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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 # 1

We thank referee # 1 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:

- *Unfortunately the authors do not really motivate the intention and importance of their study. Many questions remain open: What is the intention of this study? Why*

is the comparison with the models done? Why are the observations alone not sufficient? What is the outcome for the community? Though some are answered somewhere in the paper they are not answered in the important parts of the paper like abstract, introduction or conclusion and some answers are definitely missing. Should this paper be a scientific one or a technical one? The paper begins with a lot of technical details but then turns into a more scientific paper at the end. What definitely is missing is a discussion about the trend analyses method applied in this study. Previous results of this method or trend analyses of these species (except the Rinsland et al. paper) are not mentioned or discussed at all. How should one judge how reliable the results are if the method description and discussion is missing? I would strongly recommend to include this.
We tried to improve the points you made. Please see the answers to your specific comments for the changes we made.
Concerning the comparison with other studies, please see our answer to the second “General comment” of referee 2.

Technical comments:

- *The paper is structured to complicated. There are too many splits in subsections. I would suggest to split section two into two sections, one for the instruments and one for the model descriptions and then skip all the sub-sub sections x.x.x. The section should also be renamed. "Instrumentation" is more adequate than "Observations" since not the observations are described but the instruments. The model description can be named "Models" as done in section 2.2. or "Model Set-up". Section 4: Include section 4.1 into section 4 and then start numbering with 4.1 and so on instead of 4.1.1. etc. Paragraphs: They are sometimes very long and sometimes very short paragraphs (consisting of e.g. only one sentence). This should also be improved. Description of the figures: It would be worth to mention more on what they show in the text (e.g. figures 3-5).*

ACPD
11, C16053–C16074, 2012

You are right, the structure was complicated. We tried to improve that by following your suggestion concerning the splitting of the instruments and models description section. Moreover, we separated the trends section into two sections. The first one now describes the method and the trend sensitivity, while the second one contains the trend results. In this context, the paragraphs on page 32106, lines 26–28, and on page 32107, lines 2–5, were removed and combined into the description of the new Sect. 5 that reads: "In this section, a short description of the trend calculation method is given, especially also of the bootstrap method used to estimate the trend uncertainty. In addition, we investigate the dependence of the trend result on the type of fit function, the time period, and on sampling. The first two influencing factors are analysed using the FTIR measurements, while the sampling influence can be estimated with the help of the CTM data. The main time period chosen for the trend and its sensitivity investigations is 2000–2009. This was done because a continuous decrease is expected for the chlorine gases and an increase for HF during this time so that a linear trend fit can be assumed to sufficiently represent the temporal evolution of the total column abundances. Moreover, nearly all sites included in this study measured during this time (see Table 1)."
In this context, also the description at the end of the introduction was changed slightly. Specifically, the sentence that started on page 32090, line 4 now reads: "The trends of HCl, $ClONO_2$, and HF are presented in Sect. 6, but before, an investigation of the dependency of the trend results on some influencing factors is made (Sect. 5)."

- *Tables: Tables 2-4 are of technical nature and could be provided as a supplement rather than in the paper itself. This depends however on the intention of the study. Should this be a technical paper or a scientific paper?*
  We did put these tables in a supplement. Please see the answer to the comment concerning page 32092, line 14.

ACPD
11, C16053–C16074, 2012

- *The title is very general. Why not already mentioning there that the focus of this study is on the trend and seasonal cycle of the species? Further, the numbering of the author's affiliations is somewhat awkward. After e.g. 5 follows 15 and 17.*
  The expression "time evolution" in the title is supposed to suggest that we are analysing longer time series, with respect to trends. The focus of the paper in our opinion should be on the trend and the temporal evolution, this is why we did not mention the seasonal cycles in the title.
  The numbering of the authors' affiliations was changed.

- *Figures in general: From the descriptions in the text it would be more wise to sort the figures by species instead of what is shown (trend or seasonal cycle). As it now Fig. 7 is discussed before Fig. 5 and 6.*
  You are right, from the discussion in the text, we should sort the figures differently. But for the general structure of the paper, we think it is more logical, and also easier to find a certain figure, and to compare the different gases, if they are ordered like they are now.

