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Atmospheric Chemistry and Physics
Discussions

Interactive comment on "Global HCFC-22 measurements with MIPAS: retrieval, validation, climatologies and trends" *by* M. Chirkov et al.

Anonymous Referee #1

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General Comments

This manuscript describes the retrieval of MIPAS HCFC-22 profiles from 2005-2012, validates them, and presents some figures showing its morphology. It concludes with a discussion of stratospheric trends from this short data set. It is long and poorly written, especially the retrieval section. The validation section underused (maybe misused) the ACE data, which are a valuable tool for the validation. Two balloon profile data sets are also used but are not appropriate for validating the MIPAS data. There is little analysis of the MIPAS data. Figures are presented and described but not analyzed in any quantitative way. There is no HCFC-22 climatology in the Climatology section. No meteorological data are used to support descriptions of HCFC22 behavior or statements about processes affecting the distributions. Many statements about HCFC22 strato-

spheric structures are known from previous studies (e.g., using ACE satellite data).

The topic and the MIPAS data set used do have a place in the literature, so there is good 'raw material' here for a publication. However, this manuscript requires major revision. Some sections can be eliminated, others combined, and almost all need reorganization or rewriting. The comments below are organized by section; they note what I think the major problems are and how each section might be improved. Because of the significant revisions needed, it is premature to make many minor or technical suggestions.

The standard of English used is fair. I appreciate how challenging it is to write a scientific article in a second language! I strongly recommend having a native English speaker proofread the manuscript to improve readability. As an example 'The profiles obtained are…' sounds better than 'Obtained profiles are…' A satellite measurement is a sounding but measurements aren't 'sounded'. Also, the writing style uses many unnecessary words. E.g., '…has a more or less seasonal cycle'…more or less adds nothing, try simply 'has a seasonal cycle'. 'So-called level 1B product': why 'so-called'?

Introduction

p. 14786, Line 10. The most current determination of the atmospheric lifetime of HCFC22 can be found in the 2013 SPARC report (a WMO publication). The SPARC lifetime is the same as reported here but it's a more current assessment of the lifetime.

Section 2, MIPAS data

No mention is made of the MIPAS sampling pattern. What is its latitude range? Is the range covered daily? Does is sample more at some latitudes than others? Does it measure all latitudes in all seasons?

Section 3, Retrievals

This section is not well organized and it reads like a series of unconnected details regarding the retrievals. For example, the last paragraph of the section is on information displacement: how is this relevant to understanding the data? Stiller et al, ACP 2008, on SF6, provides a good example of a retrieval description. I suggest you consider this and other MIPAS publications to see how they explain their retrievals (e.g.,) – then rewrite accordingly.

p. 14789, Line 20. What is a 'zero-level profile'? How sensitive are the results to the a priori? p. 14790, lines 3-15. The writing does not flow well. Where do the temperature analyses come from? (state it and reference it) line 23. 'Its dominating components…'? maybe 'primary components'. 5 things can't all dominate. p. 14791, line 2. What is regularization? It hasn't been defined.

Section 4, Validation

p. 14792, line 11. What do you mean by 'unsolved problems cause by different altitude resolutions'…? All the validation comparisons you make involve measurements with altitude resolutions different than MIPAS, so this statement is puzzling. Lines 20-22. How does MIPAS sampling (latitude and season) compare to ACE?

Section 4.1, ACE comparison (figures 4-7).

By examining a limited latitude region and sorting by season, Figure 5 provides the most useful comparison of this section. The two data sets agree within their uncertainties below 20 km all the time while seasonal differences are revealed above 20 km. Why, then, do you combine all latitudes and seasons in the other figures? So much information is lost. In fact, Figure 6 gives the opposite impression as Figure 5: it shows continuous, nearly 1:1 agreement below $\sim$25 km or so. (Thus I cannot understand the statement that the points fall into 2 clusters, p. 14794, line 2). In general I find that the words written in this section do not align with what is shown in the figures. For example on p. 14793, line 11, 'The bias is significant at all altitude levels.' It clearly isn't! In Figure 7, there is a bimodal shape in the lower right histogram, but as global data are combined in this figure, who knows why this shape occurs? (But a different comparison would probably reveal the answer.) Overall, the ACE comparison is inadequate. It

could be a more valuable and useful part of this paper with comparisons that examine specific seasons and latitude ranges.

Section 4.2, Cryosamplers

p. 14795, lines 3-6. These sentences are poorly worded. They serve as the introduction to the topic of the next paragraph and should be combined with it. The coincidence criteria is so broad (1000 km and 24 h) that when the profiles do not agree you really can't know why. If you use some meteorological analyses to show the profiles are from similar environment, then you would know whether it made sense to compare them. As it is, Figure 8 shows mostly a lot of disagreement with the balloon profiles but the reason is probably geophysical variation – thus there is no point to these comparisons! And why calculate a 2005-2011 mean profile at all? This gas is increasing rapidly – at least 25% over this time period. The multi-year mean is meaningless, and should a balloon profile match it, that is meaningless too. Unless you can demonstrate that in spite of the broad coincidence criteria that it makes geophysical sense to compare with MIPAS, these comparisons could be eliminated.

Section 4.3, MkIV comparisons

p. 14796, lines 15-28. Too much detail on the MkIV instrument – use the Toon reference and eliminate most of this. p. 14797. Since there are no MIPAS data for the dates of the balloon flights, these flights are not useful for validation! Again, comparing to a multi-year mean is not satisfying or meaningful in a quantitative way. This section can be eliminated.

