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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Greenhouse gas relationships in the Indian summer monsoon plume measured by the CARIBIC passenger aircraft" *by* T. J. Schuck et al.

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We thank referee 2 for his review of our manuscript on greenhouse gas observations during the Indian summer monsoon.

It was pointed out in the referee's comment that our manuscript is rather long. We have taken this very serious, though we feel that the length of the text is not inappropriate given the amount of data presented. We revised the manuscript carefully, shortening the text where feasible. The main changes are the removal of table 3 and the corresponding paragraph in the text. Further changes include removing paragraphs from section 5.2 and removing parts of section 6.1. The introductory part of section 6 has been shortened and restructured, emphasising the limitations of the taken approach.

In the following we address the reviewer's more specific comments:

1) The key results here show an enhancement in CH_4, CO, SF_6 and H_2O associated with the anticyclone (often termed a "plume" in the paper), consistent with previous satellite observations (SF_6 is new).

Our in situ measurements should be seen to exist next to satellite measurements. Our CH_4, and CO_2 data can help to deepen the understanding of their distributions as monitored by satellite sensors. We believe that a strong point of our work is the combination of these trace gases. Often it is either CH_4, or CO, or CO_2, but rare are the studies tackling these tracers all at once, despite their strong link with the biosphere and oceans. We not only think that SF_6 is new in this respect, but also N_2O, and also CO_2 has to our knowledge not been discussed in the literature with respect to the Indian monsoon. However, its behavior in the monsoon anticyclone is highly interesting.

The latitudinal and temporal behavior of the relative maxima are reasonably consistent among these different tracers.

This consistency is highly interesting. As the reviewer pointed out, we have a limited number of one dimensional transects. Despite this, the behavior of the maxima is consistent. Considering the different sources, and their properties, this consistency is intriguing.

There is also a relative minimum in ozone, anti-correlated with H_2O (again, consistent with satellite observations). Variations of CO_2 are somewhat more complicated to interpret, because of strong seasonal variations in surface CO_2 amounts.

CO_2 indeed forms a complex case, but to estimate fluxes into the biosphere is valuable. We do not know of other measurement based studies that do this on this scale

All of these results are straightforward and interesting. The shape and position of the plume (Section 4.4) is interesting, but the analysis should be interpreted in light of

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the limited CARIBIC aircraft sampling (effectively several cross sectional snap-shots across the anticyclone).

We cut through the structure, or plume, at its lower level on the west of where maximum values have been observed by satellites. Our data give very accurate concentration levels, but indeed for a limited region of the plume. The entire chemical plume structure can only be reconstructed using meteorological, satellite and in situ observations in combination, we believe. In this paper, we attempt for the first time to get a detailed picture of the composition of the monsoon plume based on in-situ measurements, which naturally can only cover a small part of this large-scale feature. The limitations that arise from the snap-shot like monthly sampling of the westerly part of the plume have been addressed in the text, mainly in section 5.3 in which the structure of the plume is discussed but also in the context of emission estimates in section 6.

2) I find the detailed discussions regarding enhancements in the plume (Section 5) and estimates of emissions (Section 6) to be much longer than necessary, and I suggest these sections be reduced.

As long as there are no other such estimates, our estimates, even if they have a considerable uncertainty, must be presented. Data bases compiling source statistical data are notoriously prone to bias and uncertainties. Our estimates using large scale atmospheric measurements are an independent approach. (ref. paper by Levin) This is why we would maintain these two sections, that we however have made more concise. The fact that one can use SF_6 as a tracer to estimate emissions is a unique opportunity for the very type of data we have measured. We think that asking us to refrain from using our measurements would be asking very much. Both the US and Germany are planning major aircraft missions in the monsoon plume. Here we give a first taste of what can be expected, and particularly which parameters are essential. For instance, better accuracy SF_6 measurements (only few labs such as NOAA-ESRL or IUP Heidelberg can provide data with significantly higher precision) will allow better estimates. In the end the use of our data in sophisticated models of CH_4, CO_2, N_2O and CO will be their

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optimal quantitative use.

I like Figure 8, and much of the information on enhancements can be directly derived from there. However, the lengthy discussions on small details (such as the few April measurements discussed on p. 21) are less important, and take away from what could be a very concise summary.