Specific comments:

- *p32087, l6: Are these coordinates the coordinates were measurements are available or have these coordinates been chosen?*
  These are the coordinates of the northernmost site taking part in the study, Eureka, and of the southernmost one, Arrival Heights.

- *p32087, l8: Try do avoid line breaking of model names.* Ok.

- *p32087, l12: Why has this time period been chosen?*
  The following sentence was added in line 12 of this page (and the following one was slightly changed): "This period is chosen because from most of the mea-

ACPD
11, C16053–C16074, 2012

surement sites taking part in this study, data are available during these years. The precision of the trends..."

- *p32087, second paragraph: Why has this comparison be done? What is the motivation? Is this comparison intended to evaluate the models? Why have these models been chosen?*
  Following this comment, the abstract was slightly changed, also the order of some paragraphs. The paragraph in lines 16–21 was removed and the content was included in a sentence that was added in line 6: "By providing such a near-global overview on ground-based measurements of the two major stratospheric chlorine reservoir species, HCl and $ClONO_2$, the present study is able to confirm the decrease of the atmospheric inorganic chlorine abundance during the last few years. This decrease is expected following the 1987 Montreal Protocol and its amendments and adjustments, where restrictions and a subsequent phase-out of the prominent anthropogenic chlorine source gases (solvents, chlorofluorocarbons) were agreed upon to enable a stabilisation and recovery of the stratospheric ozone layer. The atmospheric fluorine content is expected to be influenced by the Montreal Protocol, too, because most of the banned anthropogenic gases also represent important fluorine sources. But many of the substitutes to the banned gases also contain fluorine so that the HF total column abundance is expected to have continued to increase during the last few years."
  The following sentences were added in line 9: "Thereby, the ability of the models to reproduce the absolute total column amounts, the seasonal cycles, and the temporal evolution found in the FTIR measurements is investigated and intercompared. This is especially interesting because the models have different architectures."

- *p32087, l13: What method is this? Please add a short description.*
  A short description of the bootstrap method was now added in Sect. 5 (old Sect. 4). Please see the comment concerning page 32106. We feel that it would

ACPD
11, C16053–C16074, 2012

be difficult to include an even shorter description in the abstract because the method cannot be explained with only one or two sentences. However, writing more would be inappropriate for the abstract, especially because it is only the method to determine the error bars, not to determine the trends. To clarify a little that the expression "trend determination" in this study is used for a fit of a linear function, the word "linear" was added at the beginning of the sentence starting in line 11, page 32087. (In other studies, this expression is used to only describe the time evolution qualitatively, for example by Rinsland et al. (2003).)

- *p32087, l18: "restricted"? I remember even a phase-out of these species was reached.*
  This comment was considered in the answer to the 4th comment already (concerning page 32087, second paragraph).

- *p32087, l24: Why is HF still increasing while the other species are decreasing?*
  This comment was also considered in the answer to the 4th comment already: Because it is contained in the CFC substitutes as well.

- *p32088, l12-17: This sentence is quite long and should be (if possible) split into two sentences.*
  It was split and now reads: "In the 1970s and 1980s, the emission of anthropogenic halogenated source gases characterised by a strong ozone depletion potential (ODP) increased massively. So in order to stabilise the stratospheric ozone layer and enable its recovery, the Montreal Protocol and its amendments and adjustments have been progressively implemented to reduce or even stop the production and release of the important chlorinated source gases."

- *p32088, l18: It should be added which species are considered as CCly and CFy. For CCly it is done a few lines below, but that sentence should be moved up.*
  This was done now, please see the comment concerning page 32117, lines 14–19.

ACPD
11, C16053–C16074, 2012

- *p32088, l20-24: This sentence is also somewhat too long and should if possible split into two sentences.*
  This sentence was changed when considering the comment concerning 32117, lines 14–19.

- *p32089, l2: Why "i.e."? Shouldn't it be a clear definition? Are there more species than these three considered as ClOx?*
  The expression "i.e." (= "id est") in our opinion is a clear definition, because the translation is "this is".

