Author Comment to Referee #3

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 #3 for the good evaluation of our paper. Following the reviewers advice we revised some parts of the paper for the purpose of clarification. Our reply to the reviewer comments is listed in detail below. Questions and comments of the referee are shown in italics.

The authors use the global Lagrangian CLaMS model, with artificial and chemical constituent tracers to quantify the contributions of different boundary Layer (BL) source regions in Asia to the Asian Summer Monsoon (ASM) anticyclone, and from there to extra-tropical lower stratosphere (ExLS), for the 2012 ASM season. Further, they illustrate the transport pathways for BL source air, accumulated in the ASM anticyclone, to reach the ExLS, via eddy shedding in the upper troposphere, subsequent filamentation and penetration into the stratosphere, associated with Rossby wave breaking along the subtropical jet. They also consider the westward shedding of air from the ASM anticyclone into the tropical upper troposphere.

The authors use the simulated artificial tracers, and simulated ozone, CO, and water vapor, to interpret small-scale structures observed along aircraft flight tracks as filamentary intrusions of BL source air associated with the ASM anticyclone into the lower stratosphere over Northern Europe. Further, they use the artificial tracers to quantify the contribution of different BL source regions to the ExLS over the 2012 ASM season, and use CLaMS simulated water vapor to estimate the contribution of Asian source regions to water vapor in the ExLS.

The study builds upon earlier studies that have illustrated troposphere-stratosphere exchange (STE) mechanisms associated with the ASM, going *back at least as far as Dethof et al. (1999), a paper which the authors cite. At the same time the use of the artificial and constituent tracers with the CLaMS model to quantify estimates of the contributions from Asian (and other) BL source regions to the ExLS is I believe a step beyond these earlier studies. Quantification of water vapor contribution to the NH lower stratosphere (p.13) is particularly interesting. The cross-section of the filamentary structure (Fig. 6) provides an illuminating illustration of intermediate (mixed) constituent and stability conditions between the tropospheric and stratospheric air mass characteristics.*

The comparison of aircraft observations of the low stratosphere over Northern Europe with CLaMS tracer maps interpolated to the aircraft flight tracks illustrates effectively that the simulation of tropospheric filaments in the low stratosphere represents real-world conditions, and supports the quantitative estimates of BL source influence in the ExLS presented later. The analysis and interpretation is reminiscent of that conducted by Fairlie et al. (2007) for INTEX-NA aircraft observations; the authors may wish to add correlation scatter plots of the observed O3, CO or CH4, water vapor to further illuminate air mass origin and characteristics of mixed troposphere-stratosphere air masses.

I think the paper could use an editorial review for the English and sentence structure. There is occasional awkwardness in the sentence structure, and some choice of wording that I find confusing, and may be a translation issue (see some examples below). Nevertheless, I think the paper is suitable for publication in ACP given consideration to these issues and the points listed below, most of which are minor and for the purpose of clarification.

We agree with the reviewer that tracer-tracer correlations are a powerful technique to study mixing processes between the troposphere and stratosphere. Tracer-tracer correlations would integrate the information from the aircraft observations and the CLaMS model to quantify and characterize signatures of mixing between stratospheric and tropospheric air (e.g. monsoon air) and therefore give insights in single mixing processes. However, in this paper we didn't show tracer-correlations because that would be material for an additional new study that we are planing. Therefore, to analyze tracer-tracer correlations of different chemical species from different TACTS/ESMVal measurements and tracer-tracer correlations from the CLaMS model using in addition tracers of air mass origin (e.g. similar as in Vogel et al. (2011)) would go beyond the scope of the paper discussed here. For the TACTS flight on 30 August 2012 an analysis of tracer-tracer correlations (O_3- CO) is already published in Müller et al. (2016).

