Authors' response to comments from Anonymous Referee #2

Referee #2

A review on 'Investigation of cirrus clouds properties in the Tropical Tropopause Layer using high-altitude limb scanning near-IR spectroscopy during the NASA-ATTREX Experiment' by Santo Fedele Colosimo et al. submitted to Atmospheric Measurement Techniques.

We would like to thank the anonymous referee #2 for agreeing to review the manuscript, providing valuable and constructive comments and suggestions for improvement. Responses to the main and specific comments are provided below.

This study shows a new method to derive IWC from ice water absorption derived from NASA ATTREX MiniDOAS limb scanning near-IR observations. A case study is used to interpret the retrievals, validations, and uncertainties. Sensitivity tests were performed using a radiative transfer model, and a conceptual model was derived to understand observations.

This paper is well-organized and well-written. However, I still have some concerns.

1. This study uses one case to show the performance of IWC retrievals from MiniDOAS, validated with in-situ measurements from Hawkeye FCDP and NOAA water instruments. Validation shows good agreement at some times but not that good at another time. The authors discussed possible factors that contribute to the uncertainty. However, with only one case, it is not that convincing how good the method is to derive IWC. I would suggest adding other cases (as complementary materials).

We thank the reviewer for bringing up this point. The main purpose of the paper was to develop and investigate the feasibility of using ice absorption to retrieve IWC within reasonable uncertainties, and to demonstrate the use of DOAS near-IR limb measurements as an additional tool to add to existing techniques for IWC retrieval in cirrus clouds. While we acknowledge that the validation would have benefited from a more extensive analysis of the entire data set, we decided to verify the methodology for a specific data segment during the ATTREX experiment, where the variability of the environmental radiative conditions of the measurements, such as solar position and altitude, allowed for a more thorough testing of the VLIDORT-QS (QuasiSpherical) RT model. As explained in Section 2.4, this RT model has been developed specifically for this project and the choice of that particular portion of the flight (Section 2.3) was driven by a continuous change in all of the RT parameters, which makes the validation of the RT code and the methodology more valuable within the scope of this study.

2. Please check the equations 10 -13.

We do not understand what specifically needs to be checked in equations 10 to 13. We do not see any errors in the equations as written.

3. Throughout the entire paper, the authors may state the same concept in various places but use different terminology, which is confusing. These include concentration, number concentration, mass concentration, particle density, also IWC and IWP. Please keep the terms consistent, correct, and easy to understand for readers.

We thank the reviewer for this helpful suggestion. We have carefully revised the manuscript to make the use of variables consistent.

4. An important conclusion in this paper is the high sensitivity of the method to detect and derive information for thin cirrus clouds. However, there is no comparison or discussion with other methods to prove the method's sensitivity.

The idea behind this study was to test the feasibility of detecting IWC in cirrus clouds from high altitude DOAS near-IR limb measurements, and a formal intercomparison of our methodology with other methods was not originally planned as part of the study. We did compare the retrieved miniDOAS IWC with other in situ IWC measurements taken on board the Global Hawk. The similarity of the results is considered enough given the scope of the paper. Furthermore, airborne IWC measurements in stratospheric cirrus clouds are limited, making a formal intercomparison between different techniques challenging.
Notwithstanding, we do understand the reviewer's point, and we have added the following sentence at line 479:

"*It is worth noting that the NOAA instrument has an IWC detection limit of 2-3 g* m^{-3} *for ideal conditions [Thornberry, 2015; Thornberry, 2017]. While our IWC detection limit is not as good as that for the NOAA instrument, our approach is not affected by sampling artefacts (i.e., ice shattering), and it is sufficiently different and sensitive to provide new insights into cirrus clouds*."

Specific comments:

Line 2: 'High altitude aircraft offer', aircraft→ aircrafts

The term aircraft can be equally used both in singular and plural form (see https://www.britannica.com/dictionary/aircraft).

Fig 1. Please define optical density and explain its usage in this study. Y titles of fig.1 'OD' is not expanded. Readers may be misled by optical depth. Please clarify it.

We thank the reviewer for spotting this typo. As explained in Section 2, the DOAS analysis implements the Beer's Law (in the form of logarithm of the ratios of near-limb and direct-sun spectra), where the attenuation of the light through the medium is defined by the optical depth. In Fig.1, OD is an abbreviation for optical depth and not optical density, as erroneously reported in the caption of the figure. The caption has been corrected accordingly.

