Interactive comment on “Airborne multi-axis DOAS measurements of atmospheric trace gases on CARIBIC long-distance flights” by B. Dix et al.

Anonymous Referee #1

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The authors report on the installation of a DOAS system as part of the CARIBIC project instrument container and present some first measurement results. The instruments are operated once a month onboard a passenger aircraft on long-distance flights and provide valuable observations of atmospheric trace gases. The integration on a passenger aircraft yields new possibilities regarding continuity, wide regional coverage, and trace gas measurements in the tropopause region. Measurements during take-off and landing give indications on trace gas profiles. In principle, observations are possible in three different viewing directions. Some previous problems in two viewing directions are discussed and have been solved now so that in future the analysis of multi-axis measurements should be possible on a regular basis. The paper gives a well understandable explanation of the technical installation of the measurement sys-
tem. Some first scientific results on the trace gas retrievals are presented. In this part, the authors do not explain their analysis well enough, open questions remain. Not all necessary background information and assumptions that enter their evaluations are given. Clarification is in any case necessary to judge the relevance and usefulness of their measurements and analysis.

After the following comments and requests have been taken into account, the paper should be published in AMT. The most important requests concern the revision of the scientific results section, especially the explanation of the HONO analysis.

**Specific comments**

Page 271, lines 6-16: Are fibre bundles with fibres of 210\(\mu\text{m}\) diameter (later changed to 150\(\mu\text{m}\) diameter fibres, p.275, l.7) a good choice for illuminating a slit of 50 \(\mu\text{m}\) width? Maybe there is a possibility for improvements here?

Page 272, lines 20-21: This sentence is misleading as the resolution (FWHM) is not determined by the pixel number of the CCD, but by the combination of spectrometer entrance slit width, the dispersion of the grating and pixel size. Maybe a sentence like this would be more accurate, e.g.: "The whole system provides a spectral resolution of 0.7 nm (FWHM)."

Page 276, line 16: The numbers given for the standard deviations of the fit residual reveal remaining systematic features. For purely statistical deviations, \(\sigma\) should reduce much more between the 30s and the 450s case (by a factor of \(\sqrt{15}\)). Some remaining systematic fraction is a common issue for DOAS retrievals. So this should be reformulated or an additional sentence included, as it now sounds as if hardly any systematic deviations remain. E.g.: "As \(\sigma\) is not as much reduced as expected for purely statistical errors, some systematic deviations in the residuals remain." By the way, this is also a good reason, why the authors’ choice is adequate to use the 30s spectra for the following analysis.
Page 276, lines 24-29 and Caption of Fig. 6: Please state more clearly which spectrum was used as background reference spectrum. The caption gives the altitude and SZA, but where was this taken, e.g. over polluted or clear region? How was this determined? This is important when judging the absolute values, because the result is a column difference with respect to the background spectrum.

Page 277, lines 4-21: The observed peaks in the dSCD of HONO, NO₂, HCHO are attributed to enhanced values while the simultaneous peaks in O₃ and O₄ are attributed to light path elongation. This finding is not obvious and needs to be properly justified as this cannot be seen in the data and information provided. The comparison with the in-situ data is surely of importance and supports the authors' interpretation, but cannot suffice as explanation for this discrimination. How was the light path enhancement calculated, what are the assumptions? Why is the influence different for the different trace gases? Here, also the answer to the previous question on the background spectrum is interesting as the dSCD of HONO within the cloud shows positive values up to \(3 \cdot 10^{15}\) molec/cm\(^2\) and falls to negative values of \(-2 \cdot 10^{15}\) molec/cm\(^2\). Does this imply that quite a large amount of HONO is also present in the background spectrum? I understand from Tab. 2 that the detection limit for these data is at \(2.5 \cdot 10^{15}\) molec/cm\(^2\). It would be interesting to see the fit quality (\(\chi^2\)) in addition to the trace gases in the time series.

Page 277, lines 23-27: If the enhanced HCHO values could be explained by "updraft of polluted air through deep convection" could this also be true for HONO?

Page 278, lines 1-8: I understand that the authors refer to a future publication for the detailed analysis of the HONO concentrations. Nevertheless, if the results are emphasized as new findings here, the procedure for the derivation needs to be clarified. I don’t think this should inhibit further publication of a more detailed analysis. Which assumptions enter these radiative transfer calculations? Please mention the code that has been used here - probably the same mentioned only later in another case (cp. page 279, line 29 ff)? The radiative transfer in deep convective clouds is a complex
procedure - does the comparison using the $O_4$ values result in reasonable cloud properties?

Page 278, line 24: The enhanced values in NO$_2$ and O$_3$ are not as clearly to identify as it sounds in the text. In the O$_3$ results, this is somewhat better visible than in NO$_2$, especially when considering its dependency on the SZA due to stratospheric light path enhancement. Maybe this connection to the SZA should be mentioned in this place, as this might support the authors’ interpretation which could be formulated somewhat more carefully.

Page 280, line 18: The following sentence is not clear: "Retrieved concentrations are likely to be underestimated owing to horizontal inhomogeneities". Inhomogeneities of which parameter are referred to? If inhomogeneous trace gas concentrations are referred to, I wonder why underestimations are more likely than overestimations. Maybe inhomogeneities in surface elevation are addressed? In case of surface elevations in the area of the aircraft ascent and descent, the retrieved columns would have less vertical extent than assumed and therefore concentrations would be underestimated as suggested. Please add a few clarifying words here.

Page 280, line 23: Owing to the problems with the ±10° directions that are openly discussed, maybe change the sentence to: "Most parts of the CARIBIC DOAS instrument performed reliably during the 30 flight operations." There is no question that the regular operation of this or a similar DOAS instrument in the presented manner is well feasible and represents a good opportunity for obtaining useful datasets. But a more cautious formulation would be more suitable here.

Page 281, line 9: I guess this statement is true for the nadir direction of the past data and might be also true for the ±10° directions in future.

Page 282, lines 9-11: What sort of changes, which different system are the authors thinking of? If a larger system is considered, wouldn’t this inhibit measurements during ascent and descent as stated on page 273, line 24? Is there a system that would fulfil
all requirements?

**Technical remarks:**

Page 267, line 24: The sequence of the references seems arbitrary, maybe this should be ordered either chronologically or by content.

Page 270, line 13: "direction" instead of "directions"

Page 271, line 11: Please remove the doubling of "the"

Page 271, line 28: I guess the authors mean "feed-through" instead of "feed-trough" here...

Page 272, line 3: ... and "trough" instead of "through" here.

Figure 1: It is confusing that the "nadir" direction has actually $98^\circ$ viewing angle and is plotted vertically in the picture. Therefore, the $98^\circ$ in the caption appears like a typo which it is not. So I recommend a slant viewing angle in the picture and quotations around "nadir" (also in the text).

Figure 6 and 7: Please add the units of the SCDs in the graphs.

Figure 10: The abbreviation FRS for Fraunhofer reference spectrum has not been used before and has not been introduced. Please use the full term or specify FRS (e.g. on page 275, l.28).