We would like to thank the reviewer for carefully reading our paper and identifying some parts of the paper that need revisions for the purpose of clarification (details see below 'Minor Comments'). For the most comments identifying ambiguities, the reviewers interpretation was correct (see below). Following the reviewers advice, we revise those parts of the text. In general, some English corrections are already made in the revised version of the paper. After the acceptance of our paper by ACP a language revision of the paper will be made by the Language Services of Forschungszentrum Jülich.

Minor Comments

1. *p.6, line 31, What is meant by "Maritime Continent"?*

   The expression "The Maritime Continent" describes the region between the Indian and Pacific Oceans including the archipelagos of Indonesia, Borneo, New Guinea, the Philippine Islands, the Malay Peninsula, and the surrounding seas.

   We added in the revised version of the paper: '...Maritime Continent (the region between Indian and Pacific Oceans)...'

2. *p.7, line 12-13: Comment: The authors will recognize that the transport is only irreversible if the tropospheric intrusion is mixed into the stratospheric surroundings. It is conceivable that an intrusion across the PV=7.2 PVU could return to the troposphere downstream.*

   We agree that the sentence can be misinterpreted, therefore we remove 'irreversible' and write in the revised version: 'Isentropic transport of air masses across the 7.2 PVU isoline indicates exchange between the tropics and extratropics due to wave breaking.'

3. *p.7, lines 14-15: Comment: I am unable to see the PV=7.2 PVU isopleth enveloping a "region of enhanced tracers"*

We agree that the sentence is somewhat unclear. We revised the sentence as follows: 'On 20 September 2012, at the northern flank of the separated anticyclone the 7.2 PVU isoline is in the region where strongest gradients of the emission tracers occur indicating the transport barrier at 380 K (see Fig. 3 and 4).'

4. *p. 7, line 31, instead of "surface that is" do you mean "surface, i.e.,"? I.e., are the authors stating the definition of "residual" here?*

   For clarification we introduced a definition for the 'residual surface' in Sect. 3 (CLaMS simulations using artificial tracers of air mass origin) as follows:

   'The most important regions for our study are India/China (= Northern India (NIN) + Southern India (SIN) + Eastern China (ECH)), Southeast Asia (SEA) and the tropical Pacific Ocean (TPO). The sum of all model boundary layer tracers ($\Omega = \sum_{i=1}^{n} \Omega_i$) without contributions from India/China, Southeast Asia, and the tropical Pacific Ocean is summarized in one emission tracer referred to as "the residual surface" (= Ω - NIN - SIN - ECH - SEA - TPO).'

   and revised the sentence on page 7, line 31, Sect. 4.1, as follows:

   'Fig. 5 (bottom left) shows the emission tracer for the residual surface (the entire model boundary layer without contributions from India/China, Southeast Asia, and the tropical Pacific Ocean, see Sect. 3).'

5. *p. 7, line 32-33, reference "no signature." Would the authors be more quantitative here? Looks like up to 10-15% is due to "residual" sources in the anticyclone.*

   We revised the sentence as follows: 'The contribution of the emission tracer for the residual surface is approximately 10-15 % within the separated anticyclone indicating that air masses originating in India/China and Southeast Asia / tropical Pacific Ocean almost exclusively contribute to the chemical composition of the separated anticyclone.'

6. *p.8, line 12, reference "indicating transport from the troposphere into the stratosphere." This is according to the definition of the authors, based on the work of Kunz et al.*

   Yes, that is correct. Therefore we revised the sentence to be more precise as follows: 'At 380 K enhanced contributions of the emission tracer from India/China are also found north of the 7.2 PVU barrier indicating transport from the troposphere into the stratosphere according to the definition by Kunz et al. (2015).'

7. *p.9, reference discussion of Fig. 9 emission tracer plots, here and elsewhere. Please confirm for the reader if "residual" includes all BL surfaces other than those identified (China/India, SEAsia/ tropical Pacific). How should the reader interpret the sum of these percentages being much less than 100%, e.g. does the remainder comprise background lower stratospheric air, unconnected to any BL surface in past 5 months?*

   As mentioned above we added a definition for 'residual surface' in Sect. 3 (CLaMS simulations using artificial tracers of air mass origin) in the revised version of the paper. The interpretation that the sum of the percentages of all boundary layer tracers is less than 100% because of contributions of aged air masses from the stratosphere is correct. For further clarification we added the following sentence in the revised version:

   '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.'

