Comment on acp-2021-307

Anonymous Referee #1

Referee comment on "Twenty years of ground-based NDACC FTIR spectrometry at Izaña Observatory – overview and long-term comparison to other techniques" by Omaira E. García et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-307-RC1, 2021

Response to Referee#1

The authors would like to thank the Referee for reviewing carefully this paper and provide valuable and constructive comments that have improved this work. In the following text, the Referee suggestions (in blue italics) are addressed in detail (the authors' responses are in plain text).

This is an extensive and well written review of the measurements and analyses programs in place at the Izaña Observatory over the past twenty years. It is a review paper, differing from most in that the focus is on the FTIR measurements at this site rather than a broad overview of some subject matter. This is not a problem as the measurement and science record is extensive. This paper and the references therein provide a useful compendium of the research carried out at Izaña by the FTIR group and their colleagues, and it will provide a good starting point for researchers looking to understand the context of the work carried out there. It discusses some of the more significant results and provides the references to the supporting papers should a more in-depth look be of interest to the reader. The assessment of instrument performance over the 20 years is comprehensive and fairly well presented. It is difficult to judge this paper in terms of "substantial new concepts, ideas, methods, or data" as presented herein, but nevertheless I feel the paper does have merit in presenting in one place the extensive data record and the use of that data record to address many of the more prominent areas of atmospheric research.

Some general remarks:

The paper refers to the IFS120/5 HR. Is this to denote that the instrument is of the family of the IFS120HR and the later IFS125HR, or is it to denote that it entered service as an IFS120HR and was later modified to the specification of the newer IFS125HR?

The denotation of the second Izaña FTIR instrument as IFS 120/5HR is due to the interferometer belongs indeed to the family of the IFS120HR, however, before installing at Izaña observatory (IZO), it was upgraded to the IFS125HR electronics. This clarification will be included in Section 3.1 of the revised manuscript.

With some of the figures that have many data points, the use of circles and filled circles (dots) of similar or the same colour can be problematic. It would be better to use clearly different symbols. Looking specifically at Figure 6, the use of gray circles and black circles is not a good idea.

The authors agree with the Referee in that the symbols and colours used in some figures can make them difficult to evaluate. Therefore, the format of Figure 4, 6, 8, 14 and 15 will be modified, following Referee#1's comments as well as the suggestions from Referee#2 and #3.

Specific comments:

For Figure 2, could some indication of the mean number of measurements per day (binned per month) be given?

The spectral range covered by the NDACC activities (700 and 4500 cm⁻¹) is routinely measured at IZO using six optical filters in a sequential manner (so-called SA, SB, SC, SD, SE, and SF or S1, S2, S3, S4, S5, and S6). Therefore, the number of measured spectra is highly variable depending on each NDACC filter, as illustrated in Table 3 of the preprint and Figure 1 below. This figure shows the monthly distribution of all FTIR observations taken from 1999 to 2018 for the abovementioned six NDACC filters. Usually, two measurements per day are routinely taken for each NDACC filter, except for SC, for which greater frequent observations are recorded given these spectra are used for water vapour and greenhouse gas studies (Schneider et al., 2012, 2016, García et al., 2016, and references therein).

The authors agree with the Referee in that this information would be useful for the paper and will be included in Section 3.1 of the revised manuscript. However, it will not be included in Figure 2 of the preprint to avoid making the figure more confusing.



Figure 1. Monthly distribution of all FTIR measurements taken at IZO from 1999 to 2018 for the six NDACC optical filters (SF-SA). The left axis corresponds to cumulative measurements per month, while the right axis represents monthly mean of all measurements.

Line 120: Was the IFS 120/5HR operated in "vented mode" at all times, or just for the time period of the comparison with the IFS 120M?

Both FTIR instruments have always been operated in ventilated mode given the especially dry conditions at IZO. This will be make clearer in the revised manuscript.

Table 2: As formatted, this table is difficult to read. Is there a difference between the Target Gas and Gas columns? Perhaps the "Gas" should also be Target Gas. It would improve readability significantly if a few more spaces or a vertical bar were added to more clearly separate the left-hand grouping of gases from the right-hand grouping

Effectively, the term "Gas" should be "Target Gas". It will be corrected in the revised Table 2, which will be formatted following the Referee's comment.

Line 223: This would be more readable if it was written: This figure depicts the rows of the averaging kernel matrix A ..."

This statement will be modified following the Referee's suggestion.