- *p32089, l16: Call it by it's name: The Montreal protocol and its amendments.*
  Was changed.

- *p32089, l17: It is not clear what exactly Rinsland et al. did and what data they used in their study. Did they only use measurements? What kind of measurements data did they use? Where these also FTIR measurements from NDACC? Why using model simulations in this study then Rinsland et al. used only measurements? What is the role of the models in both studies?*
  The whole paragraph from line 17 to 28 was rewritten to: "In the investigation by Rinsland et al. (2003), time series of HCl and $ClONO_2$ from Fourier transform infrared (FTIR) total column measurements at 9 stations belonging to the Network for the Detection of Atmospheric Composition Change (NDACC) until 2001 were compared with HALOE data at 55 km and calculations from a 2-D model. The measurements agreed on a stabilisation of the stratospheric inorganic chlorine content so that Rinsland et al. (2003) were able to confirm the effectiveness of the Montreal Protocol and amendments. The FTIR measurements within NDACC have been continued until present, and more stations have joined the network. Therefore, the present study is able to continue and extend the investigations of Rinsland et al. (2003) by reporting measurements at 17 sites until the end of 2009. The primary question addressed is whether the expected decrease of the

ACPD
11, C16053–C16074, 2012

total global inorganic chlorine abundance can be confirmed now with the FTIR measurements. Furthermore, five atmospheric chemistry models were included in the study in order to investigate and inter-compare their ability to reproduce the HCl, $ClONO_2$, and HF total column amounts, their seasonal cycles and their temporal evolution, as measured by the FTIR instruments.
This paper does not aim at explaining in detail differences between models or between models and measurements. Such an analysis requires much more detailed investigations, which are beyond the scope of this study. Instead, it intends to show the global inorganic chlorine decrease seen in the FTIR measurements at 17 NDACC sites, the increase in HF, and to analyse the overall ability of different state-of-the-art atmospheric chemistry models to reproduce these measurements."

- *p32089, l21: write " by the Montreal protocol and its amendments".*
  Was removed when changing the paragraph.

- *p32089, general: It is not clear what the intention of this study is. As it is written now it sounds like that for comparing to the Rinsland et al. study measurements would be enough. Why is then the comparison to models performed? Why have these different kind of models been chosen?*
  This comment is hopefully already answered by the change of the paragraph from line 17 to 28, please see above.

- *p32090, l10: In this chapter rather a technical description of the instruments than a description of the observations is given. I would suggest to rename this section.*
  The title was changed to "Instrumentation".

- *p32090, l12: Please add a short description on what a FTIR spectrometer is, i.e. what does the abbreviation FTIR stand for.*
  Was done.

ACPD
11, C16053–C16074, 2012

- *p32092, l13: I would suggest to write "in this study" rather than "here".*
  Was changed.

- *p32092, l14: Tables 2 to 4 consists of a lot of technical details which could be given in a supplement to the paper rather than in the manuscript itself.*
  This was done. In this context, the following changes had to be made:
  On page 32092, line 1, the sentence was changed to: "The biases between the use of the HITRAN 1992 spectral database (for analysis of the DA8 data) and HITRAN 2004 (for analysis of the 125HR data) have been quantified by Fast et al. (2011) based on retrievals from DA8 spectra recorded for one day and for the microwindows indicated for the DA8 in the Tables 1 to 3 of the supplement to the present publication."
  Page 32092, line 14/15 was changed to: "In the supplement to this publication, the most significant settings for the retrieval of HCl, $ClONO_2$, and HF at each site are described."
  Page 32091, line 23, the reference to the tables was changed to: "(see supplement to the present publication)"

- *p32091-32098: Why are the measurements of some of the instruments weather dependent and from others not?*
  They are all weather dependent, of course. But as this was nowhere mentioned properly, we added the following sentence on page 32090, line 15: "For these measurements, the atmosphere must be free of clouds and the sun needs to be above the horizon. This implies that during polar night, no measurements are possible. Of course, this dependency on direct sunlight leads to an irregular sampling of the measurements, with considerable gaps especially during the winter at the polar sites."