Line 160: 'IWP is defined as the vertical integral of ice mass concentration through a cloud'. Why not just use ice water content instead of ice mass concentration? It seems easy to confuse it with concentration, number concentration, and number density in this paper.

We agree with the reviewer about the confusion that the term mass concentration can generate. The term ice mass concentration has been replaced with IWC.

Line 163: 'Mice' why not just IWC? It will directly relate to the retrieval of IWC from SIWP later in the paper.

As explained in the previous comment, we agree that M_{ice} is equivalent to IWC and somewhat misleading to the reader. M_{ice} has been replaced with IWC.

Line 187: add ',' between 'angle One'.

The missing dot has been added to the text.

Line 190: what does 'SIWP tracks the O2 SCD' mean?

It means that SIWP behaves similarly to the O2 SCD.

Line 192: 'This is somewhat counter-intuitive, considering ….' Why that happens?

We do not have a conclusive answer for this, but we have provided a balance between two different mechanisms as a possible explanation. This is mentioned directly after the above sentence.

Line 267: ‘FCDP measurements yielded the number density of the particles’, please define number density.

Number density here refers to the size distribution of the particles, i.e., number of particles per specific radius. The sentence has been rephrased as:
" *FCDP measurements yielded the number concentration as a function of particle size.* “

Line 272-274: ‘using the ice volume related to the averaged particle number concentration n’, are you sure you mean particle number concentration, not the mass concentration as mentioned above?

The term refers to number concentration as defined in the answer to the previous comment. Hawkeye data provided the size distribution of the particles, which has been averaged for the portion of the flight considered, in order to evaluate the total volume. Knowing the density of ice, we evaluated the nominal ice water content IWC_0.

‘This resulted in a nominal ice concentration of $Nice$ = 1.22 x 10-3 g m-3’ Obviously, the unit here refers to mass not number concentration. I think it has the same meaning as Mice in Equation (5). To get grid of confusion, please keep the symbol consistent. Again, I would suggest using IWC instead of mass concentration.

The reviewer is correct. It does have the same physical meaning. The term nominal ice mass concentration N_{ice} has been replaced with IWC_0, representing the nominal ice water content.

‘n is referred to as the nominal case’, do you mean n = $Nice$? Then n is mass concentration not mass concentration?

Yes, *n* refers to the mass concentration. In line with keeping the symbols consistent, the term *n* has been deleted to avoid confusion, and the paragraph from lines 267–271 has been changed to:

“*We therefore calculated an averaged ice particle concentration over the period of interest for this study and used this to calculate the relative total volume. Knowing the density of ice* $\rho_{ice} = 0.91 \times 10^{-6}$ *g m−3, this resulted in an ice water content of* $IWC_0 = 1.22 \times 10^{-3}$ *g* m^{-3}*. If not otherwise specified,* IWC_0 *is referred to as the nominal case in the rest of the paper. This value represents the baseline, and the different ice water content values used in this study are expressed as multiplicative factors with respect to the nominal value*.”

Equations 6 and 7: Could you explain more why it needs two wavelengths to retrieve τice, but only one wavelength to retrieve $\tau O2$?

According to Beer law's, trace gas optical depth can be retrieved knowing intensity values with and without the gas of interest. This can be easily done with two separate runs of the RT code. For ice, this is not possible, because of the large difference in magnitude of the intensity with and without ice. Beer's law can however still be applied, using two wavelengths close enough to each other (1548.1 nm and 1550.3 nm in our study) to assume a similar ice absorption but far from the main ice absorption band. These two approaches are described by Eq. (7) and Eq. (6), respectively.

Table 3: it is confusing to read.

We thank the reviewer for the comment. For a better understanding of the table content, we have rephrased the caption as:

"*The table lists the parameters used for the three different sensitivity tests (with respect to geometry, surface emissivity, and cloud altitude) performed. For example: geometry test has a fixed surface albedo (0.05) and cloud extension (14–18km), and only solar and azimuth angles are varied, according to the value in the corresponding cell. Flight altitude is set to 16.5 km for all tests*."

Line 301-306: my understanding here ice concentration refers to ice mass concentration. n = 1.22*10-3 g/m3.

The reviewer is correct. As already explained in the response to Line 160 comment (and following), the nominal value was replaced with IWC_0 in line 268.

Figure 3: for y titles, replace ice absorbance with SIWP, and oxygen absorbance with SCD, if I understand correctly. Please add units for both x and y axis. For x axis, what does ice nominal x n mean? I think n represents ice nominal case.

