

Interactive comment on “Microphysics and heterogeneous chemistry in aircraft plumes – high sensitivity on local meteorology and atmospheric composition” by S. K. Meilinger et al.

S. K. Meilinger et al.

Received and published: 12 November 2004

1) Response to Referee 1:

a) Typos:

The typo in the abstract had already been corrected in the on-line version of the paper.

b) Chemistry on carbonaceous aerosols:

Reactions involving soot particles include a direct ozone destruction reaction, reduction of NO₂ to NO, and reactions involving HNO₃ and SO₂. Laboratory data on reaction rates are available, showing a large spread presumably due to the different chemical nature of the various soot samples used in the experiments. We refrain from considering reactions on soot based on direct evidence obtained from analysis of in situ

measurements of O₃, N₂O, particle surface area, and other quantities in an aircraft plume in the lower stratosphere [Gao et al., 1998]. This analysis provided compelling evidence that even a soot loading (from the Concorde) far above average values of modern aircraft engines is unable to significantly destroy ozone on time scales of hours. The reaction probability decreases thereafter because of ongoing plume dilution. The low reactivity of soot inferred by Gao et al. for ozone also implies that currently debated soot reactions cannot account for ozone trends in the lower stratosphere. Of course, if new evidence for fast reactions on aviation soot in flight conditions arises, this issue must be re-evaluated with a detailed, coupled plume microphysics/chemistry model like the one presented here.

2) Response to Referee 2:

a) Minor Comments:

Following referee 2, we included a reference for aerosol surface area density measurements in p.4457 l.10, namely Kärcher, 1998b, and Schröder, 1998. Following the referee's comment, we changed the arrow on p.4460 l. 14 into "=". The language remarks and typos on p.4465 l.23, p.4466 l.3 and p.4467 l.6 have been corrected accordingly. Thanks for the careful reading of our paper.

b) Gamma values in Kärcher (1997)

The gamma values in the Kärcher (1997) paper are not for a specific species or reaction, but are of general nature. Using calculated surface areas of plume particles and considering plume dilution, Kärcher (1997) derived dimensionless characteristic reaction timescales for emitted species. From these reaction timescales one can conclude, that heterogeneous processing on volatile aerosol and soot can only be efficient on timescales below 1 day if uptake coefficients $>0.003-0.007$. Depending on the lifetime of ice contrails, uptake coefficients >0.1 are required for rapid uptake of exhaust species on the ice particles. These results are of general nature and independent of the exact species or reaction under consideration.

c) Impact of initial HOx:

The model is not particularly sensitive to the initial HOx concentration in the plume (which is due to the very short lifetime of HOx), whereas the results differ depending on the ambient HOx concentrations as discussed in the paper. In general ambient conditions had a higher impact on the results. We also discovered a little error in the initial OH which was chosen to be 10 ppm instead of 6 ppm as indicated before (see p.4460). Sorry about that.

d) Further model documentation:

The model is also documented in Meilinger, S.K., "Heterogeneous Chemistry in the Tropopause Region - Impact of Aircraft Emissions", Dissertation, Diss. ETHZ No. 13819, Suisse Federal Institute of Technology, Zürich, 2000, We included the respective reference on p.4461 l.3. It includes a list of species and reactions in the appendix. Note that reaction rates, however, had been updated as described in the paper.

e) Overcompensation of ozone production:

Concerning the statement on p.4468, l.11, we do not see, why it should be at odds. Comparing the dashed line (background chemistry only) and the solid line (chemistry within the diluting plume) in the lower right panel, one clearly sees, that the solid line is below the dashed one. Additional ozone production due to the emitted NOx would lead to the solid line being above the dashed line. However, this is not what we find. NOx induced ozone production involves HOx. Ozone production in the plume is below background production. This is due to the fact, that the ozone producing effect of emitted NOx is overcompensated by dehydroxylation (not denoxification!!!) due to dehydration via ice formation and heterogeneous loss of HO₂ on ice.

f) Future model applications:

We agree that the next step could be an application of our results in a climatological study using typical meteorological conditions from the tropopause region. Our model

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is an adequate tool to perform such studies in the future. It also would be interesting to see how using such results as an input for global models would influence the results of aviation related global impact assessments.

Interactive comment on Atmos. Chem. Phys. Discuss., 4, 4455, 2004.

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