Section 4.4 Summary of intercomparisons

The summary states that, compared to the different data sets, the MIPAS bias is either low, high, or zero. This is not a result. It is exactly what was known before any comparisons were made. The balloon profiles really aren't adequate for this validation – that's fine, so don't use them. The validation section would be greatly improved by expanding the ACE data comparisons as noted above. After a more thorough validation using the ACE data, I think you will be able to state much more definitively where the data sets agree and where there is bias (and how much).

Section 5. Climatology

The data are presented in various ways (e.g., latitude v. time, altitude v. latitude, etc) but there is actually no climatology here. In general, the 'analyses' in the subsections are only descriptive (i.e., descriptions of what is already known) or speculative, and do not present any quantitative analyses. No meteorological data are used in support of speculative statements about processes that might be indicated in the data. This section would be improved by including a climatology (i.e., mean distributions as a function of month/season, mean cycles, etc.) and by adding meteorological analyses to give support to the processes you describe.

Figures 10 and 11 are introduced (p. 14798, line 3) but the next figure mentioned is Figure 15 (line 19). Figures must be mentioned in sequence. p. 14799, lines 7-9. While rapid uplift can explain why upper tropospheric values are nearly equal to surface values, they cannot explain mixing ratios that are higher than the surface. What would the source of the extra HCFC22 be?

Sections 5.1 and 5.2 have lots of qualitative discussion but there is no actual MIPAS data analysis that demonstrates any of the processes discussed; e.g., p. 14798, line 22: "The following scenario is suggested…". Analyses, not suggestions, are required for publication. Why not bring in meteorological data to support your ideas? Also, Figure 15, which is used to show something about monsoon transport, crams 6 years of data on 5 surfaces into a very tiny space. The panels are illegible, the font impossible to read. It is impossible to see the details of a seasonal process such as monsoon transport with a tiny panel showing a 6-year time series. Other parts (e.g., first few paragraphs of 5.2) describe what is already very well understood about long-lived trace gas structures and seasonal cycles.

Most air enters the stratosphere through the tropical tropopause, so interhemispheric (IH) differences in long-lived trace gases found in the troposphere are usually not found in the stratosphere. (CO_2 has some but they disappear quickly with height.) If you speculate about IH differences in the stratosphere, you'll need a supporting analysis to demonstrate that tropospheric IH source differences are the cause.

p. 14800, line 14. 'Interestingly, the breakup of the vortex seems to take place at all altitudes at almost the same time for the northern polar region.' The Arctic vortex final warming ('breakup') occurs in March or April and shows considerable interannual variability in how it breaks up (e.g., wave 1 or wave 2 warming). Its variability is much greater than the Antarctic vortex, so your statement doesn't make sense. The sharp discontinuities found each year in your figure appear too early in winter to be the breakup – they are probably midwinter sudden warmings, not the final warming (breakup) that occurs in March or April. Please check your figures and interpretation.

The lowermost stratosphere is generally below 16, not 20 km. It is below 380 K (below Hoskins' 'overworld'). p. 14801, line 15. 'the seasonal cycle in the SH…is not globally compensated but only weakened by the cycle of the NH': What does globally compensated mean? The results of the growth rate analysis in Section 5.3 (p. 14803) might be better displayed as a table. Too many different units are used to discuss growth rate, making it harder to compare results with previous studies cited. The Figures (16,17) use ppt/yr and%/decade. Studies cited on page 14803 use %/yr. Pick a unit and stick with it.

Section 5.4 Comparisons with surface measurements

The NOAA/GMD and AGAGE measurements are both precise and accurate: why compare in two separate sections? Please combine them into a single 'comparisons with surface' section. p. 14804. 'the troposphere can be considered well-mixed'. Yes, sort of. As you can see in your own figures there are interhemispheric gradients of ∼20 ppt (∼10%) at the surface.

Why is the first parargraph of Section 5.4 talking about the dryness of stratospheric measurements when this section is about comparisons (of dry mole fraction) ground-based measurements with MIPAS tropospheric measurements? You haven't explained whether the MIPAS tropospheric measurements are wet and whether this will impact the comparisons with ground-based data. If there is an impact, what size is it? Big enough to cause a bias, or is it in the noise compared to other differences?

p. 14805. If MIPAS measurements and ground measurements are the same to within their overlapping uncertainties, then this means they AGREE. Say so. Figures 18 and 19 have too much data. It is hard to distinguish MIPAS behavior at different latitudes. I suggest organizing the comparisons into panels showing different latitude ranges (e.g., NH extratropics, tropics, and SH extratropics) and include the relevant GMD and AGAGE stations on each panel.

p. 14805, lines 21-24. Again, this is more speculative discussion that is not supported by meteorological analyses or other trace gas measurements. This section could be very interesting if you examined the HCFC22 data as a function of altitude in the lowermost stratosphere (e.g., 12-16 km) in each hemisphere to identify seasonal transport processes in the lowermost stratosphere. Interhemispheric differences between the results may reveal process important to the cycles in each hemisphere. p. 14806. I see that you recognize that chemistry, emissions, and transport are all important to understanding HCFC22 behavior. But this means that it can't be understood as simply as discussed here. An atmospheric model is required to adequately interpret the behavior.

Section 5.5. Stratospheric trends

This paper examines a 7-year data set. Seven years is less than 3 QBO cycles. Each QBO cycle is different in length and its seasonal timing. It is not completely accounted for in the regression analysis by considering terms at 2 pressure levels. Because the regression analysis cannot adequately remove the QBO effect, the residuals (Figs. 17

and 20) have a QBO signature. To search for a trend (that indicates stratospheric circulation change) in a 7-year data set is by definition unreasonable. There are end effects (2 years out of the 7 are endpoints!), the QBO cannot be adequately accounted for, and in the case of HCFC22, there is an enormous annual growth (compared to a possible circulation change) in the species measured. Section 5.5 should be eliminated.

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