

The paper by Takeda et al reports the first attempt at a long term analysis of HFC-23 from ground-based FTIR. There are very compelling reasons to analyse this dataset based on the importance of HFC-23 as a strong greenhouse gas (100 year GWP of over 12000), and the fact that it is one of the HCFC family of chemicals whose production will not be phased out until 2030 under the Montreal Protocol for ozone protection. But its potential climate impact will remain for some time due to stockpiles and its long-lifetime. The addition of the NDACC network to long term measurements of this gas would be very welcome. However, this gas has not been a traditional target for this network as retrieving this gas from IR spectra is very difficult and challenging.

The purpose of this paper therefore is to outline, in some detail, how to retrieve total columns of HCFC-23 from IR spectra, obtain an annual trend and compare this trend against well-known and calibrated in-situ data. To retrieve this molecule is very difficult, so it will be of high interest to the NDACC FTIR community to follow this method. Indeed this will potentially unlock a worldwide dataset of measurements from Pole to Pole, linking in with the existing in-situ AGAGE network, and also offer a validation tool for satellite records.

The paper does indeed give a great deal of detail on the methodology, using one of the main software tools (SFIT4) used by the NDACC. This means the community can repeat this method without too many problems. However, the absorption signal from this species is so small (0.5%), and the spectra from filters 7/8 that are used throughout the network are relatively noisy, that without an independent dataset (AGAGE) the trends reported from this work would have been very difficult to justify.

The Japanese group leading this research have been involved in the NDACC for many years and therefore have significant experience in operating FTIR spectrometers and also analysing IR spectra.

The manuscript itself is a mixed bag in terms of its use of English language. The first 6 or so pages of text, abstract, sections 1 and 2, flow well and have been written well. However after this, section 3 has many instances of poorly written English, and use of English terms that are inappropriately used. Given that there are a number of English speakers in the coauthorship list, it would seem that they have not carefully commented and/or made changes in the manuscript that are necessary. This is an important paper to get out into the community so this referee has gone to the trouble of pointing out most of the issues. Some may have been missed; so coauthors read carefully!

This is an important paper for the FTIR community and a very useful outline of a method to analyse IR spectra for other researchers to follow. This paper therefore should be published, subject to the corrections/comments listed below

Comments:

Section 1 and 2: good background and well written. One change; section 2.2, line 29 had => has.

Section 3.1

1. Page 6, Line 20: "is a measurement noise." => "is the measurement noise."
2. Line 23: "which are often so-called Jacobian" => "also called the Jacobians"

3. Line 27: "is a gain matrix, whose line elements are so-called contribution function, which mean inversion sensitivity." => "is the gain matrix, or contribution function, which represents the sensitivity of the retrieved parameters to the measurement."
4. Line 29: "relationship of" => "relationship"
5. Page 7 line 2: "is a covariance" => "is the covariance"
6. Line 6-7: "Comparing the equation (3), which is neglected the error terms of the forward model parameters and the measurement noise" => "Comparing equation (3), which neglects the error terms of the forward model parameters and the measurement noise"
7. Line 9: "is usually non-linear problem," => "is usually a non-linear problem,"
8. Line 16: "profile is found by the iteration" => "profile is found by iteration"
9. Page 7 line 2: "More detail is described in the following." => "More detail is described in the following sections."
10. Line 8: "in maximum, about +5% relative " => "about +5% relative"
11. Line 9: "offset in measurement spectrum with second order polynomial" => "offset in the measured spectrum with a second order polynomial"
12. Line 11-12: "On the other hand, the continuum level, which is equal to 100% in transmittance, was fitted by the following, because the shape of the continuum level is caused by the optical characterization of the FTIR instrument" => "On the other hand, the continuum level, which is equal to 100% in transmittance, has a shape that is caused by the optical characterization of the FTIR instrument"
13. Line 19: "(the Fraunhofer lines)," => "(the Fraunhofer lines), and"
14. Line 26: "has always been maintained best optical alignment." => "has always been maintained with the best optical alignment."
15. Page 9, line 26 : "is temperature coefficient" => "is the temperature coefficient"
16. Line 29: "in mid-infrared region" => "in the mid-infrared region"
17. Page 10, line 3: "pressure data by National" => "pressure data obtained from the National"
18. Line 5: "profiles by the COSPAR" => "profiles from the COSPAR"
19. Line 8: "were basically used." What does this mean exactly? Are there other profiles including HFC-23 that were not from WACCM?
20. Line 9-10: "For HFC-23, the WACCM does not compute its profile and thus a priori profile of HFC-23 was based on globally and annually mean mole fraction profile by two-dimensional" => "For HFC-23, WACCM does not provide a profile and thus the a priori profile of HFC-23 was based on the global and annual mean mole fraction profile by the two-dimensional"
21. Line 12: "ppt at ground," => "ppt at the ground,"
22. line 14: "ppt at ground" => "ppt at the ground"
23. Line 22: "In retrieval of atmospheric" => "In the retrieval of an atmospheric"
24. Page 11, line 2: "respects" => "conserves"
25. Lines 8-15. Please explain exactly how the Se "ad hoc" S/N was determined. In SFIT4 version 0.9.4.4 the actual S/N is recorded in the header of the input spectral file (commonly called the t15asc) for each micro-window. So while the Se matrix should in principle reflect the S/N for each spectral point, the implementation is such that a representative S/N for the whole window is applied. The ad hoc method is to scale this S/N to produce a retrieval that is both stable and has suitable dofs. This is part of the Steck procedure (4.D) to get α . What was the effective S/N for an α of 100?
26. Line 24: "which has been shown in the Atlas" => "as suggested in the NDACC IR reference microwindow Atlas"
27. Line 26: "Since an H₂O absorption line having E'' of 586.48 cm⁻¹ is in the MW" => "Since a H₂O absorption line having an E'' of 586.48 cm⁻¹ is in the MW"

