Atmos. Chem. Phys. Discuss., 11, C1978–C1983, 2011
www.atmos-chem-phys-discuss.net/11/C1978/2011/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Trends of HCl, $ClONO_2$ and HF column abundances from ground-based FTIR measurements in Kiruna (Sweden) in comparison with KASIMA model calculations" *by* R. Kohlhepp et al.

R. Kohlhepp et al.

regina.kohlhepp@kit.edu

Received and published: 18 April 2011

Answer to the report of anonymous referee # 2

We thank referee # 2 for his/her constructive comments regarding our manuscript. In the following, citations from the referee report are written in italics.

- *page 1491, introduction: To put things in perspective it would be appropriate to*

mention the long series of measurements carried out at the Jungfraujoch alpine station with the same type of instrument as used here (+ relevant references).
The sentence (or rather paragraph) starting on page 1491, line 24, was changed to: "In this paper, measurements of vertical columns of HCl, $ClONO_2$ and HF with a Fourier transform infrared (FTIR) spectrometer at Kiruna (Northern Sweden, 67.84° N, 20.41° E, 419 m asl) are presented. They are compared with results from the atmospheric chemistry transport model (CTM) KASIMA (KArlsruhe SImulation model of the Middle Atmosphere) and also with the longest European FTIR time series recorded on Jungfraujoch (Swiss Alps, 46.5°N, 8.0°E, 3580 m asl) (see e.g. Zander et al., 2008). The key question addressed is whether the expected decrease of chlorine species can already be seen significantly in the vertical column abundances of HCl and $ClONO_2$ above Kiruna and how the HF abundances develop."
Zander et al. (2008) was added to the bibliography as a reference.

- *page 1491, line 19: Please mention the average stratospheric lifetime of HF.*
At stratospheric conditions HF does not react with most of the stratospheric trace gases. At room temperature just the reaction with $O(^1D)$ with two branches (either quenching $O(^1D)$ to $O(^3P)$ or forming FO and H) is known. By assuming the temperature-independent rate coefficient of 1.5×10^{-11} cm^3/s [R. Atkinson, D. L. Baulch, R. A. Cox, J. N. Crowley, R. F. Hampson, R. G. Hynes, M. E. Jenkin, M. J. Rossi, and J. Troe, Evaluated kinetic and photochemical data for atmospheric chemistry: Volume III – gas phase reactions of inorganic halogens, Atmos. Chem. Phys., 7, 981-1191, 2007] (please note that Sander et al., Chemical Kinetics and Photochemical Data for Use in Stratospheric Modeling, Evaluation Number 15," JPL Publication 06-2, Jet Propulsion Laboratory, California Institute of Technology, Pasadena, CA, 2006, state that this reaction branch is considered to be unimportant) for the H and FO forming branch the stratospheric lifetime of HF is in the order of $>$10 years. Thus, HF can be considered to be an inert

ACPD
11, C1978–C1983, 2011

tracer in the stratosphere. The sentence in the manuscript was changed to: “The stratospheric lifetime of HF is more than ten years so that it can be treated as an inert tracer there (see also Chipperfield et al.,1997).”

- *page 1493, line 11-21: FTIR measurements are much less sensitive in the troposphere than they are in the stratosphere, as shown in Figure 1. The proper way of doing the observation/model comparison would be to convolve the Kasima profiles by the sensitivities plotted in Figure 1. However, I understand from what is said in page 1498 (line 15) that the troposphere is completely ignored below 7 km in the model. This is a source of error that should be quantified (for instance by assuming a constant mixing ratio between the ground and 7 km, and by convolving by the FTIR vertical sensitivity).*
  Please also see the answer to the comment of referee # 1 concerning the same topic. Assuming constant volume mixing ratios between 0 and 7 km of 50 pptv for HCl, 20 pptv for HF, and 1 pptv for $ClONO_2$ as an upper approximation, the contribution of this altitude range over Kiruna is in the order of about 6×10^{14} molec/cm^2 for HCl, 1×10^{13} molec/cm^2 for $ClONO_2$, and 2×10^{14} molec/cm^2 of HF representing about 10% in the maximum with respect to the total column of these substances.

