

Interactive comment on “Trapping, chemistry and export of trace gases in the South Asian summer monsoon observed during CARIBIC flights in 2008” by A. Rauthe-Schöch et al.

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Answers to Reviewer #1:

General comments

We thank the reviewer for reviewing our paper. In the opening statement, the reviewer highlights that our paper contains “interesting analysis” with the potential for “making an important contribution”. At the same time, it is stated that the paper “contains serious

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weaknesses”, “In particular the presentation is chaotic and often poorly worded”. We do not agree with these last two statements. The presentation is not “poorly worded”. The presentation is systematic, not chaotic.

We have changed parts of the presentation and changed some wording to meet this criticism as is documented below. We note that the scientific content as such was not seriously questioned. We were a bit confused by the reviewer’s statement: “The paper includes many unnecessary descriptive details that obscure important results and analysis (sic) that fails to adequately support the conclusions drawn from it”.

Section 3.1.2. This section is considered too chaotic: (We assume that this is not meant to mean that a chaotic section would have been acceptable). We have changed this section throughout. When however the reviewer writes “some conclusions seem overly speculative” at least one example should have been given. We do not agree with the reviewer on this.

Section 3.1.3. This section contains far too many descriptive details: We have removed as many descriptive details as the order of argument allows for. Unfortunately, the reviewer does not even give one example of a descriptive detail that is not needed (out of the far too many).

Section 3.2. The section could be more meaningful showing statistics in Fig. 9 and 10: Adding more than one flight to Fig. 9 (now Fig. 10 in revised manuscript) would overcrowd the plot with too many coloured lines. It was already split in two panels to avoid this kind of overcrowding. However, we have changed Fig. 10 (now Fig. 11) to show source region statistics for all flights including panels showing the month-to-month variability. We have also followed the suggestion of Reviewer #2 to consider all trajectory points below 5km instead of only the first time the trajectory

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reaches the lower troposphere.

Section 3.3.1. The reviewer writes “..this section could be deleted with no loss to the integrity or impact of the paper”: We do not agree. Section 3.3.1. is a short subsection of Sect. 3 and deals by means of forward trajectories with the outflow of air from the UTAC for the locations for which we have measurement data. There are relatively few aircraft observation in the South Asian Summer Monsoon. The logic of this paper is to use our aircraft observations in relation to the trapping, chemistry and export. Surely, a valid question pertains to the outflow of this air. A focus for current research is the pathway of air from the regions involved into the stratosphere. We show by means of meteorological analyses where air from the UTAC is transported to. Its degree of dilution and the composition of the diluting air masses are not extractable from our data and only accessible at the moment by modeling. It would be a shame to delete this brief section from the paper. The decrease in impact would well surpass the decrease in size. We note, talking about impact, that the review spent most words on this section. We reiterate, it is at least of general interest to atmospheric chemists to know to where pollution that has been accumulated/trapped in the UTAC is exported.

“The criterion for determining the influence of air along the flight path in receptor regions is imprecise and subject to severe sampling problems”: We do NOT deal in this section with the influence of air in receptor regions. Therefore this statement in the review is void. We have, however, modified the text to mention that monsoon pollution export is not the only source of pollutants for the source regions and that its influence diminishes the further away the source region is from the monsoon UTAC.

“The analysis in this section is weak”: It must be clear that we deal here with a short subsection on the export regions. We cannot go further, and that may make

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the contents of this section “weak” in a quantitative sense. There are papers in the literature (e.g. *Scheeren et al., Atmos. Chem. Phys.*, 3(5), 1589–1608, 2003) that deal with the impact of pollution transported from the UTAC elsewhere.

Section 3.3.2. The reviewer states. ”A cynical reviewer would wonder if the sampling and analysis criteria were chosen to provide the answer that the authors wanted rather than physically meaningful results”: We find this an unworthy statement, even when anonymous. We expect facts in a valid review. Our reply: This section deals with the “age” of air masses, which is a complex concept. Two very different approaches are compared. One is that of a chemical clock based on NMHC ratios. Here a main a priori uncertainty is the choice of the amount of OH in the air masses. We justify this choice clearly (Spivakovsky’s OH distribution). The other one is that of trajectory calculations. Here a main a priori uncertainty is the starting point of the trajectories. We have now added a new Table 3 which lists the slopes of the least-squares fits for the correlations using start longitudes between 80°E and 100°E. We have changed the description of the results in the revised manuscript to: “For July, the best fit was found when comparing the time since the air had last been east of 95°E. For August, the correlations do not change much for a source region between 85°E and 95°E. Concerning the sampling, we had already mentioned in the “methods” section that air samples were only collected during the first two flights in each month to achieve a better spatial resolution given the fixed number of 28 available air samples per month. We have repeated this in the beginning of this section and explained that consistent NMHC photochemical ages could not be calculated for the June and September samples. Therefore we are restricted to the samples from July and August. At the same time, these months represent the core of the monsoon period in India. The remaining 40 % of this section is used to discuss the problems of the two approaches.

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Section 3.3.3. The reviewer writes: “This section seems disconnected from the rest of the paper”: As we outline above, and as the title of the paper communicates, this paper is about the trapping, chemistry and export of trace gases in the South Asian Summer Monsoon”. This very section – Monsoon UTAC “leak rates” – deals with the “escape” of air from the UTAC. Estimates for a “residence time” of air in the UTAC are very useful and form an intrinsic part of this paper.

Specific Comments

Page 3, line 12. “The word “remarkable” is too subjective and inappropriate and should be deleted throughout”: The word “remarkable” had been used only thrice in the old manuscript and is now only used here in the abstract to highlight the consistency of a feature over 3500 km over the entire monsoon period for a range of trace gases. We adopt the suggestion by the reviewer about the consistent north-south gradient.