- *p32098, l22: "assist"? In what sense to assist? What is the purpose of applying model simulations in this study? Why does one needs models to investigate the*

ACPD
11, C16053–C16074, 2012

trend? This can also be done (and maybe even better) by applying measurements.
You are right, the expression was not appropriate in this context. The only way the models can assist the measurements is when determining the influence of sampling. The paragraph from page 32098, line 22, to page 32099, line 2, was changed to: "In addition to the FTIR measurements, results from five different atmospheric models are used in this study, comprising a two-dimensional (2-D) altitude-latitude model similar to the one used in Rinsland et al. (2003), called the Bremen 2-D model, two three-dimensional (3-D) chemistry transport models (CTMs), KASIMA and SLIMCAT, and two 3-D chemistry climate models (CCMs), EMAC and SOCOL (see Table 2). Thereby, the influence of the differing architecture of the models on the trend estimation can be investigated. On the other hand, the two CTMs KASIMA and SLIMCAT can help to estimate the influence of the irregular sampling of the measurements on the trend results (Sect. 5.4). This is possible because those two models use reanalyses calculated from actual measurements so that the state of the atmosphere simulated by the models can be assumed to be as close as possible to reality. In contrast, the 2-D model uses only one repeating annual cycle. The two CCMs calculate their own independent and consistent meteorology and dynamics which is not necessarily or rather probably not corresponding to the real meteorological situation."

- *p32098, general: Can 2-d and 3-d model fairly compared with each other? Why has these different kinds of models been chosen for this study? What are the advantages and disadvantages of using 2-d and 3-d models for the purpose of this study?*
  This question was hopefully answered already with the comment before.

- *p32099, l2: How? What is the difference in the performance of these models?*
  Please see the comment two before. This sentence has been removed.

ACPD
11, C16053–C16074, 2012

- *p32099, l5: Why have these time periods chosen?*
  These are the time periods that were available to us.

- *p32099, l9: What scenarios have been used? Why have these been chosen? What is the intention?*
  The scenario used for the greenhouse gases (GHG) is A1B from IPCC 2001 (see line 10). It was chosen because it is one realistic possibility. The models all used the same GHG scenario. The halocarbon scenarios are now described in more detail, please see especially the answer to the comment concerning page 32117, lines 14–19.

- *p32100, l26: What is exactly done when the data is nudged? What is the difference in the results if the simulation is nudged or not-nudged?*
  The KASIMA simulation is the only simulation that is really "nudged". This means that the model calculates its own meteorology, but every 4 hours (in the simulation used here), the calculated meteorological fields are forced towards the ECMWF fields (see also Reddmann et al., 2001). In contrast, the SLIMCAT model does not calculate any meteorological fields itself, but directly uses the ECMWF fields and calculates the chemistry and the transport of chemical species from them. So the meteorological situation simulated by KASIMA is not expected to agree as well as the one in SLIMCAT with the actual situation, but to be quite similar. (A difference in architecture between KASIMA and SLIMCAT is the vertical coordinate, which is represented by pressure in KASIMA and by potential temperature in SLIMCAT.) The 2-D model only repeats the same annual cycle over and over, so that also the total column abundances of the considered gases on a single day are not at all expected to exactly agree with the measurements. Similarly, this is not expected for the CCM results because they calculate their own meteorology which is expected to be consistent in itself, but they are not forced towards any atmospheric observations. But for all simulations and all models, the long-term trends are expected to agree if the boundary conditions e.g. for halocarbons and

ACPD
11, C16053–C16074, 2012

greenhouse gases are similar.

- *p32101, l16-19: How important is it for this study to have tropospheric convection in the model? How does the model assume that the long-lived tracers are well mixed? Is that a result or and assumption? How is it then done?*
  In the real atmosphere, mixing of trace gases in the troposphere is achieved by convection. If they are long-lived, they are expected to be well-mixed. But if this convection is not included in the model simulation, it must be prescribed (assumed) that the mixing ratio of these long-lived gases is the same throughout the troposphere.

- *p32101, l23: What is the abbreviation MECCA standing for?*
  It is "Module Efficiently Calculating the Chemistry of the Atmosphere". This was added in the EMAC description.

