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Atmospheric Chemistry and Physics Discussions

Interactive comment on "MIPAS measurements of upper tropospheric C_2H_6 and O_3 during the Southern hemispheric biomass burning season in 2003" *by* T. von Clarmann et al.

T. von Clarmann et al.

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The authors would like to thank Dr Solène Turquety for her helpful comments. In this reply, the original reviewers' comments are inserted in *italics*.

Since all issues of the general comment are specified in the specific comments, we have reordered the comments for this reply by thread rather than by general vs. specific.

General comment: It would add to the scientific interest of the paper to explain how MIPAS upper tropospheric measurements are complementary with respect to ACE (which

also provides C2H6) and MOPITT CO. What is the main advantage? Why would people want to use these MIPAS data? What are the complementarities between C2H6 and CO?

Specific comment: Introduction: Since MOPITT provides well validated measurements of CO. It would be good to explain better why C2H6 is interesting for the analysis of biomass burning transport (sources, lifetime, etc.), as already mentioned in the general comments, and compare C2H6 to CO in particular.

The advantage over ACE is much better global coverage; the advantage over MOPITT is that various species are measured in the same airmass at similar spatial resolution. A short discussion of this issue will be included.

General Comment: The authors refer to the biomass burning "plume" or "plume-like pollution belt" throughout the paper. A plume usually refers to a transport event of pollution from a specific source. Here the authors discuss the transport from different source regions, mixing into a pollution belt. It would clarify the paper to explain from the beginning what is meant by "plume" in the analysis.

Specific Comment: An introduction of what the authors mean by "plume" would clarify the following discussion.

A statement about the terminology used here will be included at the end of the introduction.

P. 12068, L.21: MOPITT observed "enhanced tropospheric CO during the 2003 Southern hemispheric biomass burning season". Was 2003 particularly high? Why?

We do not mean "enhanced compared to other years" but "enhanced compared to background values". Our statement will be reworded for clarity.

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P. 12069, L.4: This statement is too strong, as ACE also provides measurements in the tropics, but with a lower coverage.

The statement will be reworded.

Section 2: It would be interesting to give an idea of the MIPAS coverage and horizontal (how many days for a global coverage?)

This information will be included.

What motivated the choice of the 21/10-14/11 2003 time period?

Since our personal and computational resources are limited, and no particular funding for the analysis of biomass burning was available, we could not afford to set up a dedicated biomass burning analysis period. Instead we had to use the C_2H_6 data which we had as a by-product of a funded project. The choice of the time period was driven by the needs of the funded project. Since this is an entirely unscientific criterion, we prefer not to discuss this issue in the paper. By the way, we realized that the time period actually was 21/10-12/11 2003. This will be corrected; affected analysis steps will be redone.

Section 2.1: P. 12071, L.2: It is not clear what is meant by the analysis of "relative distributions"...

This statement will be reworded for clarity, avoiding the term "relative distributions".

P. 12071, L.18: The uncertainty is in fact larger if the uncertainty on the spectroscopic parameters is taken into account. Does this number correspond to the retrieval error?

This number is the standard error of the mean, which represents both the random part

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of the retrieval error and the uncertainty due to the finite number of samples taken from an ensemble of a variable quantity. In the text we will replace the term “uncertainty of the mean” by the more specific technical term “standard error of the mean” for clarity.

P. 12071, L.25-26: Was the fire season in 1992 higher than in 2003? Should we expect 2003 to give lower values or can we conclude that MIPAS is biased low compared to other measurements?

A statement will be added in the text that the difference might be due to a MIPAS low bias caused by the spectroscopic data.

Section 2.2: P. 12072, L.5: The typical retrieval error is estimated to 50%: is it only the retrieval error or the uncertainty deduced from the validation exercise, i.e. obtained by comparison with independent measurement?

It is the estimated retrieval error (ex ante estimate). For clarity, we will include the term “estimated” in the paper. This estimated retrieval error proves reasonably consistent with the deviations from independent measurements.

Does the validation include comparisons for 2003 in the Southern Hemisphere?

Yes, it does.

Otherwise, are there ozonesonde measurements in the region of interest that could be used to evaluate O3 retrievals?

First, Southern hemispheric comparisons of 2003 were already included in the Steck et al. paper. Second, comparison of further single co-incidences is not very useful because, given the large random error of single MIPAS ozone profiles in the troposphere, comparison of further single measurements would not provide further

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insight.

Are there issues with the spectroscopic parameters for this molecule as well?

The validation work of Steck et al. do not hint at any spectroscopic problem with ozone spectroscopic data. Certainly, spectroscopic data are never perfect, but spectroscopic data are by no means a driving source of error in case of MIPAS ozone retrievals. The uncertainty of O_3 line intensities is 2–10%, depending on band and rotational quantum numbers. For the transitions used here, the mean error spectroscopic error is estimated at 5% only. We will include this number in the paper.

The last sentence of this section is not clear;

This sentence will be reworded.

Section 3: Section 3.1: The authors chose to run forward trajectories instead of back-trajectories started at the altitude range where the enhancement was found (which is the method usually used in this kind of analysis). However, only biomass burning sources are considered using satellite fire counts. A discussion of the importance and location of other sources would be useful for the discussion and to justify why BB is considered to be the major contribution here.

