Author Comment to Referee #2

ACP Discussions doi: 10.5194/acp-2016-463
(Editor - Peter Haynes)
‘Long-range transport pathways of tropospheric source gases originating in Asia into the northern lower stratosphere during the Asian monsoon season 2012’

We thank Referee #2 for the very helpful review. Our reply to the reviewer comments is listed in detail below. Questions and comments of the referee are shown in italics.

Summary: In this study the authors examine the transport characteristics associated with the Asian summer monsoon during September-October 2012, using both measurements of trace gases (e.g. ozone, water vapor, methane) and idealized tracer simulations that provide information about the relative contributions of different boundary layer regions to upper tropospheric/lower stratospheric air masses. The study provides strong evidence that the eastward shedding of eddies from the monsoon anticyclone provides an important mechanism for transporting boundary layer air from India and South Asia to the lower stratosphere over northern midlatitudes. While this study provides a comprehensive analysis and important contribution to the field that will make it suitable for publication, I have a few major comments that need addressing before I recommend its publication. In particular I have one major concern about the authors’ interpretation of the air-mass origin tracers that needs addressing, as it may potentially affect the interpretation of the main results. I also have smaller comments that are, by comparison, less important.

We thank the reviewer for the opportunity to remove unclarities about the emission tracer approach used in our paper. A detailed discussion follows below.

Major Comment

1. I am concerned about the interpretation of the air-mass tracers as a fraction. It is definitely constructive to look at the relative contributions of *different source regions and I commend the authors' use of the diagnostic. However, more care should be taken in the interpretation of the tracer concentration as giving the fraction of air that was last at the earth's surface in a given source region. In particular, the simulation only covers 1 May 2012 - 31 October 2012. If the tracers are to be interpreted as fractions (as the authors intend them to be) then the sum of the air-mass tracers corresponding to all of the source regions must equal 1 (since the union of the source regions is the entire planetary boundary layer (PBL)). This is not the case, however, as shown in Figure 9. The sum of the red, blue and orange lines should, in principle, equal 1 (but does not). What this tells me is that the tracers have not been integrated to equilibrium so that there is a large amount of air that is not accounted for by the source fractions. This is a known issue when dealing with air-mass origin tracers (Orbe et al. (2015)) and I am concerned about what this means for the main conclusions in the study. Please either start the simulation much earlier (to ensure tracer equilibration by September 2012) or remove all references to "fraction" because this interpretation is not correct. Alternatively, it is possible that I am missing something important in the authors' definition of "residual" (by which I interpret the rest of the PBL) - if this is the case, please clarify in the text.*

Orbe, Clara, Paul A. Newman, Darryn W. Waugh, Mark Holzer, Luke D. Oman, Feng Li, and Lorenzo M. Polvani. "Airmass Origin in the Arctic. Part I: Seasonality." Journal of Climate 28, no. 12 (2015): 4997-5014. (Figure 3b)

The reviewers interpretation of the residual is correct, however we believe that there is a misunderstanding regarding an important point in our approach: The composition of air in the UTLS is always a combination of aged air masses from the stratosphere and free troposphere and young freshly injected air masses from the boundary layer. Only for the young air masses, our characterization of air mass origin is conducted.

In our approach we cannot run the atmosphere to equilibrium as we are interested in working out the contribution of young, freshly injected air to the composition of both the Asian monsoon anticyclone and the extratropical lower stratosphere. In so far our approach is different but complementary to the approach by Orbe et al..

We are aware that we had not brought this point across very well in our original paper (ACPD Version). We have now improved the presentation of our modeling concept and its impact on the results throughout the paper (see in particular Section 3 in the revised version).

To go in more detail, the idea of using artificial tracers to identify transport has been used for a long time in both Eulerian (e.g. Mahlman and Moxim, 1978; Stone et al., 1999) and Lagrangian models (e.g. Bowman and Cohen, 1997; Bowman and Carrie, 2002; Günther et al., 2008; Vogel et al., 2011). In our approach, the artificial emission tracers in CLaMS are designed to identify possible boundary source regions in Asia that could contribute to the composition of the Asian monsoon anticyclone and the extratropical lower stratosphere during the course of the monsoon season 2012 considering advection and mixing processes.