- *p32103, l3 and following paragraphs: The description of these figures is somewhat short.*
  These figures only present the original data sets and give a first overview. The focus of this paper is more on the results deduced from them, especially on the trends, which are discussed in more detail especially in the discussion section.

- *p32103, l3: How have the mean relative differences been calculated?*
  The first sentence in the caption of Table 6 was changed to: "Mean differences between models and FTIR measurements in % (calculated as (model-meas)/meas) averaged over all sites, and their standard deviations, for HCl, $ClONO_2$, and HF."

- *p32103, l222-25: Is that discussion not also part of this study? At least a short summary of the discussion should be given here. Why is it shown here if this has already be done?*

ACPD
11, C16053–C16074, 2012

We hoped the short summary given here is enough because it was already discussed in Kohlhepp et al. (2011) and the other publications mentioned there. The investigations in Kohlhepp et al. (2011) considered only one site, Kiruna, so the present study extends the KASIMA–FTIR comparison to more sites. Furthermore, it is also very interesting to see the KASIMA results directly compared with other model calculations, and also to see a possible latitudinal dependency.

- *p32104, l3-7: Why? Do you have any explanation for these differences? Why does the 2-d model not get it? Can this already be expected due to the missing dimension of the model?*
  Of course, it could be (at least in part) due to the missing dimension, but this cannot be confirmed without further investigations, which are beyond the scope of this study.

- *p23104, l6: The differences in the seasonal cycle concerning stations and latitude regions should be discussed more.*
  We added some information on this on page 32103, line 16: "In all three gases, a seasonal cycle is expected that is connected with the seasonal variation of the tropopause height. This variation results from the stratospheric general circulation transporting air from the summer to the winter hemisphere. The higher the tropopause, the smaller is the relative contribution of the stratosphere to the total column abundance. This again results in a lower total column abundance of HCl, $ClONO_2$, and HF in summer, because they are all mainly produced in the stratosphere. In the high latitude regions, HCl and $ClONO_2$ are in addition influenced by the absence of solar irradiation in winter. Chlorine activation on the surface of polar stratospheric clouds (PSCs) leads to a decrease in the total column abundances of the chlorine reservoir species and a strong peak at the end of the winter due to the deactivation of active chlorine. So the seasonal cycle is expected to exhibit its largest amplitude at the polar sites."

ACPD
11, C16053–C16074, 2012

- *p23104, l20: Why is the performance of the 2-d and the two CTMs better than the performance of the two CCMs?*
  This is not the case. The mean differences for $ClONO_2$ are smallest for the 2-D model, the CTM KASIMA and the CCM SOCOL. However, this comparison is difficult for the CCMs anyway (for all gases) because the CCM simulations are not nudged, that means their meteorology does not correspond to the real state of the atmosphere on a certain day. They are expected to agree with the measurements only on climatological time scales, so also the comparison on monthly time scales shown in Table 6 can only give qualitative information about the overall agreement.

- *p32104, l22: Reference to Fig. 7 is missing here.* Was added.

- *p32105, l20-28: This too descriptive. Some discussions what the reasons for these differences could be are missing.*
  We tried to improve the discussion on reasons for the differences in the discussion section (now 8), please see the comment concerning page 32117, lines 14–19.

- *p32106, l19-20: Why is it ok to just use this approach?*
  By using a first order Fourier series in the fitting function, the seasonal cycle is represented by one maximum and one minimum whose positions are not independent from each other. A third order Fourier series is able to represent the seasonal cycle in more detail, e.g., a broader maximum and smaller minimum which are not exactly half a year apart, or something similar. So it can be expected that including the third instead of only a first order Fourier series gives a lower root mean square difference between the data and the fit. This finally results in a smaller uncertainty for the trend. However, at the northernmost polar sites, the regular gaps occurring due to polar night are quite long so that using the third order Fourier series in the fit may result in large unphysical oscillations.

ACPD
11, C16053–C16074, 2012

The trend result is not trusted then either. By using only a first order Fourier series and thereby accepting larger root mean square differences, these oscillations can be avoided.