We agree that Figure 8 shows immediately the “extent and intensity” of the plume in terms of latitude at cruising altitude, and the consistency in view of a limited number of transects is telling. The small details, however, we consider as essential to point out that deviations can occur and can be understood on the basis of meteorological information.

Also, Section 5.3 (Structure of the Plume) attempts to derive much detail, but ends up with speculation regarding mixing, etc.

We see this point and have removed part of this section.

While the trajectory analysis in Fig. 9 is interesting, these results for one flight may be difficult to generalize to the entire anticyclone structure (and the discussion comes across as speculative). Perhaps such analysis synthesizing results for many flights could be the subject of a future paper.

The mixing ratios measured appear to be tightly connected to meteorology (see Figure 7). We have kept the meteorological treatment however to a bare minimum. We would like to point out that the trajectories shown in Figure 9a are typical and representative examples and reflect the general pattern observed during all summer flights. Figure 9b is meant to show the overall circulation for this month and date but again, this looks similar for all months during the monsoon period. We think that this representation based on actual meteorological data is a very good illustration of the structure of the anti-cyclone. Here we also remark that we introduce the use of the concept “plume” instead of using “anti-cyclone”. The former is by definition pertaining to chemical composition, whereas “anti-cyclone” is a pure meteorological concept. Thus Figure 9 is meant to help the reader to form a mental picture of the anti-cyclonic structure. We also reiterate that despite the few transects the “picture” of the plume is highly consistent.

Likewise, I find Section 6 to be extremely qualitative and based on speculative assumptions.

We would consider our estimates to be semi-quantitative at least. "Extremely qualitative" as the reviewer coins, is in our opinion over-critical. We use two methods that have been used by many others before and were mentioned in the review by Andrea and Merlet. We have, in response to the criticism of the reviewer, more clearly elucidated that the correlation method has large errors and is not used in the final emission estimates. However, the intercepts obtained by this method, for which we also present a strict statistical analysis, show an agreement with independent estimates of background values. Thus we do not want to leave Figure 10 out, but we did omit Table 3.

Obviously, footprint of sources, the relation flux and mixing ratios all are hard to nail down, yet, the results we obtain need to be discussed. The agreement of the obtained emission estimates with the database (which has its own problems) is in fact surprisingly good. The reasons for agreement and disagreement must be discussed and this altogether makes the value of chapter 6. One important point here is the low flux of CO. A reason why the estimated flux is so low, may be the assumption about the background. In addition, CO is shorter lived, and it has a north-south gradient, which makes perhaps this estimate qualitative. CO is a widely studied gas and it is often used as a reference for emission estimates for example in biomass burning studies. Therefore is it important to understand its sources and sinks. Another interesting point is the net flux of CO_2. For CO_2, we do not know of any literature values for the monsoon region, although this gas is widely known to be of importance. Thus, we think this discussion is valuable and we hope to stimulate further studies using sophisticated transport

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models..

I think a more rigorous error analysis would result in very large uncertainties for emission estimates, given the uncertainties in the observed correlations (Fig. 10) and lack of knowledge of pathways and origins of air in the anticyclone, plus emissions for SF_6.

We certainly had not sufficiently clearly stated that we do not use these correlations but we use the calculations based on Figure 8. Having stated this, we are aware that we do make a number of assumptions that lead to inherent uncertainties that are hard to quantify. The resulting uncertainties may not even be that much larger than those in for instance estimating how much methane is emitted during the summer monsoon from this region using statistical data for rice paddies, ruminants, city sewers and so forth. The only other way is to use 3D models, and even in 3D models assumptions about sources have to be made. We think that using our extremely simple approach, plausible orders of magnitude for the fluxes of important trace gases have been obtained. In addition, we are not aware of any other flux estimates based on large scale measurements during the summer monsoon but hope that with our results we can stimulate further work on this using sophisticated transport models.

These uncertainties are acknowledged several times in the text, but not quantified. This section tends to ramble with far too many details (aspects of crop emissions from different countries, etc.), and this takes away from the quantitative focus of the first part of the paper. Overall I think the paper would be much improved by shortening or eliminating Sections 5-6, so that the resulting paper would be much more focused and quantitative.

As mentioned above, sections 5 and 6 have now been shortened and the discussion of emissions has been cut. However, as we have argued above, chapter 5 and 6 are essential results of our interpretation of the measurements. We have tidied up the text and made it more concise.

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 2031, 2010.