AUTHOR'S RESPONSES TO RCC2347 : CHRIS SIORIS REVIEW

The authors wish to thank the reviewer for his thorough reading of the discussion paper, and subsequent extensive corrections and suggestions. We consider the modifications applied have helped to produce a much improved manuscript. We reply to his points below with the reviewer's comments in bold, and our responses in italics.

I found the section on the optimization of the spectral fitting window to be very good (incl. Fig 3). The homogenization of vertical columns by applying the reference sector method is an interesting result. The idea to have study regions is also meritorious. I was also pleased to see the use of orthogonalized temperature-dependent O_3 absorption cross-section spectra.

There are other superior spectral fitting techniques that have not been mentioned such as weighting-function DOAS. One criticism is that the authors have some minor difficulties communicating in English. Also, there are several instances where the authors claim to be providing analysis of results, but they are simply speculating because they do not offer proof to support their statements or the proof they provide is inadequate. There are other occasions where the reader requires further elaboration by the authors to understand the points they are making.

I am also troubled with their use of statistics. For example, they look at the RMS of the residuals to decide whether the inclusion of a basis function is warranted. However, any basis function has the capability of reducing the RMS of the residuals, so an appropriate statistic is required. They have one available in terms of the standard deviation of the mean but they could explore even more appropriate statistics. I am not sure if QDOAS provides a χ^2. The statistic used to judge whether the inclusion of an additional basis function is warranted must account for the additional degree of freedom used.

We do have the χ^2 statistic available for the work, and agree with the reviewer's comments that RMS will always decrease in parallel with an extra number of fitted parameters (noted in the original manuscript – P7101L20). Therefore in order to take this into account, where necessary we have now replaced the RMS residual with fit χ^2, and included a description of the χ^2 statistic relevant to our work in section 2.

Eq. 1 – The quantity in the brackets in the numerator should be squared. This must simply be a typo otherwise the RMS values in Table 2 are not very good.

Correct, this is a typo. The RMS description is now removed from the text following updating to use of χ^2 statistics.

P7101L26 "...three step pre-fitting" → "...three step fitting..."

Amended

Section 3.1.1 – The authors have tested the idea of pre-fitting but have not used the non-DOAS method of Chance et al. (2000), which has the advantage over the DOAS method used in this work in that a polynomial scales the absorption optical depth as a function of wavelength (for both pre-fitting and fitting cases). This should be noted. The issue with pre-fitting, particularly with a DOAS approach, is that BrO and O_3 slant columns are fitting window dependent primarily because of the wavelength dependence of atmospheric scattering. For O_3, this will be a more significant problem since the mean wavelength for its pre-fitting window is significantly shorter than the mean wavelength of the HCHO fitting window. A comment here about using WF-DOAS or any more sophisticated fitting technique that does not simply fir cross sections to (differential optical depth spectra) in the future would be well placed. Limiting to SZA <60 mitigates this issue.

Section 3.1.1 has been substantially revised with the reviewers comments in mind. In particular the third paragraph has been re-written to accommodate comments on different fitting regions producing different results for their respective trace gas, as well as mentioning the application of the WF-DOAS method to minor absorber retrievals in the UV. We have also emphasised the use of only those scans observed at <60 deg to alleviate the pre-fit problem. However, with regards to the WF-DOAS method, the O_3 cross sections applied in our retrieval are weighted by a mean slant column amount to correct for the I_0 effect. This will mitigate for the saturation of the stronger absorbers, but not the difference in radiative transfer WF-DOAS also accounts for. We also take on board the referee's comments regarding non-DOAS fitting modes. However, given the authors lack of experience *with intensity fitting methods, testing of this technique is beyond the scope of this paper.*

Section 3.2.1 – This section was essentially pointless to me because an apple-to-apples comparison of Ring basis functions was not because of the slit function issue.

We have now generated Ring spectra following the Chance (1997) method, convolved to the same RAL slit function as applied elsewhere in the paper. This section and the results table are updated with these new results. Results are poorer with the single Ring cross section compared to the two cross section Vountas method.