- *p32106: It still has not been explained what the "bootstrap" method is.*
  The paragraph from line 4 to 11 was rewritten as follows: "A linear trend function, combined with a Fourier series accounting for the seasonal cycle, is fitted to the time series using a least squares method. The precision of the fit parameters and thus of the trend is determined with the bootstrap resampling method. This method is only described shortly here, however, more detailed information can be found for example in Gardiner et al. (2008), and references therein. It has been used in previous trend studies from FTIR measurements (e.g., Gardiner et al., 2008, Mikuteit, 2008, Vigouroux et al., 2008, Kohlhepp et al., 2011). From the main fit to the data, the differences between fit and data are calculated. These residuals are then added randomly (with replacement) to the fit result in order to create a new, artificial data set. Another fit using the same function as for the real data is performed on the artificial data set, giving an artificial value for every parameter, including the trend. This procedure is applied 5000 times in this study, following the example of Gardiner et al. (2008). From the 97.5 and 2.5 percentiles of the resulting 5001 trend values, the 95 % confidence interval characterising the trend uncertainty can be estimated. The reason for choosing this method to determine the trend uncertainty is that it does not assume that the residuals of the fit to the data are normally distributed. Instead, it only assumes that there are enough data points for the residuals to sufficiently represent their own distribution. The assumption of a normal (Gaussian) distribution of the residuals might not be valid because the fit is not always able to capture the complete annual cycle, e.g. the strong peak in $ClONO_2$ at the polar sites in spring. The main reason for this is that the peak does not always occur at exactly the same time of year."

- *p32107: put section 4.1 into section 4 and then continue with 4.1.1 as 4.1 and so*

ACPD
11, C16053–C16074, 2012

on.
Please see the answer to the first of the technical comments.

- *p32108: The measurement instruments and models are described in detail while the method used for the trend calculation is not described at all. What are the experiences of other scientists using this method for trend calculations? References to other studies are missing.*
  Hopefully this comment has been answered already with our answer to your comment concerning page 32106.
  The following sentence was added on page 32106, line 13: "As also found by Gardiner et al. (2008), this approach represents most time series very well and at the same time avoids over-fitting the data. But at the sites poleward of..."

- *p32108, l11-12 and l15-16: Why theses paragraphs consisting of a single sentence?*
  We put the discussion for all three gases together now into one paragraph.

- *p32109, l3: Why have these time periods been chosen?*
  In line 4, the following sentences were added: "The longest period was selected because not all, but more than half of the stations performed measurements during this time, while only a few did before. The longer the time series, the smaller the error bars and the better the trend estimate is expected to be. During the second period, all sites except La Réunion and Toronto performed measurements (see below). From earlier studies, e.g. the one by Rinsland et al. (2003), we expect the inorganic chlorine species total column abundances to have reached their plateau around 1996–1999 and not to strongly change during this time. So the decrease expected due to the Montreal Protocol may be weaker if these four additional years are included in the trend calculation. The latest and shortest period was included in this study in order to investigate whether there was a change in the rate of increase (of HF) or decrease (of HCl and $ClONO_2$) during the last

ACPD
11, C16053–C16074, 2012

few years."
To complete this discussion, the following sentence was added in line 25 of the same page: "The expected tendency of a weaker decrease in the chlorine species during 1996–2009 that was discussed above can indeed be found in the presented results, especially for HCl. It is probably due to the stratospheric chlorine loading reaching a plateau at the end of the 1990s and only then slowly starting to decrease."

- *p32110, l6: Seems that this one reason why the model simulations have been taken into account. This should have been mentioned already much earlier.*
  This was done now, please see the answer to the comment concerning page 32098, line 22.

- *p32110, l14: Why has this time period been chosen?*
  This issue was addressed in a general way in the new text describing the content of the new Sect. 5. Please see the answer to the first technical comment.

- *p32110: Up to now it has not really been explained in how far these instruments are dependent on these factors.*
  It is mentioned now, please see the answer to the comment concerning pages 32091–32098.