We thank the reviewer for this comment. The Y-axis for both the ice and O_2 tests refer to absorbance (dimensionless), as shown through the explanations in Eq.(6) and Eq.(7), and not SIWP or SCD. The purpose of this sensitivity test is only to show how ice and O_2 absorbance behave when the ice mass concentration is varied. We do state in line 302 that ice absorbance and SIWP are proportional, implying that the behavior of ice absorbance is proportionally related to the SIWP.

We agree with the reviewer's comment on the X-axis. The X-label has been changed to "*x* IWC_0 *[g* m^{-3}*]*" and the sentence "*The X-axis values represent the different ice mass concentrations, expressed as multiplicative factors with respect to the nominal value* IWC_0

(i.e., 10^0 = IWC_0, 10^{-1} = 0.1xIWC_0, …)." has been added to the caption of Fig.3 for a clearer understanding of the results.

Equations (10) and (11): the radiation being reflected by surface or atmosphere below the aircraft may transmitted through clouds. Why is the extinction of clouds ignored? Which is to say, instead of $Is(\lambda)R(\lambda)$, it may be more realistic using $Is(\lambda)R(\lambda)T(\lambda)$, T is the transmissivity.

We are aware of the limitation of not considering lower clouds in the model (as stated in line 262). However, our choice has been validated by the CPL data, which showed very little presence of lower clouds for the portion of the flight considered. We agree that Eq. (10) and Eq. (11) are simplifications. However, we would like to point out that the purpose of the equations in Section 3.1 is to provide a conceptual model to explain the mechanism of limb measurements of stratospheric cirrus clouds. The quantitative interpretation of the observations is performed using the RT model that can explicitly describe low altitude clouds.

Line 379 '$Nice$ identifies the ice particle number density' I think it is called nominal ice concentration in Line 273, which should be mass concentration, not particle number density. Again, please keep the terms and symbols consistent if you are meaning the same thing.

We corrected this point here as well, following the previous comments regarding the ambiguity of these terms.

Equation 13: I think τ=ln [$IobsIS$], based on Equation 7. Then Equation 13 misses a negative sign.

We thank the reviewer for this comment. Eq. (13) represents the generic expression for the optical depth (as stated in line 382) from which the different cases are derived, and it is expressed as an absolute value. We agree with the reviewer about the ambiguity of the sign; this has been resolved through introducing the minus symbol for the derived optical depth in Eq. (13), Eq. (15), Eq. (17), Eq. (18), and Eq. (20).

Table 4: size distribution 'bimodal 3-5 um', change as 'bimodal peak 3-5 um' might be clearer.

We thank the reviewer for the suggestion. This has been changed in the text of the caption of Table 4.

Line 446: 'the average ice particle number' is it the ice particle mass concentration as mentioned in Line 273? Note that particle number and particle mass are two different terms.

The reviewer is correct. It refers to mass concentration. The sentence has been modified in line 442 as:

"*The average ice particle mass concentration*".

Line 447- 448: 'SIWP and number concentration are proportional', be sure if you mean the number concentration or mass concentration.

The reviewer is correct again, this refers to mass concentration. This sentence has been modified in line 444 as:

"*SIWP and mass concentration*"

Line 460: 'This difference is likely due to an actual IWP higher than the modeled IWP'. This is confusing. SIWP? Ice concentrations?

The sentence has been rephrased as: "*This difference is likely due to an actual ice mass concentration higher than the modeled value."* in line 454 to clarify this point.

Line 463: 'the model favors an ice density', is it the observation favors an ice density of about half of the nominal value? I think the nominal values is used in the model.

We confirm that the nominal value is used in the model. The sentence refers to the 7:26–7:40am portion of the flight only, where the simulation with half of the ice mass concentration is more in agreement with the observed measurements.

Line 470-480: Please explain more specifically how to get IWC from SIWP? Is there any equations or references to point the readers to a better understanding of the methods?

To improve the understanding of the methodology used to retrieve IWC from SIWP the following paragraph has been added in lines 463–468:

"*Based on the knowledge gained from our sensitivity calculations, we then used a linear interpolation of the modeled SIWP to the observed SIWP, in order to determine the IWC corresponding to the mini-DOAS measurements. The IWC for every observation is thus evaluated by scaling the nominal ice water content* IWC_0*, in order to achieve the best fit between the observed and modeled SIWP. The retrieved IWC varies between 0.001 gr* m^{-3} *and 0.0025 gr* m^{-3} *and is independent of the variability imposed by RT effects on*

SIWP, these values are consistent with typical cirrus cloud ice water content levels observed in the TTL [Thornberry2017, Schiller2008]."