Section 3.2.2 – The authors speculate that the larger fitting window (extending to >350 nm) are "substantially affected by unquantified fitting interference with O_4". The source of interfering spectral structure could be from the instrument or Earth / atmosphere for example. to demonstrate that O_4 is the likely source of the interference, the authors may choose to show the exponential decay of the residual feature as a function of cloud top height or simply remove the speculation. As a result, I disagree with the authors' conclusions regarding whether O_4 should be included in the optimized fit.

We agree with the comments above, and therefore remove the last two sentences in section 3.2.2. We include the reviewers comments on the unspecified instrumental and / or atmospheric effects on the fit, as well as enhancing the discussion on the interplay between the polynomial, fitting region and O_4.

I think that the authors should state their literature reference for the absorption spectroscopy of the $(O_2)_2$ collisional complex (O_4), and for OClO if they don't remove OClO from the paper (see below).

Greenblatt et al., (1990) O_4 and Bogumil et al., (2003) OClO cross section references added (note for typesetter, these have also been added to bibitems at the bottom of the source document).

Section 3.2.3 – This section could be omitted since OClO is not expected to be found for SZA<60. Including this absorber has no physical basis.

As OClO has been commonly included in previous HCHO retrievals for similar instruments (e.g. GOME-1, SCIAMACHY (De-Smedt (2008)), we use this section to confirm that its inclusion actually appears to be unnecessary for this instrument.

P7106L26 "concentration" → "slant column"

Amended

Section 3.2.5 – The use of 'add back' is misleading in my opinion. I believe the authors are referring to the slant column that is used in generating the I_0 corrected cross section following Appendix A of Aliwell et al. The SCD used to generate the I_0 corrected cross section should be stated for each species. Test 3f is apples-to-apples only if the O_3 slant column used to generate the I_0 corrected cross section is 0.8×10^{19} moles / cm^2. A dynamic I_0 correction might be more appropriate, where the O_3 SC used to generate the I_0 corrected cross section is obtained from the BrO pre-fit but this is probably not too important for HCHO since the data of the latter species is probably discarded in post-processing for highly perturbed O_3 conditions. Figure 5 should extend to 2.3×10^{19} moles / cm^2 to cover the O_3 SC shown in Figure 6.

We have replaced 'add-back' with 'slant column' where relevant, and emphasised the Aliwell reference. Values used for the slant column correction are included in the text for all fitted absorbers, with the O_3 value indeed being the same as specified in Table 1. Figure 5 is extended to cover the correction range retrieved for Figure 6.

Section 3.2.6 – To determine the suitable range of cross-section temperatures, an extreme atmosphere should be considered (tropical) rather than a mid-latitude one since temperatures at the tropopause near the equator can range as low at 195 K and are warm in the upper stratosphere. At these latitudes, 228 K may be too warm as the lower T of the cross-section pair.

We agree with the reviewers observation, and revise the temperature ranges studied (200–230 K lower, 230–260 K upper). Please also note, the author identified an error in the plotting code for Figure 6 which meant the surfaces were previously plotted inversely. This has now been rectified. Text in section 3.2.6 has been modified accordingly. We have also removed references to fit RMS in the HCHO cross section temperature discussion.

P7109L7 "...mean tropospheric temperature..." → "a single effective temperature for tropospheric HCHO absorption..."

Amended

Section 3.3 – Remove "reduction in width" (L14) since reducing the width reduces the need for a higher order polynomial. The authors should not use residuals as a statistic to justify whether the polynomial order is sufficient (see introductory comments). The last sentence of this section should be removed. The can discuss 1sigma instead or use more relevant statistics. P7109L25 "sufficient" is not quantified. Proper statistical tests demonstrate sufficiency.

Section now amended to discuss χ^2 values.

P7110L9 "lower UV" is unconventional. Suggestions : "UV-B" (for <320 nm), "short UV wavelengths".

