

**Response to reviewers' comments on the manuscript "New and improved infrared absorption cross sections for dichlorodifluoromethane (CFC-12)" by Jeremy J. Harrison**

I wish to thank the reviewers for their helpful comments. I respond to their comments, which are in bold text:

Referee #1:

**General comments**

**Introduction:**

**They are, in fact, two (not explicitly differentiated), sub-sections, i.e.:**

- a) First sub-section on the CFC-12 history and uses facts, which could be shortened;**
- b) Second sub-section related with atmospheric concentration measurements, using remote sensing experiments and required retrieval algorithms. The differentiation of these two topics should appear explicitly. Related with b), more details should be given on the direct (forward) and inverse modeling processes and their related requirements; this could be illustrated by the ACE-FTS experiment (related with section 4 concluding comments).**

The reviewer is correct that the introduction consists of two parts, one on the history and uses of CFC-12 (my view is that a discussion on the history of CFC-12 is interesting) and the other on the remote sensing of CFC-12. The introduction is in this form to provide a background on the importance of this molecule and the importance of its quantitative IR spectrum in satellite retrievals. I am sure the majority of readers already have an understanding of forward and inverse modelling, so an in depth discussion on this is probably not required. The primary purpose of this paper is to provide details of a new spectroscopic dataset for use in retrieving CFC-12 from EO instruments. The manuscript outlines a number of general spectroscopic requirements for absorption cross sections; it is these data which feed directly into the forward model. Errors in the spectroscopy, i.e. in the widely used cross-section dataset of Varanasi, propagate into retrieval errors. This point has now been explicitly added to the text. I do not wish to split the introduction into two distinct sections, but I suspect the reviewer's issue is that I haven't provided enough 'bridging' text to differentiate the sections more clearly. I have now made some additions and changes to the text to remedy this.

**Section 4:**

**As for the introduction, two not differentiated sub-sections appear, i.e.:**

- a) First sub-section devoted to quality comparisons with Varanasi's former measurements, involving experimental methods and subsequent results. In the assessments lines 8-13, page 2833, try to provide numerical values, to better illustrate the enhanced quality of the presented new work.**

Lines 8-13 of page 2833 contain a discussion of channel fringes in the Varanasi cross sections. I have added some additional text as requested.

- b) Second sub-section on the ACE-FTS experiment CFC-12 mixing ratios data retrievals, mentioning spectroscopy quality requirements to lower the residuals. This section should be better associated with b) of the introduction. I would suggest that the related topics on: forward, inverse modeling, and CFC-12 atmospheric content**

**retrievals, should be provided a clearer and more precise emphasis, especially illustrated by the ACE experiment expected performances. The subsequent progresses in CFC-12 atmospheric quantity retrievals, thanks to the new data, could be numerically illustrated (“substantial improvement in the  $1\sigma$  retrieval errors”).**

The reviewer’s point isn’t quite right. The discussion of ACE-FTS spectra was not intended as a discussion on improvements of the ACE retrieval, or atmospheric retrievals in general, but as an extension of the comparison between the two cross-section datasets. A high-resolution atmospheric spectrum with strong CFC-12 absorption features, as measured by the ACE-FTS, is ideal for illustrating the better quality of the new measurements compared with the previous Varanasi dataset. There are two main points:

1.) The derivative-shaped features observed in ACE-FTS v3.0 residuals are a direct consequence of the poor wavenumber calibration in the Varanasi dataset. The fact that these features are minimised when using the new dataset illustrates the improved accuracy of the wavenumber calibration.

2.) The minimisation of the residuals associated with the strong features of the  $\nu_8$  band Q branch illustrates a major defect in the Varanasi dataset, a combination of poor wavenumber calibration and saturation of the Q branch, which the new dataset resolves.

I have modified the text to make these points more clear.

I believe the main point of the reviewer here is that the subsequent improvement in CFC-12 retrievals should be numerically illustrated. This is a good point, however one that is beyond the scope of this work. As mentioned before, the point of this paper is to describe a new and improved absorption cross-section dataset and make it available to the wider remote-sensing community. The ACE retrieval scientist informs me that the new cross sections have improved the  $1\sigma$  errors, although retrievals have only been performed for a handful of occultations. A proper investigation into this requires processing of a large amount of data. Currently a new processing version (v4) of the ACE data is under preparation, and it is not anticipated that the actual processing will begin until early 2016. With various hardware / resource issues at ACE SOC, re-processing v3.0 data with the new cross sections is not a priority. Once v4.0 ACE-FTS processing is complete (hopefully in 2016), I have plans for a detailed study on the improvement of CFC-12 retrievals. I have now explicitly stated this in the text.

