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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Observed and simulated time evolution of HCl, $ClONO_2$, and HF total column abundances" *by* R. Kohlhepp et al.

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Answer to the report of anonymous referee # 2

We thank referee # 2 for his/her constructive comments regarding our manuscript. We feel both referee reports were really very helpful to improve the paper. In the following, citations from the referee report are written in italics.

General comments:

- *An almost identical analysis has already been published in Chapter 6 of the SPARC Report on the Evaluation of Chemistry Climate Models, June 2010. In*

this report, the observations were used to evaluate various CCMs. For HCl and ClONO2 the SOCOL, EMAC, and SLIMCAT models did not do as well as other models presented in that study. For instance, the report states that SOCOL overestimates HCL in the 500-600K region. No mention of the results from the SPARC report is made in this paper which is somewhat egregious considering the comparisons and analysis are so similar. In fact, the SPARC report does a better job of describing issues pertaining to the models. If the model configurations presented in this paper differ significantly from those in the SPARC report or the conclusions the authors infer from their model efforts are different then some comment needs to be made regarding this to differentiate this work from SPARC. If the result presented here are similar to SPARC then that needs to be stated as well. We added the following sentences to the introduction, page 32089, line 17: "In the framework of the SPARC CCMVal (2010) initiative, a comparison between different chemistry-climate models (CCMs) was performed for HCl and $ClONO_2$. The two CCMs taking part in the present study, EMAC and SOCOL, were also involved in this activity. The CCM calculations were compared with FTIR measurements above the Jungfraujoch and with satellite data sets with respect to the mean annual cycle, mean profiles and total column abundances. So the present study extends the SPARC CCMVal (2010) comparison with respect to additional geolocations and compares the CCM results of EMAC and SOCOL with those of other kinds of models (a 2-D model and two chemistry-transport models, CTMs)."

- *While this paper provides a good synopsis of the ground based measurements, it is not clear what scientific questions are being answered here or how this work relates to some of the larger issues being discussed in our field. For instance, the authors report a decrease in HCl and ClONO2 during the period of study, as expected. Do these results also agree with reports of the initial recovery of stratospheric ozone? This brings up another point, which is that the analysis in the paper would be greatly enhanced through the use of other available data*

sets, such as those from the satellites (SAGE I/II, HALOE, etc.). For instance, Newchurch et al., 2003 compare the trend in satellite observations of ozone to HCl and conclude that stratospheric ozone loss is decreasing. Further confirmation (or otherwise!) from ground based observations of the Newchurch et al., study would be very valuable to the community.
In the introduction, we added the following sentence on page 32089, line 17: "Later studies confirmed stratospheric HCl or total Cl_y to have reached a plateau at the end of the 1990s and to be decreasing since (e.g., Newchurch et al., 2003; Froidevaux et al., 2006; Lary et al., 2007)."
To the discussion, the following paragraph was added: "The present study is also able to confirm the results of many other preceding investigations on stratospheric inorganic chlorine and fluorine measurements. The stabilisation of the stratospheric HCl content at the end of the 1990s described in Sect. 3.1 was already seen for example by Considine et al. (1999), Rinsland et al. (2003), and Newchurch et al. (2003). Also the subsequent negative trend in the stratospheric chlorine abundance was reported before by other studies. For example, Lary et al. (2007) found a decrease towards the end of their study in which they analysed a stratospheric Cl_y time series between 1991 and 2006 that was created from a combination of many different (mainly satellite) measurements. For HCl, a trend was estimated from measurements made by the MLS (Microwave Limb Sounder) instrument aboard the Aura satellite between 50 and 65 km height and 60°S and 60°N by Froidevaux et al. (2006). They reported a decrease in the volume mixing ratio of (-0.78±0.08)%/yr between August 2004 and January 2006. This value also agrees very well with those found in the present study. A similar result for HCl was published by Jones et al. (2011) who combined HALOE (Halogen Occultation Experiment) data with ACE-FTS (Atmospheric Chemistry Experiment Fourier Transform Spectrometer) results between 35 and 45 km to form a time series of HCl from 1993 to 2008. They found a significantly negative trend of about -5.1%/decade to -5.8%/decade for the time period 1997–2008,