- *p32112, l5-6: The temporal model output is quite different. How does that affect the results? How does the application of a 2-d model instead of a 3-d model affect the results?*
  The differing temporal model output should not influence the trend results because it is a regular output in all cases, in contrast to the irregularly sampled measurements. But the less data points, the larger the error bars of the calculated trends, of course. Concerning the effect of the model architecture on the trend results, this was supposed to be investigated in this study. It is being stated that the 2-D model does not show a large latitudinal variation, but the overall

ACPD
11, C16053–C16074, 2012

agreement of the trends with the results from the other models and the measurements is good, especially for HCl and $ClONO_2$.

- *p32112, l19: "do not show" instead of "not showing".*
  We must admit that the sentence structure is quite complicated. "not showing" refers to the "due to", like "indicating".

- *p32112-32113: What are the reason for the differences found between model and measurements? How do the trend derived in this study compare to other studies? Especially, how do they compare to the Rinsland et al. study?*
  We tried to improve both points you made. Especially the discussion section now contains more discussion on that. Please see the comment concerning page 32117, lines 14–19.

- *p32114, l3: It has still nowhere been mentioned why this time period has been chosen.*
  Now it has, please see the answer to the comment concerning page 32110, line 14.

- *p32114, l24: Finally an explanation. This really should have been mentioned much earlier!*
  Part of this discussion is indeed mentioned earlier, on page 32108, line 1 and lines 20–25.

- *p32115, l15-18: This in important fact which should have been mentioned already much earlier!*
  This has been changed now, please see the answer to the comment concerning pages 32091–32098.

- *p32117, l8-10: It should have also been written like this already in the introduction.*

ACPD
11, C16053–C16074, 2012

It was added to the introduction on page 32089, line 9: “Because fluorine is also contained in CFC substitutes, the HF total column abundance is expected to have continued increasing during the time range considered in the present study.”
This information was also added in the abstract, please see the comment concerning page 32087, second paragraph.

- *p32117, l14-19: What is the reason for this differences? This is despite the discussion not becoming clear.*
Overall, we tried to improve the discussion on reasons for the differences between the trends of models and measurements. Therefore, we especially added more detailed information on the scenarios used by the models and how differences between them might affect the trends. Specifically, we made the following changes:
The paragraph on page 32088, lines 17 to 23 was removed and replaced by: “Figure 1 shows the time development of the mean global surface volume mixing ratios of total organic chlorine (CCl_y) and total organic fluorine (CF_y) according to the halocarbon scenarios that were used as boundary conditions for the model simulations, between 1992 and 2010. Additionally, their relative annual growth rates are shown. CCl_y is defined here as 3 CFC-11 + 2 CFC-12 + 3 CFC-113 + 2 CFC-114 + CFC-115 + 4 CCl_4 + 3 CH_3CCl_3 + HCFC-22 + 2 HCFC-141b + HCFC-142b + Halon-1211 + CH_3Cl and CF_y is represented by CFC-11 + 2 CFC12 + 3 CFC-113 + 4 CFC-114 + 5 CFC-115 + 2 HCFC-22 + HCFC-141b + 2 HCFC-142b + 2 Halon-1211 + 3 Halon-1301 + 2 Halon-1202 + 4 Halon-2402. According to the A1 scenario from WMO (2007), CCl_y is assumed to have reached its tropospheric maximum in 1993, whereas CF_y is expected to have reached a plateau with a small positive growth rate of about 0.1 % per year in 2010 (Fig. 1). The older scenario Ab from WMO (2003) assumes CCl_y to have reached its maximum only in 1995. In contrast, CF_y peaks earlier than in the A1 scenario, already in 2005, so that the growth rate in the Ab scenario in 2010 is

ACPD
11, C16053–C16074, 2012

already negative. A small part of the difference between the two scenarios probably results from the fact that in the WMO (2003) Ab scenario, Halon-2402 and Halon-1202 are not considered."

