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## ***Interactive comment on “Satellite observations of stratospheric carbonyl fluoride” by J. J. Harrison et al.***

### **Anonymous Referee #2**

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#### General Comments

The paper of Harrison et al. reports the first global measurements of carbonyl fluoride (COF<sub>2</sub>) from infrared sounders (ACE-FTS and Envisat MIPAS) and a comparison with chemistry-transport model simulations (SLIMCAT). The paper provides detailed descriptions of the COF<sub>2</sub> retrievals as well as the global distributions (zonal means, time series, trends) found in the three data sets. The paper presents an interesting topic within the scope of ACP. It is well written and should be published once the following comments are addressed.

1) I would recommend adding a paragraph in the introduction that provides the scientific motivation for measurements and modelling of COF<sub>2</sub> from a broader perspective.

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Why are global COF2 measurements needed? Are they mainly needed to validate and improve modelling of COF2 in the stratosphere? Do atmospheric models such as SLIMCAT have specific deficiencies representing the photochemistry of COF2 or its source gases? Or are the measurements particularly useful to validate the representation of transport and mixing in the stratosphere? Are there other scientific motivations to measure and model COF2?

2) From the perspective of a retrieval scientist I would like to ask for a more detailed description of the vertical resolution and sampling of the ACE-FTS and the SLIMCAT data sets. For MIPAS the averaging kernels and estimates of the vertical resolution are presented, which is fine. For ACE-FTS it would be good to provide these information in section 3.2, for example. I also got the impression that the averaging kernels (or at least some estimates of vertical resolution) are not considered in the comparison of the different data sets (section 4-6)? This might be okay, if differences in vertical resolution are not too large, but this should be made more clear in the paper, I think.

#### Specific Comments

p18129, I5-9: Perhaps add a sentence explaining the distinction between "organic" and "inorganic" forms and why this is important here?

p18130, I25-p18131, I10: Suggest to add references to the individual sections of the paper in this paragraph (which is describing the structure of the paper).

p18131, I20: Does "vertical resolution" refer to the vertical sampling of the ACE-FTS measurements or the vertical resolution of the COF2 retrieval?

p18131, I25-26: "ACE-FTS records [...] spectra over a large portion of the globe" sound a bit vague, I think.

p18132, I18-20: It would be good to mention the vertical sampling (1km?) of the retrieval grid here.

p18134, I9: Suggest to replace "error covariance" by "covariance", as S<sub>a</sub> refers to the

variability of the atmospheric state (in optimal estimation theory).

p18135, l19-21: This paragraph is too short. It provides a detail on retrieval grids (related to interpolation of data) in the middle of other information. Perhaps this information should be given earlier, e.g. somewhere near the description MIPAS tangent heights and retrieval grid?

p18138, l5-6: "MkIV and ACE-FTS v2.2 profiles from 2004 and 2005 agree well within the measurement errors." Which relative or absolute difference would that be?

p18139, l1-17: Did you calculate the median absolute deviation or the standard deviation of the individual fractional retrieval errors to get the statistics?

p18139, l18-p18140, l8: I found this description of the approach to handle the top column data in the ACE-FTS retrieval a bit long and confusing. I understood that an a priori profile from ATMOS was used above a latitude-dependent upper boundary, which was scaled by a factor that was retrieved for each profile. However, this approach causes errors, which are currently not considered in your systematic error budget, right? Please clarify.

p18141, l2: The term "retrieval gain matrix" is used, but it was not explained before. It may be introduced with Eq. (5) and could be used to simplify Eq. (6).

p18141, l6-8: The term "Spectroscopic errors in interfering molecules" confuses me. Do you refer to all the spectroscopic errors from trace gases other than COF2? However, at the end of the sentence you are referring to COF2?

p18141, l11-14: This is another very short paragraph referring to the measurement and retrieval grids somewhere in the middle of other information. I think it would be better to collect these information in fewer places. Please try to sort this out.

p18141, l15: I would not say that the averaging kernel matrix describes the "quality" of a retrieval. Perhaps "sensitivity" (to measurement information and a priori information) is a better word?

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p18141, I22-23: Why do you show polar profiles for spring equinox? Wouldn't it be more reasonable to show polar profiles for January or July, representing the different atmospheric conditions of polar summer and polar winter more clearly?

p18142, I13-15: "... near-coincident profiles are expected to be similar but not necessarily identical" is a bit vague. Perhaps clarify by saying "... are expected to agree within the error budget and vertical resolution (AVKs) of the retrievals"?

p18142, I18-19: Are these error bars representing the full (systematic) error budget or just the retrieval covariance?

p18142, I21-23: You could convolve the ACE-FTS profile with the MIPAS AVKs to test this hypothesis. Further, I noticed that Fig. 3 (bottom, left) shows a dip in the MIPAS profile which is not present in the ACE-FTS profile. What is the reason for that?

p18142, I24-p18143, I1: The approach for outlier filtering for the ACE-FTS data remains unclear. How are "significant outliers" defined?

p18143, I8-9: Would this diurnal variation be expected based on COF2 photochemistry? (If this is relevant I think the solar local times of the ascending and descending Envisat orbits should be mentioned in the paper.)

p18143, I19-20: A forward reference to section 5 of the paper would be good. (I was first tempted to think the secondary maximum is a measurement artifact, but you found that nice explanation that it is caused by the HCFC-22 source.)

p18144, I3-6: The ACE-FTS zonal mean cross-sections appear noisy also at mid-latitudes (e.g. 10/2009 or 03/2010), and not just in the tropics, in particular?

