e.g. two levels n and $n + 1$
- L275-305: I understand this part as a formulation of the iteration procedure shown e.g. in Fig 5. but it can certainly be improved (see major point).
- Fig 6, source latitude distributions: this quantity is not well-defined in the paper, please put a reference to the equation in the method description
- Fig 7: please explain how profiles of the path integrated lifetimes were calculated. Is every τ_i a result of iteration step i?

- General: I think that your partition of the source regions into NAM and tropics is a good approximation to demonstrate the method. However, I think the air composition in the tropic during boreal summer is strongly determined by sources within the Asian summer monsoon region. Maybe something for discussion.