8. *p. 10, discussion of Figs. 9-11. It would be helpful if the authors labeled the locations of flight segments "1", "2", "3", etc. on the maps in Figs. 7-8, to help the reader identify the features highlighted in the flight data to features on the CLAMS maps. Additionally, the flight data appears to be higher temporal resolution than the CLAMS profiles (e.g. the profile of FISH H2O). It may be helpful to add a time-averaged data profile at the same resolution as the CLaMS for better comparison. Tracer-tracer correlation plots may also be a useful addition (see above).*

   We agree that numbers in Fig. 7-8 can help the reader to identify the regions that are impacted by young tropospheric air masses. We revised figures 7-8 and also Fig. 10 and 11 as shown below.

   Within Fig. 9-11, the measurements of CO, CH_4, H_2O and O_3 are shown in the highest resolution that was available for each species to demonstrate the variability of the measurements. For the interpolation of CLaMS results as described in Appendix B, backward trajectories are calculated every second along the flight path.

   As discussed above an analysis of tracer-tracer correlations would be beyond the scope of this paper.

9. *p. 11, lines 13-15. Suggestion: I think the authors mean to emphasize the locations (Atlantic and Pacific Oceans) here, rather than the mechanism (Rossby wave breaking). They may want to leave out "Rossby wave breaking" in this sentence to keep the stress on the locations.*

   That is a good point. We changed the sentence as follow: 'Finally, Fig. 11 shows that transport from air masses originating in the Asian monsoon anticyclone into the lower stratosphere take place most frequently over the Pacific and Atlantic Ocean ....'

10. *p.11, lines 18-20. This sentence seems a bit out of context here. Perhaps reference to SE Asia/ Tropical Pacific contribution (the appendix) would sit better after the introduction to Fig.12 (p.11, line 2, after "September 2012").*