The choice of forward trajectories was driven by the fact that our analysis is based on data averaged in time. Backward trajectory calculations are appropriate if the measurements can be assigned to exact times. For our mean distributions, which cover approximately a month, there is no obvious way to decide at which time the backward trajectories should be started.

In the revised version we mention the possibility of C_2H_6 sources other than biomass

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burning. We consider biomass burning the major contribution here, because the plume nicely coincides with the biomass burning plume seen be Edwards et al. Furthermore, we consider it very unlikely that the C_2H_6 plume seen by MIPAS would coincide so well with biomass burning trajectory density, if other sources were dominating. Nevertheless, this issue needs further work but this can hardly be achieved with the data available by now.

How was the 12 days limit chosen?

The 12 days limit is long enough to monitor intercontinental transport in the altitude range of interest. In a preliminary study, 8 days 2D-trajectories at 275 hpa already proved sufficient to represent the plumes and to transport polluted air once around the globe in many cases. For the subsequent 3D-trajectory study, where only segments of the trajectories were considered which fell into the altitude region of interest, we decided to have an additional margin of 4 days. Stohl et al. (J. Geophys. Res., 2002) suggest longer times (but 10-15 days still sufficient to monitor intercontinental transport) but these values represent the entire troposphere, which is dominated by lower altitudes with lower windspeeds.

The authors should provide a short discussion of the trajectory uncertainty.

Agreed, will be added.

Section 3.2: How was the avg + 2sigma criterion chosen?

In case of normal distributions this criterion is a traditional one used to decide if the difference of an actual value from the ensemble mean is significant or not. Although we cannot assume a normal distribution here, this criterion is not unreasonable as long as it is not assigned to a particular confidence limit.

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General Comment: In the trajectory calculation of the origin of C2H6 enhancements, the possible mixing of pollution from different origins is not clearly addressed, although one could assume that it will be an important factor for ozone production, especially above the Indian Ocean and downwind. The authors then discuss the differences between Amazonian and African plumes. What is the confidence that there is no South American contribution to the "African plume"? Are they always well separated (not clear from Figure 9)?

Specific comment: The possible mixing of plumes from different origins needs to be discussed here. Can a specific enhancement of C2H6 be attributed to only one source region? Is there no mixing above South Africa and downwind? Did the authors choose to analyze only specific parts which are clearly dominated by specific source regions? Can the authors associate a percent confidence to the plume origin?

Each data bin was assigned to the source of which the sum over related trajectories times residence time within the bin was largest. In this context it should be noted that it is not always mixing in its physical sense if a bin is touched by trajectories of different origin. If the circulation is not stationary, a bin can be crossed by a trajectory of South America on the one day, and by a trajectory of Africa on the other day. In this case the mixing is a purely numerical effect via calculation of time averages of MIPAS data. We will include an explanation in the paper on how we assign the ambiguous bins to the sources and have reworked Figure 9 according to the recommendation of reviewer Dr Chabrillat.

Section 3.3: The second sentence may be confusing as ozone is generated by photo-chemical reaction with two main precursors: VOCs (mainly longer lived species - CO and CH4 - in the remote atmosphere) and NOx, although it depends critically on the level of NOx (NOx-limited and NOx-satured regimes).

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This statement will be corrected. Many thanks for clarification!

In section 3.3 and 3.4, and in the following discussion, the authors refer to Amazonian and African plumes. Again, the question of mixing in the pollution belt should be addressed.

see above.

Section 3.5: Considerations on injection heights could be moved to section 3.1

The injection altitude concept including the reference to Labonne et al. were already introduced in Section 3.1. of the original paper, where the more technical aspects of the trajectory calculations are described. We mention this concept again in Section 3.5. where we discuss the not fully solved problem of uplifting of air, because the injection altitude concept is directly relevant to this issue.

Section 3.5:
The emission ratios could also help understanding the difference in O3/C2H6? According to Andreae and Merlet [2001], C2H6 emission factor is 0.32 $\pm$ 0.16 for savanna/grassland and 0.5-1.9 for Tropical forest (usually higher VOCs EF for tropical forests), and NOx emission factor is 3.9 $\pm$ 2.4 for savanna/grassland and 1.6 $\pm$ 0.7 for Tropical forest. Could O3 production also be more efficient for savanna/grassland?

Our ratios of American and African C_2H_6/trajectory densities are somewhat but insignificantly smaller than that one would expect from the different emission factors. Using C_2H_6 as quantitative plume tracer leads to an overestimate of the Equatorial American plume. Our ozone/C_2H_6 ratios in the Equatorial American and African plumes are consistent with the different ratios of C_2H_6/trajectory.

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Do the altitudes of trajectories show differences in altitude reflecting different residence time in the lower troposphere? Or is it something that would be missed by the trajectories?

We see no systematic difference in the trajectories: both for South America and Africa there exist trajectories where air is immediately uplifted, as well as trajectories where the air resides a few days in the lower troposphere before it is uplifted.

Was there thunderstorm activity during the time period studied?

Over Africa there was more thunderstorm activity than over tropical America. This information will be included in the paper.

Conclusion: The authors could state more precisely how the MIPAS and MOPITT data are complementary.

Agreed; this will be included.

P. 12079, L.10: small scale convective processes are needed but also chemistry. The authors could mention need of chemistry-transport model calculations for further analysis of ozone production within the plume.

Agreed. This will be added.

Interactive comment on Atmos. Chem. Phys. Discuss., 7, 12067, 2007.