Table 3: For readability rather than spacing the columns equally, I would suggest adding more space between the columns for the combined pairs, and removing a space between the M and σ , so the first line would look like:

C2H6 13625 1.48 0.15 1.81 0.39 5.44 0.17

Table 3 will be modified following the Referee's suggestion.

Figure 3: As there are many similar averaging kernels, it would be of interest to see some explanation of why the particular kernel was selected for display in the figure.

Figure 3 depicts, as an example, all the rows of the averaging kernel matrix (A) for all NDACC products for typical measurement conditions at IZO (spectra taken on 20^{th} July 2013 at a solar zenith angle of $\sim 30^{\circ}$). Note that, for each trace gas, the A rows describe the altitude regions that mainly contribute to the retrieved profile and, therefore, the vertical distribution of the FTIR sensitivity. Given that the retrieved altitude information is not independent (Rodgers, 2000), only those A rows at altitudes representative of the layers discernible by the FTIR instrument were highlighted in Figure 3 as coloured lines. Therefore, for each trace gas (i.e. the frame in Figure 3) the number of highlighted A rows corresponds to the number of independent layers discernible by the remote sensing instrument, which is equivalent to the trace of A or Degrees Of Freedom for Signal (DOFS), as summarised in Table 3 of the preprint. This will be further explained in Section 3.4 of the revised manuscript.

Line 236: Are "great values" to mean those values that show large statistical uncertainty?

Yes, this statement refers those values that show large statistical uncertainty. It will be re-written in the revised manuscript to avoid confusions.

Line 241: maximum instead of maximal

This will be modified following the Referee's suggestion.

Line 243: The systematic uncertainty budget is dominated by spectroscopic errors (instead of "led").

This statement will be modified following the Referee's suggestion.

Figure 4: The choice of lines and lines + circles in the same colour makes the plot difficult to evaluate for some constituents. Making the plot wider would help. Also, use just symbols for those constituents that have sufficient points and are fairly "straight" such that the lines are not necessary. H2CO and HCI are both shown in cyan with the former being a thinner line. I can't distinguish a difference in thickness in the lines in the plot.

Figure 4 will be modified, according to the Referee's comments, to make it easier to evaluate.

Line 250: Do the authors mean to say that the predominant errors are located in one of the troposphere, upper troposphere/lower stratosphere or middle/upper stratosphere, or that the errors are in all three regions? If the latter, they do not appear to be equally concentrated in the regions and that should be discussed.

The estimated uncertainty vertical profiles are largely linked to the vertical distribution of each trace gas and FTIR vertical sensitivity, as shown in Figure 4 of the preprint. Therefore, depending on each gas, they are predominantly located in only one of the layers (troposphere, upper troposphere/lower stratosphere, or middle/upper stratosphere), but not in all three regions. This statement will be clarified in the revised manuscript.

Line 257: Particularly, high error profiles ... would read better as "large" error profiles unless the authors mean to suggest that there is also an altitude dependent component which does not seem to be the case as the lines are straight in Figure 4.

Effectively, this statement should refer to large values in the estimated error profiles. It will be modified following the Referee's suggestion.

Figure 6: Caption refers to 'grey-white dots" which is, I think, a grey circle that is not filled? See earlier comment about the use of circles and dots. Also the grey circle is easily confused with a black circle. Note also that the RXCO2 quantity is used here but not yet defined in the text.

The term "grey-white dots" refers to grey circles with a white centre. The symbols and their colours will be changed in Figures 4 and 6 in order to make them clearer, following the Referee's comments.

In addition, R_{XCO2} will be defined in the caption of Figure 6.

Line 312: I don't think that 'considering' is the proper word here. I would suggest 'using'. For example: "... using a scaling retrieval with a fixed WACCM a piori VMR profile, and PROFFIT software."

This statement will be modified following the Referee's suggestion.

Line 366: At the resolution the data are plotted with, it is not possible to discern a seasonal cycle in Xair N2. If the authors consider this to be a significant finding, then overlay a trace on the data that would display it. Otherwise, the statement could be left out without weakening the section.

Effectively, the Xair N_2 seasonal cycle is not clearly discernible in Figure 6. However, to avoid make this figure more confusing, the authors prefer to keep it as is and remove this statement in the discussion.

Line 384: "... causing punctual downward and upward shift of the UTLS region,..." The use of punctual doesn't make much sense. Punctual implies arriving on schedule. Is that what the authors mean to say?