depending on latitude. Measurements with the McMath-Pierce solar telescope on Kitt Peak (Arizona, U.S.) showed a slightly larger decrease of the HCl total column abundance of (-1.8±0.4)%/yr between 1997 and 2007 (Wallace and Livingston, 2007). Concerning the HF total column abundance, a strong increase of (10.9±1.1)%/yr above Kitt Peak between 1977 and 1990 was reported by Rinsland et al. (1991). It weakened during the 1990s so that the trend for the period 1977–2001 amounted to (4.30±0.15)%/yr only (Rinsland et al., 2002). A leveling-off could be seen by Zander et al. (2008) above Jungfraujoch around 2003–2004. This agrees very well with the results of the present study at some of the northern hemisphere sites, where the HF trends are much weaker for the period 2004–2009 than for 2000–2009 and 1996–2009, or even negative (Sect. 4.1.2)."
In this context, also the order of the paragraphs in the discussion section was changed.

Specific comments: Model analysis

- *The models presented here are used for comparison to the observations and the authors do a good job of describing the differences in trends between model and measurements. It is a little confusing as to whether the measurements are being used to validate the models or vice versa, though. Please provide a clear description of purpose as to why the models are being used in this study and why these particular models are best suited for this work. It is also not clear why multiple models are needed. If one particular model has shortcomings then why is it used for this effort? What insight into the atmosphere is gained by having multiple models?*
  You are right, this was a little confusing. We tried to improve the reasoning as to why models and especially those were used, and to the purpose of the paper in general. Please see for example the answer to the comments of referee 1 concerning page 32087, second paragraph, or concerning page 32098, line 22.

We do not really expect to get much more insight into the atmosphere by having multiple models, but maybe something can be learned about the models. Through the direct comparison with other models and the measurements, it can be detected whether a particular model has shortcomings in the first place.

- *There is little discussion as to why the model trends differ from the observations. Do the models not represent the chemical or dynamic state of the atmosphere accurately enough? The authors state on page 32099, Line1 "These different types of models can help to evaluate the contributions of dynamics and chemistry to observed trends" but do not really use the models to determine these contributions.*
  The sentence you mentioned was removed when dealing with the comment of referee 1 concerning page 32098, line 22.
  Our answer to the comment of referee 1 concerning page 32117, lines 14–19 tries to answer part of the question concerning the difference between modelled and observed trends.

- *The models are all using outdated chemical kinetics, with some models still using recommendations from JPL 2002. The JPL 2010 compendium has been available since June 2011. While it is unknown whether updating the chemical kinetics will change the results, the use of the older recommendations leaves some question as to the validity of the model analysis.*
  The JPL (2010) compendium was published only very shortly before the present paper was submitted in Sept. 2011. So it would not have been possible to use the new recommendations, especially because the model simulations take a very long time. This was also the reason why already existing CCM climate simulations were used in this comparison. The EMAC (and SOCOL) data stem from the mentioned SPARC CCMVal comparison. In order to ease the comparisons, the CTMs KASIMA and SLIMCAT used the same JPL (2002) recommendations.

- *Please provide a brief description of the bootstrapping method then refer to Gardiner et al., 2008 for more information. Some readers may not be familiar with this method.*
  We did this, please see the answer to the comment of referee 1 concerning page 32106.

- *The models use boundary conditions from different studies, EMAC considers the IPCC A1B scenario and SOCOL considers the CCMVal2 REF-B1 scenario. How do these scenarios differ and what effect will this have on the final model output?*
  The scenarios do not differ strongly, but all CCMVal greenhouse gas scenarios base on the IPCC A1B scenario. The only difference can be found in CH_4, which has been changed from the middle of 2002 on in the Ref1 data according to newer measurements. So only SOCOL differs slightly from the other models, for the last 2.5 years of its simulation only, which is not expected to have a strong influence on the results presented here.
  The difference between the halocarbon scenarios is discussed in more detail now, please see the comment of referee 1 concerning page 32117, lines 14–19.

Specific comments: Instruments

- *While the differences between instruments may be obvious to the authors and other experimentalists (i.e. A Bruker 120HR vs. a Bruker I20M FTS) it is not obvious to others in the community, such as modelers. If it is important enough to mention the different instruments used in this study, it is important then to describe salient differences between these instruments and how this will affect observations.*
  The following explanation was added on page 32092, line 4: "Different kinds of FTIR instruments have been used for the measurements analysed in this study (please see the description of the sites below), called Bruker 120M, 120HR, and 125HR, and Bomem DA8. The instrumental differences between the Bruker