The CLaMS transport scheme consists of advection, which is the reversible part of transport (the trajectory), and mixing, the irreversible part of transport. At beginning of the CLaMS model simulation each air particle is marked by a boundary layer tracer (Ω). That means, for example, that the boundary layer tracer (Ω) for air parcels in the boundary layer (BL) is set equal to one and everywhere else (free troposphere and stratosphere) equal to zero. Mixing in CLaMS between an air parcel from the boundary layer ($\Omega = 1$) and an air parcel from the free troposphere or stratosphere ($\Omega = 0$) is implemented by insertion of a new particle. The boundary layer tracer (Ω) of the new particle is then equal to 0.5. Successive mixing processes between boundary layer air ('young air masses') and air masses from the free troposphere or stratosphere ('aged air masses') during the course of the simulation yield a tracer distribution differing from the initial distribution ($\Omega = 1$ or $\Omega = 0$). In the CLaMS simulation used here, air masses in the model boundary layer are marked by boundary emission tracers every 24 h (the time step for mixing in CLaMS) during the simulation period from 1 May 2012 until end of October 2012. Thus in our simulations, the age spectrum is relatively simply constructed from the response to a single pulse during $(t_i, t_i + \Delta t_i)$ for times $t > t_i$ (see Holzer et al., 2009), with t_i equal to 1 May 2012 and Δt_i equal to 6 months. This pulse marks 'young air masses'. E.g. a value of Ω equal to 0.4 means that 40 % of the air parcel is younger than Δt_i (= 6 months; 'young air masses') and 60 % are older than Δt_i released from the model boundary layer before 1 May 2012 (emission time $t_e < t_i$; 'aged air masses'). We used this single pulse approach because the focus of the paper is to analyze the influence of fresh emissions from different boundary regions using the meteorological conditions of the year 2012. Here, the air-mass fraction $f(r, t \mid \Omega)$ is defined to be the fraction of air at a location r and time t

(with $t_i \leq t \leq t_i + \Delta t_i$) that was emitted since t_i within Δt_i from the model boundary layer (BL).

To get the information about the origin of young air masses within the model boundary layer, we divided the boundary layer (BL) in different regions, i.e. different artificial tracers of air mass origin, referred to as "emission tracers" (Ω_i of the number n = 17) are introduced in CLaMS (see paper table 1). Within the boundary layer, the sum of all different emission tracers (Ω_i) is equal to 1 ($\Omega = \sum_{i=1}^{n} \Omega_i = 1$). During the course of the simulation, the air-mass fraction $f_i(r, t \mid \Omega_i)$ is defined to be the fraction of air at a location r and time t (with $t_i \leq t \leq t_i + \Delta t_i$) that was emitted since t_i within Δt_i from the model boundary layer (BL) in region Ω_i. Note however that emission tracers only describe the contribution of the young air masses.

The fraction of air from different boundary regions $\sum_{i=1}^{n} f_i(r, t \mid \Omega_i) = f(r, t \mid \Omega)$ shown in Fig. 9 and also Figs. 10 and 11 (red, blue, orange) is less then 100 % because only "young air masses" emitted since 1 May 2012 are counted. Air masses originating in the free troposphere or stratosphere ('aged air masses') also contribute to the composition of the extratropical lower stratosphere. Thus in Fig. 9 region No.2 (gray area), the sum of air masses originating in India/China, in Southeast Asia/tropical Pacific and in the residual surface ($\sum_{i=1}^{n} f_i(r, t \mid \Omega_i)$) is roughly 45 %. The remaining 55 % of the composition of the lower stratosphere in this region is from the free troposphere and the stratosphere. Fig. 14 illustrate how the contribution of the model boundary layer ($f(r, t \mid \Omega)$) to the extratropical stratosphere rises to 44 % at 360 K, to 35 % at 380 K, and to 23 % at 400 K (see Sect. 4.4.2) from May until the End of October 2012. The remaining percentages to 100 % are contributions from "aged air masses" originating in the free troposphere and stratosphere.