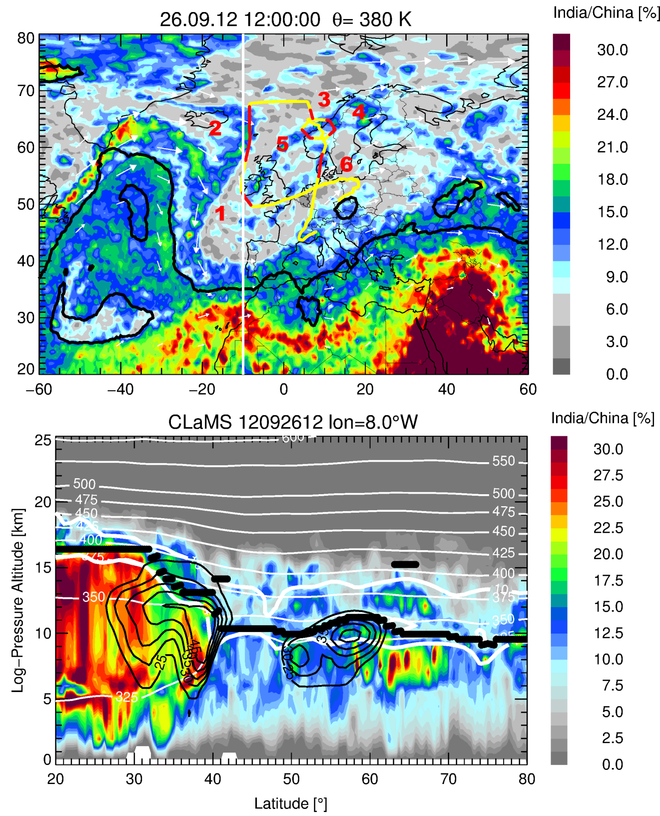

Figure 1: (= **Fig. 7 within the ACPD paper**) Horizontal (top) cross-section of the fraction of air originating in India/China over Europe om 26 September 2012. The flight path transferred to noontime of the TACTS/ESMVal flight is shown as yellow line. Segments of the flight in the lower stratosphere with enhanced measured CO, CH_4, and H_2O and reduced O_3 compared to the stratospheric background are highlighted in red and numbered for clarification (the same numbers are used in Fig. 9 The climatological isentropic transport barrier of 7.2 PVU at 380 K is shown as thick black line. The white thick line marks the position of the vertical (bottom) cross-section at 8°W longitude which is similar to the cross-sections shown in Fig. 5 and 6.

We agree. The paragraph is shifted as proposed.

11. *p.11, reference discussion of "transport pathways": The title of 4.4.1 is "transport pathways into the lower stratosphere." The authors have illustrated that "eastward eddy shedding" on the NE side of the anticyclone, and subsequent transport and filamentation of material can be a pathway to reach the stratosphere (pathway 1, line 10). But, what about the "westward eddy shedding" (pathway 2, line 11) from the anticyclone to the TTL? I find no discussion of this as a potential pathway to the stratosphere, via e.g. diabatic ascent (Garny and Randel, 2015).*

    We agree that pathway 2 needs some further discussion and added therefore the following paragraph: 'Transport of air masses from the Asian monsoon anticyclone southeastwards into the tropics by e. g. westward eddy shedding causes increase of contributions of air masses originating in the boundary layer in India/China within the TTL. These air masses could penetrate into the upwelling in the deep branch of the Brewer-Dobson circulation and thus could be transported further up into the stratosphere (e. g. Garny and Randel, 2016).'

12. *p.12, discussion of Fig. 14 and Table 2. Please clarify how these metrics are computed. Are they achieved by area weighting daily isentropic "fraction of air" maps for areas north of 30 o N and for PV greater than the "transport barrier" PV? Are you saying for example that by end October 2012, almost 20% of the air in the NH at 360K north of these delimiters originates in the India/China BL within the previous 5 months?*

    The reviewers interpretation of the percentages in Fig. 14. is correct. For clarification we revised the text as follows:

    'The accumulation of young air masses from Asia since 1 May 2012 in the extratropical lower stratosphere is calculated using the isentropic transport barrier at different levels of potential temperature derived by Kunz et al. (2015) as shown in Fig. 14 and Tab. 2. To calculate the percentages of different emission tracers within the lower extratropical stratosphere at a certain level of potential temperature, we use the following approach: For each day between 1 May and 31 October 2012 a mean value for each emission tracers of all CLaMS air parcels is calculated for PV values larger (lower) than those at the transport barrier (see Tab. 2) and for air masses poleward of 30° N (30° S) at a specific isentropic level ($\Theta \pm$ 0.5 K). End of October 2012, the contributions of all boundary emission tracers on the composition of the extratropical northern lower stratosphere are at 360 K ≈44%, at 380 K ≈35%, and at 400 K ≈ 23%, with the remaining fraction of air consisting of aged air. The highest contributions of the boundary emission tracers are from India/China uplifted within the Asian monsoon anticyclone, from Southeast Asia, and from the tropical Pacific Ocean. The contribution of all other regions of the Earth's surface (residual surface) are of minor importance (Tab. 2).'

13. *p.14, lines 12-14, reference "A mixing layer .... (see Fig. 5)" It is unclear to me what feature is being identified in Fig. 5. I see no discussion of such a mixing layer in earlier discussion of Fig. 5. Indeed 2 lines earlier (p.14, line 11) the thermal tropopause is described as a "strong transport barrier above the separated anticyclone." Do you really mean Fig. 5? Are you referring to the thin layer of strong vertical gradients in fractions of air and in simulated CO at the thermal tropopause south of 40 o N, 370-400K)? What is the evidence for mixing, and what is the mechanism? Or do you mean Fig. 6 instead where mixed troposphere-stratosphere characteristics of BL source fractions, CO, and buoyancy frequency are evident between the double thermal tropopauses? The discussion of PAN (p.14, lines 16-23) suggest you are discussing Fig. 5, but what I see is a strong vertical gradient at the tropopause, not a zone of mixed tropospheric and stratospheric characteristics. I read reference to a "small mixing layer around the tropopause" (p.14, line 33) which seems to minimize the significance of mixing here; if it's not significant (strong transport barrier), why spend a whole paragraph (lines 10-23) describing it?*