#### **General comment on the paper:**

**My opinion is that these new results on CFC-12 cross-sections, thanks to improved measurements and producing a much more balanced dataset with wider PT combination coverage, than the previous ones of Varanasi et al., represent a substantial contribution for an improved knowledge of CFC-12 concentration in the atmosphere. Anyway, it has to be mentioned that continuous recommended experimental improved measurements will be necessary, for their use in future planned hyper-spectral remote sensing experiments. This could be shortly documented and illustrated in the very final conclusions of the paper.**

I am glad the reviewer agrees that the new dataset is superior to that of Varanasi. CFC-12 absorption is stronger in the limb (compared to nadir), so most CFC-12 measurements are from limb sounders. I do not believe that there are any planned after ACE-FTS (the so-called ‘limb gap’). Although a nadir sounder like IASI can ‘see’ a weak CFC-12 signature, there is very little effort in actually retrieving total columns of CFC-12. Therefore, I am unaware of any planned missions that are actually focussed on measuring CFC-12.

#### **Technical corrections:**

**Page 2826 line 6: ODSs (Ozone-Depleting Substances).**

This has been corrected.

**Page 2829 line 20: Give the link: <https://www.bruker.com/fr/products/infrared-nearinfrared-and-ramanspectroscopy/opus-software/overview.html>, providing OPUS software information; line 24 MCT (Mercury Cadmium Telluride)**

The link should be <https://www.bruker.com/products/infrared-near-infrared-and-raman-spectroscopy/opus-software/overview.html>. This has now been added to the text, and MCT defined.

**Page 2831 line 13: Give intensity unit.**

The intensity units of the absorption cross sections are given in line 8. If the reviewer means the units of integrated band strength, these are already given in Eq. 2.

Referee #2:

**The new set of CFC-12 cross sections appears to represent a significant improvement over the existing set and will be a valuable addition to the databases.**

**The article seems clear enough and is generally well organized. The depth of the discussion on the history of refrigeration in the Introduction felt somewhat excessive.**

**There is much discussion on how the new cross sections improve the residuals in the analysis of atmospheric spectra, specifically for preliminary studies performed with the ACE-FTS, but this aspect ends up being vague because it is unclear from the text what the extent of the improvement actually is. Ideally, the paper would include figures showing examples of these preliminary studies, to best convey the magnitude of the problem with the original set of cross sections and the degree to which the new set of cross sections fixes the problem. Stating that the systematic features “go away” has less impact than a figure showing the actual residuals.**

**The author has chosen instead to reference a conference presentation that contains figures showing the spectral residuals from ACE-FTS studies. While this seems mostly calculated to avoid sharing authorship, it is a viable approach because the conference has assigned each presentation a “doi.” However, there is no text associated with the figures (being an oral presentation that the author described verbally at the conference), which could make it difficult for the average reader to interpret what they are seeing. I suggest it would be appropriate to mention in the current paper which specific slides are of interest and to provide some text in the current paper describing the salient details of the figures in the presentation. While not as convenient as having the figures in the article itself, this should make the discussion less vague.**

I am glad the reviewer agrees that the new dataset is a significant improvement to that of Varanasi. As explained above, the point of this discussion is not to illustrate how the ACE retrievals are improved by the new dataset. The characterisation of this improvement is a study in itself, one which I plan to carry out in the future. Please note that nowhere in the text do I state that systematic features “go away”.

I have now added text to the manuscript identifying the two slides from the HITRAN conference presentation which are applicable to this work. The reference to this presentation is intended to keep the number of figures down, particularly as I have stated earlier that the improvement in ACE retrievals is not the focus of this paper.

As discussed above, once v4.0 ACE-FTS processing is complete (hopefully next year), I have plans for a study on the improvement of CFC-12 retrievals, where authorship will be shared as appropriate.