Page 11, line 12: We refer now to *Randel and Park, J. Geophys. Res., 111(D12), D12314, 2006*, and use the word suggested by the reviewer, namely “interesting”.

Page 11, lines 18-19: We now write: “In general, the centre of the UTAC is observed to be furthest north during July (Fig. 4). This is consistent with meteorological studies of monsoon development, its northward propagation and recession (IMD, 2009).”

Page 12, lines 13-16: As suggested, we have now changed the old Fig. 5 (now Fig. 6) to include data from all flights together with the means and the 1- σ standard deviations. The numbers for mean and standard deviation in the southern (Δlat -7.5° to -2.5°) and northern (Δlat 2.5° to 7.5°) section of the UTAC are now mentioned in the

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text. They show that there is a significant difference between the two sections except for CO which has its maximum around $\Delta\text{lat } -3^\circ$ and decreases both northwards and southwards.

Page 13, lines 1-7: It is well known that CO mixing ratios are higher in the troposphere than in the stratosphere and vice versa for O₃. Therefore in-mixing of stratospheric air would also lead to an increase of O₃ and a decrease of CO mixing ratios. The trajectories and NMHC ages both show that the air has been transported for a longer time since it was loaded with pollutants when we measure it in the northern part of the UTAC compared to its southern part. We also discuss the difference in nucleation mode particle number concentrations in the Supplement Sect. S5. We therefore do not understand well where the “speculative” part is.

Page 13, lines 7-9: We have now added the thresholds used for this filtering, namely 1.3 PVU and 150 ppb. The filtering has been done in the same way as for our previous monsoon studies by *Schuck et al. (2010)* and *Baker et al. (2011)*.

Page 13, lines 9-10: We have changed this to “The distinction between freshly polluted air in the southern section and more processed, aged air in the northern section is also supported by a previous study of CARIBIC NMHC data (Baker et al., 2011).”

Page 14, lines 3-11. Please explain (briefly) why positive correlations indicate ozone formation...” We refer on page 14, line 5 to the paper by *Fishman and Crutzen (1978)*. Later we refer to other papers on ozone trends. This all is sufficient for a paper in an atmospheric chemistry journal. We have changed the wording to “Positive correlations”.

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Figure 8: We have added the mean and $1-\sigma$ standard deviation to the plot. They support the “C”-shaped profile for CO, the decrease of water vapour with height and the increase of the aerosol particle concentrations with height. In the case of O₃, the increase with altitude from 4 km to 12 km is at the limit of significance, i.e. the upper bound at 4 km is only slightly lower than the lower bound at 12 km. If we were to use the “patterns” or “trends” quantitatively, we would have done more statistical analyses. However, the main features are clear in Figures 8 and 5 (now Figures 9 and 6 in the revised text).

Page 16, line 29 to Page 17, line 7: No, the “C”-shape is a typical vertical profile structure. It is like someone smoking a cigar in a room. We see smoke near the person and we see smoke higher up in the room (often stratified). We thus see a “C”-shaped profile. This is basically due to an observational bias, because the pollution at higher levels in the room has passed in a fairly narrow single vertical corridor at relatively high speed to the higher level (see e.g. Fig. 2 in *Barret et al., Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2015-1011*). In aircraft observations in the troposphere we have a similar effect. It is noteworthy that pilots do not wish to fly in the region where the pollution from below is transported rapidly upwards (e.g. inside thunderstorms).

Page 19, line 7: The choice of 5 km is arbitrary, but it is a sensible choice. We have repeated the calculations for 3 km and 1 km altitude and obtained very similar results. This is now mentioned more explicitly in the text together with an interpretation of this fact, namely that there must be rapid more or less directly vertical transport. The reliability of backward trajectories close to the surface is generally considerably less than backward trajectories in the free atmosphere. For the question “where had the pollution been picked up” the choice of 5 km is reasonable.

Page 25, lines 19-20: The proposed word “reduced” is unsuitable. Reduced means

that a quantity is lesser than in a previous state. This does not apply here. We deal here with a vertical distribution pattern with higher mixing ratios at those levels where the outflow peaks and where dispersion over matter of days is less. We have changed the formulation to make it clearer. See above for an explanation of the higher pollutant levels above than below.

Page 25, lines 23-24: We have changed the formulation as it was indeed not clear.

Page 25, line 26 to Page 26, line 2: The statement by the reviewer “**for example, it could be interpreted as saying that limiting the number of chemical species observed is expected to make a data set less consistent**” is not correct. We have reformulated this part.

Page 27, lines 16-26: We disagree that this paragraph makes no logical sense. For clarification we have added at the end of this paragraph: “In other words, the difference between the two regimes is due to the time elapsed since fresh pollution was injected into the UTAC.”

Page 28, lines 12-13: Colleagues from other research institutions when dealing with the Asian Monsoon have introduced the concept “trapping”. In the current tone of reviewing the reviewer would point out that the air is not trapped, but escapes. So, this concept that is in use now would have been rejected. Meteorologists have used the concepts like “Meteorological bomb” (not rejected) or “Stratospheric fountain” (not rejected)” and we used “merry go around” (hopefully not ejected). As long as humans do science and it is communicated in language not all qualifiers will be perfect descriptors.

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Page 28, lines 16-24: The reviewer is mistaken. It does not refer to an artifact of the flight path.

Page 28, lines 26-27: We have removed this piece of information.

Selected technical details

We have incorporated the changes suggested by the reviewer. Please see the revised manuscript and the track-changes version for details.

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 6967, 2015.

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