    We agree the discussion about the mixing layer is redundant. Therefore, we remove this paragraph within the discussion section as well as the statement "small mixing layer around the tropopause" (p.14, line 33) within the conclusions.

14. *There are some places where the English is a bit obscure to me, e.g.*

on p.15, line 15, I don't know what "yield predominantly" means in this context. I wonder if the words "yield to" (e.g. on p.15, line 23) is intended to mean "serves to" or "results in", i.e. "serves to increase," or "results in increasing."

The sentence is revised as follows: 'This second transport pathway is mainly caused by westward eddy shedding, however this transport pathway enhances predominantly the contribution of fresh emission from India/China to the composition of TTL air at 380 K in summer and autumn 2012.'

References

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Kunz, A., Sprenger, M., and Wernli, H.: Climatology of potential vorticity streamers and associated isentropic transport pathways across PV gradient barriers, J. Geophys. Res., 120, 3802–3821, doi:10.1002/2014JD022615, URL http://dx.doi.org/10.1002/2014JD022615, 2014JD022615, 2015.

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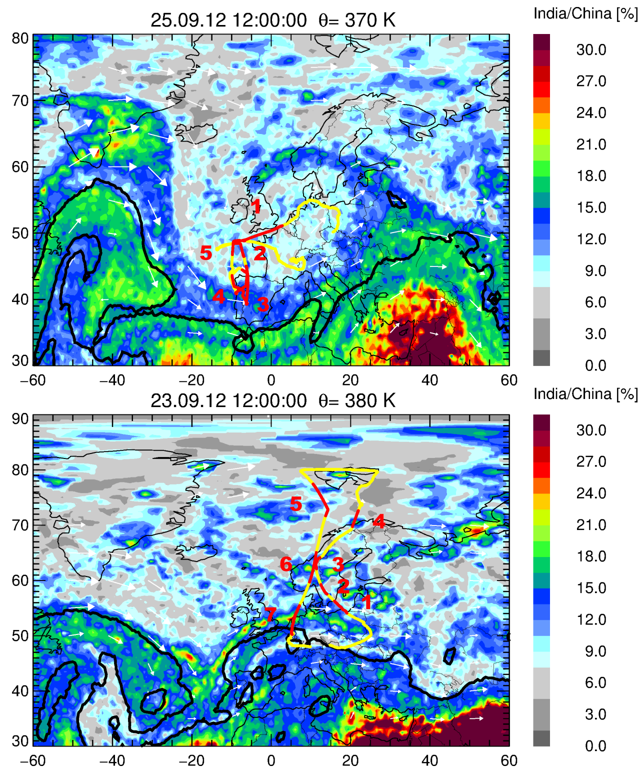

Figure 2: (**= Fig. 8 within the ACPD paper**) Horizontal cross-section of the fraction of air originating in India/China over Europe on 25 September at 370 K (top) and on 23 September 2012 at 380 K (bottom). The flight paths transferred to noontime for the TACTS/EsmVal flights 2012 over northern Europe are marked as yellow lines. Segments of the flight in the lower stratosphere with enhanced measured CO, CH_4, and H_2O and reduced O_3 compared to the stratospheric background are highlighted in red and numbered for clarification (the same numbers are used in Fig. 10 and 11). The climatological isentropic transport barrier of 6.0 PVU (at 370 K) and 7.2 PVU (at 380 K) are shown as thick black lines.