p18144, I14-16: I would first like to clarify that you indeed plotted the "standard error of the mean" ( $\sigma/\sqrt{n}$ ) and not the standard deviations ( $\sigma$ ) of the data? I think the calculation of the standard error of the mean is based on the assumption of independent data? Is this applicable here, where individual profiles are likely correlated due to atmospheric chemistry and dynamics? I was wondering if it wouldn't make

sense to show standard deviations instead of standard errors to illustrate atmospheric variability?

p18144, l18-19: It seems the ACE-FTS error bars are largest at upper altitudes, but not necessarily at the upper boundary of the altitude range of the retrieval? Is this due to the upper boundary of the retrieval varying with latitude?

p18144, l18-29: I found it a bit difficult to identify the different altitude ranges you are referring to in this paragraph. There are the "highest altitudes" and the "very highest altitudes" and several times you refer to "at these altitudes". Perhaps give some numbers to clarify?

p18144, l21-22: It seems not all of the ACE-FTS mean profiles are flat? For instance, the profile at 25-30S has a distinct dip at 35-38 km, which is not captured by MIPAS. I was wondering if this dip in the ACE-FTS profiles is related to the approach used to handle the top column data in the retrieval? (If the top column is over-estimated for some reason, the retrieval would likely compensate by underestimating the COF2 vmrs just below the top column?)

p18145, l2-4: Would be good to mention the actual number of profiles in the averages (e.g. in the plot title or the figure caption).

p18145, l15-16: What is the vertical grid spacing of the model/simulation in the stratosphere? Is it comparable to ACE-FTS and MIPAS?

p18146, l22-24: What are the relative contributions of the photolytic and O(1D) loss mechanisms?

p18147, l3-4: It is not clear to me why the analogy between COF2 and NOy is important or relevant here?

p18147, l4-6: Is it important to mention the compact correlations between COF2 and CFC-12? (The tracer-tracer method was not used here to estimate the COF2 lifetime.)

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p18147, I19-21: I found it surprising that the ACE-FTS two-year means (Fig. 11) show the same noise-like features as the one-year data (Fig. 8) in some cases. For example, in both figures there are noise-like structures/oscillations at 10-30N and 30-45km in June. (Are these features related to individual outliers in the ACE-FTS data rather than noise?)

p18148, I1-12: Following the order of the figures it may make more sense to shift this paragraph (which is discussing Fig. 9) before the preceding paragraph (which is discussing Fig. 11)?

p18149, I1-22: A separate figure or additional plots for SH/NH polar summer in Fig. 10 may help to illustrate that the HCFC-22 source is causing the secondary polar summer maximum.

p18151, I8-10: I recall that the MIPAS data coverage also varies with latitude, with maxima occurring at high latitudes?

p18153, I2-4: It seems the ACE-FTS trends are more noisy than the MIPAS and SLIM-CAT trends? Please comment.

p18155, I21-22: I think the 30% high bias of MIPAS needs some more discussion. Is it related to uncertainties of the spectroscopic data (noting that ACE-FTS and MIPAS retrieval use rather different microwindows for the retrievals)? Or is the bias related to the different vertical resolutions and/or a priori influences of the different retrievals? Or are there other reasons?

Table 1 and Table 3: The ACE-FTS and MIPAS retrievals use rather different microwindows. Except for one window near 1234/cm all other microwindows are different. I found that a bit surprising. Please comment.

Fig. 2: How many spectra have been averaged?

Fig. 3: The retrieval error due to spectroscopic errors is rather small. I thought it would be about 15-20% according to the discussion presented in section 3.1? I guess "day"

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in the plot title refers to "mid-latitude day-time"?

Fig. 4-6: All the curves are rather thin and the colors are hard to distinguish on my printed version of the paper. "DFS" in the top left plot is clear (although not explained in the paper), but what is "INF" referring to? Do you show the median profile to illustrate that the selected profile is close to it and that its diagnostics can be considered to be representative for a large fraction of profiles? The retrieval error at 80S (Fig. 4, top, right) is at 8-10%, whereas Fig. 3 shows that the estimated retrieval noise is >15%. I guess this is due to the different atmospheric conditions (mid-latitudes vs. spring equinox)? The integral of the AVKs should be displayed in the response plot (bottom, left) to indicate the fraction of measurement information in the retrievals. Fix plot title in vertical resolution plots (bottom, right).

Fig. 9: It is a bit difficult to see the mean ACE-FTS curves due to the broad error bars.

#### Technical Corrections

p18128, l18: spell out SLIMCAT acronym

p18128, l25: suggest to use brackets for trends, i.e. "(0.85+/-0.34) % year<sup>-1</sup>" rather than "0.85+/-0.34 % year<sup>-1</sup>" (also in other places below)

p18130, l26: "...and the investigation more fully of COF2 trends," may need to be reworded (?)

p18131, l3: spell out SCISAT acronym

p18137, l10: "Q branch" rather than "Q-branch" (to be consistent with other places)

p18137, l20: there is no "on the one hand..." before "on the other hand..." (?)

p18141, l29: suggest to use " $\Delta z_i$ " or "dz<sub>i</sub>" as symbol for grid spacing

p18143, l24: suggest to replace "very low" by "low"

p18146, l16: write "...\_at\_ ~20-40 km" (?)

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p18147, l21: perhaps "averages of monthly averages" or just "averages" would be more clear?

p18151, l26: write "\_which is\_ stronger.." (?)

p18152, l21: "that" instead of "which" (?)

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Interactive comment on Atmos. Chem. Phys. Discuss., 14, 18127, 2014.

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