We added the following sentence: 'On 26 September 2012 (see Fig. 9, top), a very pronounced signature of tropospheric air in the lower stratosphere is found between 09:05 UTC to 10:17 UTC (No. 2). Here, the contributions of the emission tracer for India/China and Southeast Asia /tropical Pacific Ocean are up to 20 % and 23 %, respectively (up to 5 % from the residual surface). Thus, the sum of all emission tracers for model boundary layer is roughly 48 %. The remaining 52 % of the composition of the lower stratosphere in this region is from aged air masses originating in the free troposphere and the stratosphere at the beginning our the CLaMS simulation on 1 May 2012.'

The technique used in our paper is different compared to the approach used in Orbe et al. (2015). A detailed comparison of our approach and the approach used in Orbe et al. (2015) is published in (Vogel et al., 2015, Sect. 4 Discussion). For clarification we repeat the corresponding paragraphs published in Vogel et al. (2015) here in our authors' comment:

"In contrast to our study, Orbe et al. (2015) used tracers of air mass origin in model simulations to infer the impact of boundary regions in Asia to the tropical lower stratosphere with an approach that the different tracers have equilibrated so that the sum of all tracers of air mass origin is equal to unity within the entire atmosphere (using a spin-up of 20 years). This approach provides for each air parcel the information about the origin within the boundary layer. However, this approach provides no information on the transport times from the boundary layer in Asia to the tropical lower stratosphere. To infer the transport times in the approach by Orbe et al. (2015), they introduce a boundary impulse response that marks air that left the boundary layer on a certain day (1 July). They infer transport times of 1 month from the boundary layer in Asia into the tropical lower stratosphere in July when the Asian monsoon is active. In this respect, their pulse serves a similar purpose as our seasonal tracer set-up. However, our approach considers all air masses that left the boundary layer over the course of the 2012 monsoon season since May and thus reflects the meteorological conditions of the entire 2012 monsoon season."

Specific Comments

1. *Line 29, Page 3: I am wary of the use of the term "transport pathways." The air mass origin tracers only tell you where air was last in contact with the boundary layer. They do not provide a sense for how the air arrived at the receptor location, so please remove all references to pathways. To infer pathways you would need to use idealized tracers similar to those used for inferring the age spectrum or, most appropriately, the path density tracers examined in Holzer (2009)*

   Holzer, Mark. "The path density of interhemispheric surface-to-surface transport. Part I: Development of the diagnostic and illustration with *an analytic model." Journal of the Atmospheric Sciences 66, no. 8 (2009): 2159-2171*

Our approach allows the relative contribution of air masses of varying origin to be quantified for every individual CLaMS air parcel on a certain location (r_j) and for each time (t_l) during the course of the simulation. Thus, transport pathways from the boundary layer into the UTLS (occurring since 1 May 2012) can be inferred analyzing the spatial-temporal evolution of artificial emission tracers during the simulation period. The change of the fraction of air originating in region Ω_i on different locations (r_j) for different times (t_l) $f_{ijl}(r_l, t_j \mid \Omega_i)$ define in our approach the transport pathways. However, we agree with the reviewer that the term "pathway" might have been used a bit to excessively and have removed the word on various locations in the paper.

2. *Line 8, page 6: Again, reservation about the word "pathway."*

   okay, we dropped "pathway"

3. *Lines 33-35, page 6: The sum of all of the air-mass fractions does not equal 1, leaving a large fraction of air unaccounted for. Therefore, I am not confident in the statement that "that air masses originating in India/China and Southeast Asia/Pacific Ocean almost exclusively contribute to the chemical composition of the separated anticyclone." Please either start your simulation earlier (to ensure equilibration of the airmass tracers) or do not use the word "fraction".*

   We agree the air masses originating in India/China and Southeast Asia/Pacific Ocean do not exclusively contribute to the chemical composition of the separated anticyclone. For clarification we added the following sentence to the paper:
   "Further, the emission tracer for Southeast Asia / tropical Pacific Ocean contributes to the composition of the Asian monsoon anticyclone, however to a smaller extent compared to the emission tracer for India/China as shown in Fig. 4 (right). Contributions from the residual surface are of minor importance (see Sect. 4.2). Note that contribution of air masses originating in the free troposphere and stratosphere

> ('aged air masses') als contribute to the composition of the Asian monsoon anticyclone of about 25 % at the end of September 2012 (see in Vogel et al., 2015, Fig. 9)."

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