Amended

P7111L9 – Standard deviations are slightly higher globally with undersampling correction. The authors have not demonstrated using appropriate statistics that the use of undersampling spectra is justified in the spectral fitting. Again, they have failed to account for the fact that they are using an extra basis function. Until some revision is made, "strongly" should be removed from the last sentence in this sub-section.

χ^2 discussed instead of RMS residual, taking into account extra absorbers, se we are now able to verify fit quality decreases without undersampling correction.

Section 3.4.3 – It is not clear how a linear offset term in the DOAS fit is not already included by virtue of the 5^{th} order polynomial. Again, the use of the RMS statistic (P7111L25) is not appropriate to make a judgement on the utility or necessity of a basis function. The standard deviation could be used, after the authors have clarified the above issue (re: 5^{th} order polynomial) to the reader.

The linear offset term is included in order to fit structures in spectra at a higher frequency than the polynomial term is able to subtract in analysis. From the tests this remains valid even at higher polynomial orders (e.g. 5^{th}). Figure 1 details the polynomial (5^{th}) and linear offset term (1^{st}) for the retrieval plotted in Figure 2 in the AMTD paper; both of which (polynomial and linear offset) differ significantly from each other. We also update our fit quality statistics as previously mentioned, which also demonstrate fit quality significantly decreases without a linear offset term. Similarly, fit quality decreases with application of lower order polynomials in conjunction with the same linear offset. From this we infer the 5^{th} order polynomial alone does not model high frequency structures in the spectra to the same level as our offset term.

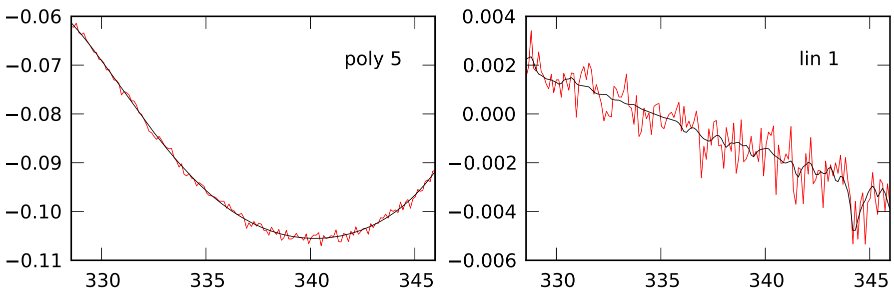

FIGURE 1. Polynomial and linear offset terms from the same retrieval illustrated in Figure 2 of the ACPD text.

Section 4.4 – This section needs substantial revision. It is not really the resolution that enables different cloud fraction thresholds to be tested but rather the spatial sampling frequency. Suggested wording: "The high spatial sampling frequency ... provides the opportunity to test a higher rejection..." *Amended*

" To which "statistic" the authors are referring: "..the statistic is affected..."

Amended

The authors claim significant "above cloud enhancements" are occurring in the AMA region. I doubt this since the majority of the clouds are at 600 hPa (4 km) or lower pressure (higher altitude) as shown in Figure 10 and as stated in the text. Then the authors contradict themselves by stating that the Amazon has dense, low cloud cover (on 9 Aug 2007). This should be removed. Figure 10 shows that the highest formaldehyde columns in the AMA region are occurring when the cloud tops are low for partly cloudy scenes. This suggests that formaldehyde is below 600 hPa (near the surface... as expected).

Agreed, and removed.

The authors also state that the "AMF (takes) a greater fraction of cloud albedo into account". Why would the cloud fraction be higher in the AMF than in the observations? My guess is that there is an issue with the AMF simulation (oversimplification of cloud radiative transfer) for mostly cloudy scenes that leads to the increase in vertical columns with increasing cloud cover. An increase in the vertical column due to 'above cloud enhancements' would also be seen on the slant column, which is not the case (Figure 9) in either hotspot region.