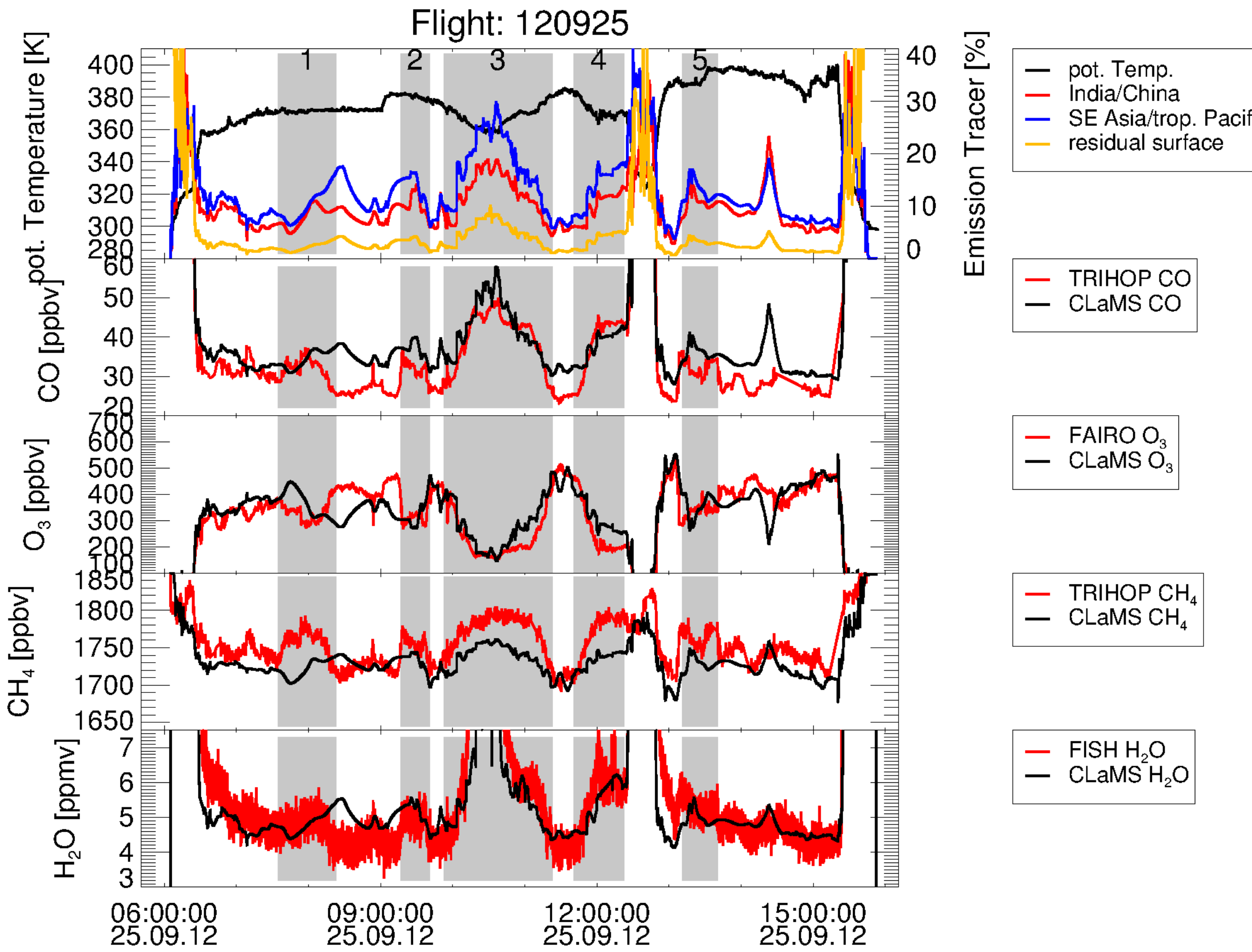

Figure 3: (= **Fig. 10 within the ACPD paper**) As Fig. 9, but for the flight on 25 September 2012.