Line 472: 'Four different cases were chosen…' please add details (date, location, flight altitude etc.) about these four cases.

We thank the reviewer for the suggestion. The four cases were chosen during Science Flight 2 in February 2014. The following paragraph has been added in lines 471–476:

"*Four different cases were chosen to represent the variability of the retrieved IWC from the measured SIWP. The four cases were chosen within the same Science Flight 2, recorded between the 16th and the 17th of February 2014, for the same geographical location (13°N,144°E). The four cases had similar altitudes (16.6 km) but different solar zenith (57.5°, 56.5°, 25°, 30.5°) and viewing azimuth (53.5°, 138.9°, 343.8°, 138.4°) angles. IWC for these four cases range from 5.4×10^{-3} g m^{-3} to 0.02 g m^{-3}, capturing a wide range of cirrus cloud conditions*".

Line 473: 'retrieved IWCs for these four cases range from 5.4x10^-3 g/m3…', how well is this limit compared to lidar?

We chose to focus on the IWC from the in-situ instruments since a comparison of mini-DOAS with LIDAR data is challenging due to the different viewing strategies, i.e., limb vs nadir. The in-situ IWC observations are also more sensitive. We agree with the reviewer that a more formal intercomparison of the various methods would be advantageous. However, this was outside the scope of this study.

Section 4.3: the whole section discusses IWC rather than IWP.

The discussion of the interpretation of the results (related to SIWP, IWP, and IWC) starts at Section 4.2, following on to the next section. We kept IWP in the title only for consistency with the previous section. We agree with the reviewer's suggestion, and we renamed the section title in line 448 as: "*IWC retrievals*".

Figure 8, panel b, the unit of IWC 'gr m-3' → g m-3 to keep it consistent in the paper.

We thank the reviewer for spotting this typo. The label has been corrected.

Line 512: Fig. 8 shows SIWP not 'retrieved IWP'. Do you mean IWC?

"Shown" refers to the portion of the flight and not the IWP. To avoid confusion, the sentence has been modified in lines 514–515 as follows:

"*In order to evaluate the uncertainties introduced by a change in the RT input parameters to the retrieved IWC for the flight segment considered, we performed sensitivity tests for different ice cloud heights and particle size distributions*".

Lines 521-525: It is confusing how to get IWPs and their uncertainties in this section. What is the best retrieval? How do you get the percentage uncertainty?

The following paragraph has been added in lines 525–529 to provide a better understanding of the IWP uncertainties in the retrieval:

"*IWP can be obtained from the simulated SIWP, which provides IWC that is then integrated over the known cloud extension (definition of IWP). In order to get the IWPs and their uncertainties , we ran the RT model starting with the nominal case (assumed as the best retrieval), defined by a cloud deck from 14–18 km, and then modifying the extension of the cloud and the azimuth angle by fixed amounts. IWP uncertainty is then expressed as a percentage of the variation of the retrieved IWP from the nominal case, for a variation of a specific parameter in the RT model (i.e., cloud height, SZA).*"

Line 538: 'as IWP is the IWC integrated over the vertical extent' move the definition of IWP before the discussion of IWP uncertainty and explain a little bit on why discuss IWP not IWC.

As mentioned in the reply to the previous comment, ice water path IWP is first defined in Section 2.2 (line 159), and we repeated the definition here in order to strengthen the point we were making. However, we did move the IWP definition before the discussion of the IWP uncertainties, as suggested by the reviewer, adding the following sentence in lines 525–527:

"*As IWP is the IWC integrated over the vertical extent of the cloud, we discuss IWP uncertainties rather than IWC uncertainties. In addition, IWP is related more closely to the variation of the cloud extent, as modeled in this sensitivity test*."

Line 573: 'lower detection sensitivity for IWC in limb geometry'. Do you mean higher detection sensitivity? Compare to what other approaches to demonstrate a better sensitivity of your approach?

The point of this sentence was mostly to remark on the use of limb scanning near-IR measurements as a tool to infer information on the IWC and the detection of subvisible cirrus cloud. The sentence has been rephrased in lines 576–578 as follows:

"*Finally, this approach has showed the ability to detect IWC in limb geometry, and its potential use as alternative method for the detection of cirrus at low ice water concentration.*"