Of course, the corresponding figure (Fig. 1) was also changed accordingly and now shows the different scenarios used by the models, not only from WMO (2007) as before. The figure caption now reads: "Time series of monthly mean CCl_y (left) and CF_y (right) surface volume mixing ratios (in pptv) and growth rates (in % per year) from different halocarbon scenarios between 1992 and 2010. The one called "WMO (2003) REF2" corresponds to the scenario Ab in WMO (2003) and was used by KASIMA and the 2-D model. The "WMO (2003) REF1" time series of CCl_y and CF_y are based on that same scenario until the year 2000, but were corrected by additional measurements between 2000 and 2004. It was used by SOCOL. The "WMO (2007)" scenario is the one called A1 in WMO (2007) and was used by SLIMCAT and EMAC in this study."

Concerning the halocarbon scenarios, the information was corrected in Table 5.

On page 32099, the paragraph from line 6 to 23 was rewritten to: "For all five models, the time evolution of the greenhouse gases and ozone-depleting substances was prescribed as a boundary condition at the lower model boundary. The emission scenario for the most important anthropogenic greenhouse gases, i.e. CO_2, CH_4, and N_2O, was the IPCC scenario A1B for all simulations considered here (see also Table 5). It assumes very rapid economic growth, low population growth, and the rapid introduction of new and more efficient technologies (Nakicenovic et al., 2000). The time evolution of the global surface volume mixing ratios of the ozone-depleting substances (ODS) was prescribed according to different so-called baseline scenarios. This means the scenarios represented the best guess for both past and future source gas emissions at the time of their publication. A comparison between them is shown in Fig. 1. KASIMA and the 2-D model applied the Ab scenario from WMO (2003), called REF2 in Fig. 1, while SOCOL used the REF1 modification where updates from newer observations for

ACPD
11, C16053–C16074, 2012

some source gases were made between 2000 and 2004. The ODS in SLIMCAT and EMAC follow the scenario A1 of WMO (2007). All the ODS data were provided in the framework of the SPARC (Stratospheric Processes And their Role in Climate Change) CCMVal (Chemistry-Climate Model Validation activity) initiative (Eyring et al., 2006, 2007) and were recommended for use as lower boundary conditions in the simulations for the 2006 and 2010 WMO Ozone Assessments."
In the discussion, the following paragraph was added on page 32116, line 4: "Part of the discrepancy between the modelled trends can be explained by the different halocarbon scenarios used in the simulations. Considering a time shift of a few years due to the transport of CCl_y and CF_y from the surface to the stratosphere, the weakest chlorine decrease in 2000–2009 would be expected from the WMO (2007) Ref 2 scenario used by KASIMA and the 2-D model (Fig. 1). KASIMA indeed at most sites shows the weakest decrease in HCl and $ClONO_2$, along with SLIMCAT (Fig. 12 and Tables 7 and 8). In contrast, the 2-D model shows a stronger decrease than expected from the scenario. Above most of the measurement sites, also EMAC and SOCOL show stronger HCl and $ClONO_2$ decreases than expected from the surface halocarbon scenarios they used. For HF, the 2-D model calculated much weaker increases than KASIMA and SLIMCAT did (Fig. 12 and Tab. 9). This cannot be explained by the different scenarios because as already mentioned, KASIMA and the 2-D model used the same one. A possible reason for a part of this discrepancy in the HF trends is the fact that the 2-D model does not treat all halogen-containing species explicitly (please see Sect. 3.1). Instead, the mixing ratios of some are added proportionately to those of others with similar lifetimes by considering the number of chlorine atoms. But this means that for some gases, the contained amount of fluorine is not represented correctly so that very roughly about 50 pptv are missing in the surface CF_y mixing ratio. As the missing amount of CF_y increases with time, the trend of the HF total column abundance is expected to be slightly too small in the 2-D model."

ACPD
11, C16053–C16074, 2012

In context with the missing CF_y in the 2-D model, the following sentence was added to the description of the 2-D model, page 32100, line 9: "In this simulation, the following halogen-containing gases are treated explicitly: CFC-11, CFC-12, CFC-113, CCl_4, CH_3CCl_3, Halon-1301, Halon-1211, HCFC-22, and CH_3Cl. In contrast, HCFC-141b is not treated explicitly, but proportionately added to CH_3Cl_3 so that the additional chlorine atoms are accounted for. Analogously, the CFC-114, CFC-115, and HCFC-142b surface mixing ratios are considered in the HCFC-22 value."

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