Indeed, CF is not higher in AMF than observations, this section has been removed from the text. We acknowledge the reviewer's comments; including the following at the end of section 4.4: "The vertical column is seen to increase in parallel with an increased cloud cover at CF >0.4, which is not noted for the corresponding slant columns. Whilst this offers a useful indicator of an appropriate CF threshold, it also shows the AMF is providing a sub-optimal conversion value for such scans, due to an un-realistic interpretation of radiation transfer above, within, and below clouds. "

P7116L9 The offset correction (tests 5c-d) appears to not "be a smaller effect" than the I_0correction (test 3f), so I suggest that one of these concluding statements if revised.
P7116L11 "... equal importance ..." is probably too strong. How about "... high importance ... "

Amended to ".. and offset corrections also have an significant impact on the fit residual. Subtle effects on fit quality and HCHO slant column are also noted with instrumental corrections, such as wavelength calibration, undersampling and scan bias correction, although these should be probably weighted with a similarly high importance..."

Technical Amendments:
All the following points have been amended as per the reviewer's comments
P7096L20 "... occur to ..."→ "... occur near ..."
P7097L17 "... its' ..."→ "... its ..."
P7098L16 "... on an assumed / known ..."→ "... on assumed or known ..."
P7099L22 "... example ..."→ "... sample ..."
P7099L26 "... statistic ..."→ "... statistics ..."
P7101L13 "... in Theys et al. (2011)'s original retrieval ..."→ "... in the retrieval of Theys et al. (2011). ..." [avoid the possessive form]
P7102L16 "... serve ..."→ "... serves ..."
P7102L27 "... removal ..."→ "... exclusion ..."
P103L1 "... and ..."→ "... an ..."
P7105L2 "... minima ..."→ "... minimum ..."
P7105L9 "... finding ..."→ "... where it is found that ..." P7105L15 "higher"→ "wider" [this unconventional usage appears in several instances]
Section substantially re-written, the above are no longer present
P7106L19 "... 0.31 ..."→ "... by 0.31 ..." *Amended (value amended to χ^2 also)*
P7106L22 "high"→ "large"
P7108L22 "... 0.11% per extra K. ..."→ "... 0.11%/K. ..."
P7108L28 "... the the ..."→ "... the ..."
P7109L2 "... x 10^{-7}."→ "... 10^{-7}."
Line removed from manuscript
P7109L4 "... spectra recording. However... likelihood ..."→ "... observation. However... weak likelihood ..."
P7109L18 define "SC"
P7110L11 "... vectors ..."→ "... spectra ..." [also at P7110L15, use polarisation spectra]
P7110L22 "... retrieval based ..."→ "... retrieval-based ..."
P7111L7 "... as a pseudo-absorber ..."→ "... as pseudo-absorbers ..." [there are two of them right?]
We calculate our own undersampling spectra as per Chance (2005) and fit only one in the retrieval.
P7112L17 "... accounted for ..."→ "... taken into account ..."
P7113L7 suggested rewording: "SCs from the reference strip ... are fitted with a 3^{rd} order polynomial over all latitudes, and the latitude-dependent fitted SC from this strip is subtracted globally from each day's measurements to serve as a daily correction."
P7114L17 It is not what is meant. Suggested rewording: "... due to the much smaller relative differences when vertical columns are considered instead of SCs."
P7114L22 Don't start a new paragraph

P7115L9 "... higher signal to noise ratio ..."→ "... less variability ..."

P7115L10 "... in slant columns whose ..."→ "... in the number of observations for which the ..."

P7115L26 "... the contrasting vertical column increase" . The vertical columns increase relative to what or as a function of what? I assume the authors mean: "... the contrasting behaviour of the vertical columns versus cloud fraction threshold between the two regions..."

Table 1: "I_0-corrected to ..."→ "I_0-corrected using ..."

Figure 9 caption: "Removing..."→ "Altering..."

Figure 10 caption: You should define "high CF" since it appears that not all high CF pixels are in the 300–600 hPa region. One point with a high CF, for example, is near 900 hPa and shows a HCHO SC of 1×10^{16} molec / cm^2.