Atmos. Chem. Phys. Discuss., 12, C11852–C11855, 2013
www.atmos-chem-phys-discuss.net/12/C11852/2013/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "HO_x measurements in the summertime upper troposphere over Europe: a comparison of observations to a box model and a 3-D model" *by* E. Regelin et al.

Anonymous Referee #1

Received and published: 21 January 2013

This is a study of HOx chemistry in the upper troposphere aboard the Learjet at altitudes up to 9 km over Northern Europe, and represents an important addition to the very limited dataset in this region, where levels of VOCs are low and there are a limited number of HOx sources and sinks. This region is typically dry and so the reaction of O(1D) with water vapour is less important, and peroxides and carbonyl species are more important, and sometime dominant. Also NOx levels are low and so the dominant HOx sink is peroxy radical self and cross-reactions.

In this work Regelin et al compare OH and HO2 measurements, which are made using a laser induced fluorescence instrument housed in a wingpod of the Learjet, with

Full Screen / Esc
Printer-friendly Version
Interactive Discussion
Discussion Paper

calculations from a highly constrained box model (CABBA) and an unconstrained 3D global model. There is good agreement with a slope close to 1 for the box model and high correlation factor. In contrast, there is poorer agreement with a global circulation model (GCM) (EMAC) that uses the same chemical mechanism, with some quite large measured to modelled ratios. This demonstrates that there is a difficulty with using an unconstrained GCM even when the chemistry is validated via the constrained box model. H2O2 and NO (which are also measured on the aircraft) cannot be calculated accurately by the GCM either – and using the modelled values for these is stated to be responsible for the errors in HOx. It is suggested that small scale meteorological events (e.g. convection which is prevalent in N Europe at that time of year) and micro-physical processes for wet scavenging are not captured well by the GCM, leading to the errors in H2O2 and NO, with the knock on effect for the calculation of HOx. There is usually a correlation between the ability of the GCM to calculate H2O2 and NO accurately, and the ability of the CTM to get HOx right.

The aircraft is well equipped with state of the art methods for the determination of HOx and supporting parameters, including HOx sources: J (NO2), H2O, H2O2, O3, NO, organic peroxides and sinks : CO, CH4, HCHO. Many of these were real time in situ (rather than off-line using canisters). The HOx instrument has been described in detail in a previous paper (except for aspects of the calibration, notable the temperature dependence, which is very important, see below). CFD calculations have been performed to show velocity field between inlet nozzle and detection chambers in order to visualise the flow and account for radical losses. The conclusison is that there is no wall loss in the inlet tube but some further downstream. The values reported as very heavily dependent upon the calibration of the instrument, which is difficult to perform as a function of pressure and temperature.

The authors should address the following points.

Specific questions/comments

ACPD

12, C11852–C11855, 2013

Page 30626. Line 1. It is not clear how the pressure dependent calibrations are performed. Were different pinhole diameters used to simulate the change in pressure in the cell owing to sampling at different altitudes? Or was a constant size pinhole used with a varying pumping speed. This procedure, even if it is described elsewhere, needs some attention there as the pressure dependence is critical. Is the assumption made that or sampling pressures (different altitudes) any losses of HOx on the sampling assembly is the same? A way around this assumption is to perform an in flight calibration, even being able to see a relative change in sensitivity with altitude is useful. In the GABRIEL paper an in flight calibration was discussed for this instrument (but not here).

Page 30626, line 12. Is there a reason why a significant H2O dependency in the calibration was not seen in HOOVER compared with GABRIEL? It is a very similar instrument?

Page 30626, line 12. It is interesting that the CFD calculations show no wall contact until after the OH cell when the jet broadens and there is wall loss prior to the HO2 detection cell. Extending the calibration to different temperatures is an important landmark, as this is not typically reported. Although the ambient temperatures varies quite a bit, the temperature within the instrument sampling tube within the wingpod varies much less. It is interesting that the OH sensitivity did not really change much with lowering the temperature (a small increase was seen) but it dropped quite a bit for HO2 (about a factor of 2 , which is a lot considering that the reported concentrations will directly mimic this). It is interesting that it is thought that the cooling down of the calibration tube or the electronics. I think there will be uncertainties in translating this dependence found in the laboratory to those that may prevail on the aircraft . However, the fact that a temperature dependent calibration has been performed on the ground is a step forward, but further studies of the temperature dependence are needed, particularly in flight.

Page 30632. For INTEX-A where the model underestimates HO2 by up to a factor

ACPD

12, C11852–C11855, 2013

Interactive Comment

of 3 where there is a higher load of pollutants. Is this due to an interference for this instrument, was HO2* being measured which is significantly more than HO2 (as in the model)?

Fig 5. In order to calculate the flux some assumption needed to be made about the average photolysis time in the flow-reaction calibrator. What was this time?

Fig 6. Say how the pressure was varied (changing pinhole, or same pinhole with different pumping speed?)

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 30619, 2012.

ACPD

12, C11852–C11855, 2013

Interactive Comment