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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Satellite observations of stratospheric hydrogen fluoride and comparisons with SLIMCAT calculations" *by* J. J. Harrison et al.

Anonymous Referee #2

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Review of the manuscript by J.J. Harrison et al. (ACPD, 34361-34405, 2015) "Satellite observations of stratospheric hydrogen fluoride and comparisons with SLIMCAT calculations".

In this manuscript, the authors combine the two multi-year satellite infrared solar occultation data sets available for hydrogen fluoride (HF), the main stratospheric reservoir of fluorine, in order to determine its global distribution and trend over the 1991-2012 time frame. The version 19 set (the latest release to my knowledge) derived from the HALOE (HALogen Occultation Experiment) observations and covering the 1991-2005 period is used, complemented with several subsets (v2.2, v3 and v3.5) derived from the ACE (Atmospheric Chemistry Experiment)-FTS instrument, in operation since 2004. Furthermore, ACE-FTS data available for the main F-bearing source gases (CFC-12,

CFC-11 and CFC- 113) and two intermediates of their degradation (COF2 and COCIF) are presented. Model results by the TOMCAT/SLIMCAT 3D Chemical Transport Model are included for comparison with the observations and to support the interpretation of the results.

The manuscript is generally clear and well written (although some figures (as Fig. 6) remain tiny and of limited use), the data sets and the results are important and the subject is clearly of relevance for this journal. In my opinion, there are however a few drawbacks that need to be fixed before publication. They are identified and listed below, together with suggestions for improvement.

While one of the aim of this paper is to characterize and understand the evolution of HF over two decades or so, with two different instruments (already a challenge with only a few months overlap between the two missions...), three -possibly inconsistent!- versions of the ACE-FTS data are used, version 2.2, version 3 and 3.5. Moreover, these versions are incompletely described, with e.g., Table 1, 2 and 3 providing information as to the settings for v3 and v3.5, but nothing for v2.2. There is no effort to characterize a possible systematic bias (because different HF lines might be used, the interferences accounted for might be dissimilar...) and to merge the ACE data sets. The same is true for the combination of ACE-FTS with HALOE results, despite a well-known bias. The authors state (section 3.2, page 34371): "There have been no detailed comparisons in the literature between ACE-FTS v2.2 and v3.0 HF datasets, however Duchatelet et al. (2010) state that first comparison exercises involving ACE-FTS v3.0 products indicate a decrease of close to 5 % in HF amounts". If the bias is not well known while perhaps non-negligible (5%), the authors have to characterize it, this is certainly not beyond the scope of this study, given its aims. They have at hand all what is needed and my recommendation is to use significant subsets of occultations available for v2.2, v3 and v3.5 to determine their consistency and correct for a possible systematic bias. The next step will require a careful combination with the HALOE set, following e.g. the method developed for the generation of the GOZCARDS data product.

In the present version of the manuscript, the GOZCARDS ensemble appears useless or underutilized. Added "for completeness" (section 2.3, page 34370), it is only included in Figure 7 and little is learnt from these comparisons. Indicatively, neither the abstract nor the conclusions mention findings resulting from its use. The statement on page 34380-34381 "Had v3.0/v3.5 ACE data been used instead, the GOZCARDS dataset would have been shifted lower in VMR by several percent" further adds to the confusion, leaving the reader unsure about the consistency of the data sets used for the trend evaluation. Trend evaluations which btw do not consider the GOZCARDS merged data set, while it is covering the 1991-1997, 1998-2005 and nearly the 2004-2012 (2004-2010) time intervals. Therefore, my recommendation would be either to discard the GOZCARDS set (saving one figure), or to keep and exploit a merged set for the trend investigations, i.e. supposedly an asset with this respect.

One of the conclusions of this study is that changes or variability in stratospheric dynamics are responsible of variations in the HF trends with altitude and latitude. Several recent papers have identified and investigated these changes (e.g. Ploeger et al., 2015, doi:10.1002/2014JD022468, a reference to it might be useful to the reader), or their impact on significant stratospheric composition changes with time (e.g. for ozone, hydrogen chloride...). As a possible result, the evolution of HF in the stratosphere might well not always follow a smooth route, as is the case in the troposphere, complicating the interpretation of its trend in the stratosphere (upper or lower, in SH or NH), to e.g. support the Montreal Protocol. Indeed, how are the circulation changes and the reduction/variation in source gases emissions contributing to the derived trends? I believe it is therefore important to provide elements allowing to fully characterize these contributions. The support of SLIMCAT is key here, and the figure 8 (and similar) provide an important input, showing the net and contrasted effect of stratospheric dynamics on the HF trend over the 2004-2012 time period. But there is no information as to the temporal development of HF with altitude/latitude. I think that adding the "fixed to 2000 dynamics" SLIMCAT time series to Figure 6 would be very useful to identify in the various subsets the most significant departures from a smooth unperturbed HF evolution as

driven only by surface emissions of the source gases and their subsequent conversion to inorganic fluorine.

Minor comments/corrections

Abstract

P34363-L3: suggest adding "involving" to get "...nature, involving e.g. chlorofluorocarbons (CFCs),..."

Introduction

P34364-L7: "source molecules are CFC-12, CFC-11, CFC-113" instead of "source molecules are CFC-11, CFC-12, CFC-113"

P34364-L16: suggest changing to "Certainly, in addition of HCl, monitoring the growth..."

P34365-L5: suggest adding a blank line between R2 and R3

P34366-L15: suggest changing to "..., based on solar spectra recorded by balloon-borne and from the ground at Jungfraujoch,"

Section 2.1

P34368-L22: change to "an atmospheric density of 9E15 or 2E16 molecules cm-3"

Section 2.2

P34369-L19: Change to "For the HF channel, the spectral bandpass..."

Section 3.1

P34371: I am questioning the relevance and usefulness of the paragraph between lines 8 and 14, starting at "Recently"

Section 4

P34374-L12: Could the switch from ECMWF to ERA-I reanalyses be responsible of a bias/change in quality in the SLIMCAT simulations? With a significant impact on the respective HF trends?

Section 6

P34383-L7: a reference such as Ploeger et al. JGR, 2015 might be relevant/useful here

Table 1. Wouldn't it be more useful to quote the upper approximate altitudes in the last column, for all cases, and mention the density unit threshold in the foot note for the relevant cases?

Figure 7: the GOZCARDS symbol should be "empty" (instead of a black diamond)

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 34361, 2015.