28. Line 27: "temperature dependence of line strength are small" => "temperature dependence on the line strength are small"
29. Page 12, line 2: "on the HITRAN" => "on HITRAN"
30. Line 23: "and fixed profile" => "and a fixed profile"
31. Page 13, line 3: "values in profile" => "values in the profile"
32. Line 6: "with the one" => "within one"
33. Line 7: "with" => "within" ; both instances in this line.
34. Line 18: "for both all retrievals" => "for all retrievals"
35. Section 4.1: on the question of the dofs for HCF-23. The regularization strength α is 100 which is quite strong. So this is a column scaling in effect, so the dofs will always end up being 1.0, regardless of how much information there is in the spectrum. There should be no surprise that the total column will be the only possible product from this method. That is, the dofs are being entirely driven by the method (Tikhonov), rather than the inherent spectral information.
36. Line 21: "Subtract" => "Subtracting"
37. Line 28: "from target gas" => "from the target gas"
38. Page 14, line 2: "established well" => "well established"
39. Line 23-24: "and the ones for other species were set to the values calculated from the used WACCM datasets." => "and the uncertainties for other species were set to the values calculated from the appropriate WACCM datasets."
40. Line 28: "uncertainties on the" => "uncertainties reported from the" ; NCEP has standard errors that are reported for their data which you can reference
41. Page 15 line 1: "we set" => "we set to"
42. Line 4: "and then their uncertainties" => "so that their uncertainties"
43. Line 5: "more affects" => "has a larger effect on"
44. Line 6: "error to" => "error on"
45. Line 12: as mentioned earlier above, what is the ad hoc S/N exactly quantitatively?
46. Line 19: "closed" => "close"
47. Lines 21&22: "to the" => "on the"
48. Line 31: the caption for table 4 is a little misleading. The word "uncertainty" is not correct, replace this with "error"
49. Page 16, line 17: "observation (Vömel et al., 2007) which was executed" => "observations (Vömel et al., 2007) which were flown"
50. Line 26: "and of a" => "and a"
51. Line 29: "biases lead underestimation to the trend on the retrieved" => "biases lead to an underestimation of the trend on the retrieved"
52. Line 30: "curvature is considered to the background correction." => "curvature of the continuum is considered when applying the background correction."
53. Page 17, line 17: "as the" => "as in the"
54. Page 18, line 6: "to the HFC-23" => "to HFC-23"
55. Page 19, line 27 "remaining of" => "remaining"
56. Line 32: "region" => "regions"
57. Page 20, line 4: "negative difference of average" => "negative average difference of"
58. Line 10: "from 260 K to 300 K approximately." => "from approximately 260 K to 300 K."
59. Line 11: "ppt as" => "ppt as the"
60. Line 26: "of lower" => "of the lower"
61. Line 27: "this should be done to further understand the reason for the negative bias and the seasonal cycle by intercomparison with the retrievals using the observed spectra at other"

=> "further study should be undertaken to understand the reasons for the negative bias and apparent seasonal cycle by an intercomparison with HCF-23 total columns at other"

62. Page 21, line 3: "as green" => "as a green"
63. Lines 5, 7, 9,11: "annual change rate" => "annual growth rate"
64. Line 14: "Considering with the above, it is obviously indicated" => "Considering the above, it would seem"
65. Line 20: "strategy basically agree well" this is a very vague assertion. How do they agree?
Perhaps it is better to write something like "strategy agree well within the errors"
66. Line 21: "These results indicated" => "These results indicate"
67. Page 22, line 22: "from the Eurasia." => "from Eurasia."
68. Line 28: "of lower" => "of the lower"
69. Lines 29,31: "annual change rate" => "annual growth rate"
70. Line 33: "ground-based measurements." => "ground-based in-situ measurements."
71. Page 31, table 4: It might be useful to add to this table the assumed uncertainties for each parameter that led to the computed error terms. Note the comment earlier about changing the caption. If however the uncertainties are included then the caption would read "errors and uncertainties"
72. Page 38, figure 7: what exactly are the R values computed with, ie, the FTIR data against all in situ data for each site?