spectrometers are small, especially between 120HR and 125HR. The latter is the newer version with improved electronics which in the end helps to reduce the noise in the spectra. The 120M instrument is the mobile version which is therefore smaller and more compact than the 120HR. In general, it is more difficult to adjust the 120M than the 120HR spectrometer, which may lead to a slightly worse ILS of the 120M. However, this would affect mostly the profile retrievals, not the total column abundances dealt with in this study. So there are no significant discrepancies expected between the different Bruker instruments. Furthermore, when a new instrument was installed at one site, if possible, an intercomparison was performed with the old one so that the here presented data sets can be assumed to show self-consistent time series. The discrepancies between a Bruker 125HR and a Bomem DA8 instrument were investigated in detail for the total column abundances of the three gases analysed here by e.g. Batchelor et al. (2010) and were found to amount to less than 3.5%."

- *For the comparisons between observational sites closer to the poles a major concern is whether the models can reproduce the polar vortex well. If they are not able to reproduce the extent of chlorine activation and denitrification then comparisons to the ground based data are problematic. Was any screening done to make sure the model and instruments were "seeing" the same atmosphere?*
  No, it wasn't. But the focus of the study is on the trend analysis, and even if a model does not reproduce the polar vortex well, this should not have a significant influence on the resulting trend. A trend only documents the mean change over a larger time range, so a single year when the polar vortex is "wrong" in a model should not have a strong influence. And even if the model is not able to reproduce some important vortex feature above a site every year, the mean change over many years could correspond to the measurements, too. Such an error in a model would maybe be visible in the mean monthly means (Figs. 6–8).

- *The authors state that NCEP analysis fields of temperature and pressure are*

used for the instrument retrievals yet ECMWF met fields are used in the KASIMA and SLIMCAT models. No mention is made as to whether the EMAC and SOCOL models use assimilated winds. Can ECMWF met fields be used for the observational retrievals? Does the use of NCEP instead of ECMWF lead to some of the differences between model and observations?
In the changes following the comment of referee 1 concerning page 32098, line 22, the information was now added that the EMAC and SOCOL runs did not use any meteorological reanalyses, but are freely running and thereby creating their own atmospheric dynamics which (probably) does not correspond to the real state of the atmosphere on a specific day.
We think the difference between NCEP and ECMWF does not lead to a significant difference between measurements and models. Comparisons have been done, but not published, which showed that there is no significant difference in the FTIR results when using NCEP or ECMWF data. Furthermore, for every gas, the spectral microwindows and the lines where the inversion is performed are selected such as to be the least possible temperature dependent. The NCEP data were chosen to be used for the FTIR measurements primarily because they are very easily accessible through the Goddard Automailer system, while it is more difficult to obtain a permission to use ECMWF data.

Technical corrections:

- *JPL 2002, 2006, IUPAC 2004, 2005 are referenced in Table 5, yet there are no corresponding references to Atkinson or Sander in the references section.*
  The references were added.

- *Tables 7, 8, and 9. It would be useful to provide another column that contains the average model trend to give the reader an overall impression of how well the models are doing.*
  We prefer not to do that for two reasons. Firstly, not all models cover the same

time range so that averaging the trends would pretend they are exactly comparable. Especially the SOCOL trends which often differ from the other model results, but only cover 2000–2004, would have a strong impact on the resulting average. In addition, technically, it would be very difficult or impossible to include another column in these tables.

- *Page 32088, Line 18. . . I believe it should be "the World Meteorological Organization".*
  Was changed.
- *Page 32101, Line 23 Define MECCA.*
  Was done.

Further changes:

- The sentence on page 32089, lines 6–9 was very long and therefore split into two so that it now reads: "Due to its long stratospheric lifetime, fluorine, and in particular HF, is not involved in catalytic ozone destruction. It is often used as a tracer for stratospheric dynamics and transport, and hence as a reference for chemically more active trace gases like HCl (Chipperfield et al., 1997)."
- On page 32093, line 7, the name "Spitsbergen" was changed into "Svalbard".
- On page 32112, line 9, the sentence was extended by: "[...], following the conclusions of Sect. 5.5."
- The sentence on page 32116, lines 6–8 was changed to: "This was already described by Kohlhepp et al. (2011) for Kiruna and in the SPARC CCMVal (2010) report for the Jungfraujoch. In Kiruna, the trends between 1996 and 2009 of these two gases differ by about a factor of four both for the FTIR measurements and the KASIMA model calculations (Kohlhepp et al., 2011)."

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 32085, 2011.