- *page 1495, line 5: Please rephrase the first sentence, which is unclear. What are these "relaxation terms"? Is this an experiment nudged towards the ECMWF analysis ? but then, what does the 18 km-altitude refer to? this whole paragraph is rather confusing. Also, an important missing information is the scenario of emission of long-lived species (including chlorine- and fluorine-bearing species) which has been used in the Kasima simulation. Long-term series of measurements of HCl and $ClONO_2$ provide a very good test of the validity of scenarios of emissions. It is therefore important to describe precisely the scenario used in this study.*
  We have rephrased the complete paragraph:

ACPD
11, C1978–C1983, 2011

"For the present study, the KASIMA version as described in Reddmann et al. (2001) which yields realistic stratospheric age of air values (Stiller et al., 2008) is applied. The necessary meteorological data of temperature, vorticity and divergence are taken from the European Centre for Medium-Range Weather Forecast (ECMWF), using ERA-40 data until 2002 and operational ECMWF analyses from 2003 on. In this version, the KASIMA model is relaxed (nudged) toward the ECMWF data between 18 and 48 km pressure altitude using forcing terms with a timescale of 4 hours. Below 18 km, the meteorology is based on ECMWF analyses without nudging, and above 48 km pressure altitude, the prognostic model integrating the primitive equations without additional forcing from ECMWF data is used. The model consists of 63 vertical layers between 7 and 120 km and has a horizontal resolution of approximately 5.6×5.6 degrees (T21).

The time evolution of the global surface volume mixing ratios of the ozone-depleting substances is prescribed at the lower model boundary according to the so-called baseline scenario Ab which is a best-guess scenario following the Beijing Amendments in 1999 of the Montreal protocol (for more information on the scenario see chapter 1 of the UNEP/WMO Scientific Assessment of Ozone Depletion: 2002). These data which were recommended to be used as lower boundary conditions for the WMO Ozone assessment 2007 are provided in the framework of the SPARC (Stratospheric Processes And their Role in Climate Change) CCMVAL (Chemistry-Climate Model Validation activity) initiative.

The photolysis rates are calculated online in KASIMA using the Fast-J2 model of Bian and Prather (2002).

For the comparison of the KASIMA model data with the FTIR measurements, the model data were interpolated to the location of Kiruna from the circumjacent grid points."

- *page 1495, paragraph 5.1: It would be useful to the reader to explain in a few sentences what is the origin of the strong seasonal cycle of HF.*

ACPD
11, C1978–C1983, 2011

A new reference was added: Duchatelet et al., 2010.
The following text was added: "At the International Scientific Station of the Jungfraujoch (Swiss Alps, 46.5°N, 8.0°E, 3580 m asl), the seasonal cycle of HF measured by the FTIR instrument there was investigated (Duchatelet et al., 2010). Especially the seasonal cycle of the lower stratosphere partial columns and of the total column abundances were found to be anti-correlated with tropopause height, which is due to the high stability of HF. This is why we assume that the main characteristics of the seasonal cycle of the HF total column abundances above Kiruna result both from the varying tropopause height and from the polar vortex."

- *page 1496, line 13: Why are the months of June and July not included in the so-called "summer" period ?*
  Please see the answer to the comment of Reviewer # 1 on the same topic.

- *page 1497, third paragraph: Why is the trend for $ClONO_2$ so much larger than that of HCl ? how does this compare to similar measurements performed at Jungfraujoch for instance ?*
  The following paragraph was added: "We have not yet found an explanation for the different trends of HCl and $ClONO_2$ above Kiruna. A comparison of FTIR and modelled total column abundances of HCl, $ClONO_2$ and HF and their temporal development at 17 measurement sites around the world belonging to the NDACC is in preparation at the moment (Kohlhepp et al., 2011). At some other northern polar FTIR sites, the signal seems to be similar to the one at Kiruna described here, both for models and measurements. For HCl, measurements from the midlatitude site on Jungfraujoch show a trend of (-0.87 ± 0.10)%/yr between 1996 and 2009 (Mahieu et al., 2010), which very well matches the HCl trend at Kiruna presented above. In contrast, the $ClONO_2$ trend at Jungfraujoch amounts to (-0.90 ± 0.27)%/yr. It is thus much weaker than the one at Kiruna and agrees with the HCl one within errors. Mahieu et al. (2010) also compared their FTIR

ACPD
11, C1978–C1983, 2011

data with KASIMA model results when sampled like the measurements. KASIMA simulates a relative $ClONO_2$ decrease of (-0.72 ± 0.21)%/yr for Jungfraujoch that is about a factor of two stronger than the HCl one of (-0.29 ± 0.11)%/yr."
Mahieu et al. (2010) was added in the bibliography.

Other changes

- In the acknowledgements, the following sentence was added: "We acknowledge support by Deutsche Forschungsgemeinschaft and Open Access Publishing Fund of Karlsruhe Institute of Technology."
- The tables 1 and 2 were changed slightly: In the captions, we added the sentence "The time range considered is 1996–2009 (unless otherwise identified)." and removed the column containing the time ranges because especially for summer/autumn (June–November), the notation was a little confusing.

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 1489, 2011.

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