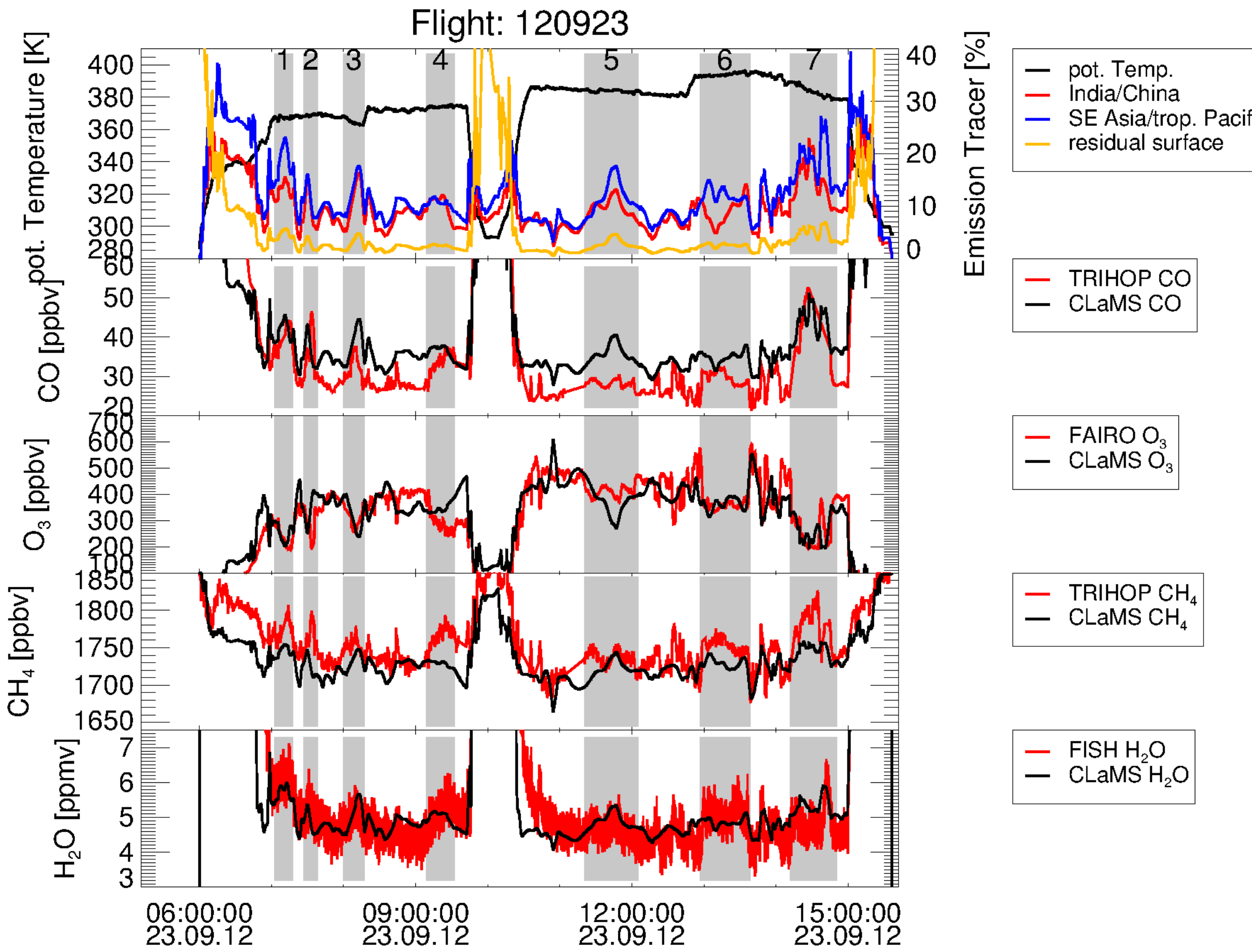

Figure 4: (= **Fig. 11 within the ACPD paper**) As Fig. 9, but for the flight on 23 September 2012.