Effectively, the term "punctual" has been wrongly used here. It will be replaced by "sporadic".

Line 390/Figure 7: It would be a good idea to move the HF and N2O frames to be next to each other to make the anti-correlation of the two easier to detect.

The frames of the trace gases are displayed in alphabetical order in Figure 7. However, as the Referee suggests, placing the HF and N_2O time series together could help reader to better follow the discussion. This will be changed in the revised manuscript.

Line 559: There is a set of ellipses between CCMVal initiative and Schneider et al. Are there more missing from the list? There are 8 models listed and 10 papers referenced. It might make it clearer to list the CCM followed directly by the reference.

As far as the authors know, there are no more studies dealing with the evaluation and development support of global atmospheric chemistry climate models at Izaña observatory, thereby the ellipsis will be removed in the revised manuscript and the statement will be modified following the Referee's suggestion.

Lines 561-562: This is an awkward and confusing sentence and the details are lacking. There is mention of "attribution of sources/sinks", but no comment on whether or not there is agreement. Similarly for the "representation of moist processes" evaluation. As the paper is already quite long, it might be best to make a general statement about the comparison to the CCMs and referring the reader to the above references for details rather than to try to call out these areas without further discussion.

The authors agree with the Referee in that this explanation is vague and does not provide helpful information. Therefore, it will be removed in the revised manuscript.

Line 693 and below: Generally, when speaking of time the word used is coincident rather than collocation.

The word "collocation" will be changed by "coincident" following the Referee's suggestion.

Line 690: It is unclear whether a actual single temporal criterion is applied here as multiple gases are being discussed. Sentence should read either "Similar temporal criteria ..." or "A similar temporal criterion...." as the authors see fit.

For CH₄, CO and N₂O, the same temporal criterion has been applied for both NDACC-GAW and NDACC-TCCON comparisons (i.e. matching the daily means). This statement will be changed following the Referee's suggestion.

Line 699: Over what time interval are the FTIR observations averaged? Are the total columns derived from a series of sequential spectra, averaged?

For the DOAS-FTIR intercomparison, all available FTIR measurements taken before and after 12 UT were averaged with the only restriction of not considering FTIR data at very high solar elevation angles (\geq 85°) to avoid imprecise retrievals (mainly caused by misalignments of the solar tracker or spectroscopic issues) (see Section 3.3 of the preprint). As shown in Figure 2 below for O₃, the FTIR measurements at IZO are mostly concentrated (\geq 90% of the total observations) in the interval 8:00-16:00 UT, i.e., ±4 hours around noon. Therefore, the latter can be considered as a representative time interval for FTIR averages.

The FTIR total column means are computed from single total column retrievals, not from an averaged solar spectra.



Figure 2. Hourly distribution of the FTIR O_3 measurements taken at IZO in the period 1999-2018. The number of measurements (left axis) and the cumulative percentage (right axis) are shown.

Line 703: Need a comma after "correction"

This statement will be modified following the Referee's suggestion.

Line 706: If I understand what is being said, this might read better as "... each FTIR measurement is only paired once to the reference observation that minimizes the time difference within the temporal collocation window." And I would use coincidence rather than collocation.

This statement will be modified following the Referee's suggestion.

Figure 14: Presumably the "TRO" quantities refer to the tropospheric portions?

Effectively, the term "TRO" refers to the comparison of the tropospheric quantities (GAW in-situ records and FTIR VMR averages). This clarification will be included in the caption of Figure 14.

Line 785: In what manner are the PWV values over IZO "slowing down"?

Both climate models and observations suggest that an upward trend in water vapour is expected to appear as a response to the surface temperature increase (Chen and Liu, 2016, and references therein). Nevertheless, it has also been shown that water vapour scales not everywhere to temperature as expected and that large regional differences exist (Bernet et al., 2020, and references therein). Over continental areas, correlations between surface temperature and integrated water vapour changes are smaller than over oceans, showing in some regions even opposite trends (Wagner et al., 2006). In this sense, as mentioned above, albeit the net response of climate system points to an increase in the atmospheric water vapour concentrations, numerous studies report large trend variabilities, especially at a regional scales (e.g. Nilsson and Elgered, 2008; Alshawaf et al., 2017, Bernet et al., 2020). Another key factor largely contributing

to trend variabilities is altitude (Bernet et al., 2020). Given the less water vapour content present at higher altitudes, the trend estimation is more sensitive to uncertainties.

The FTIR water vapour observations at IZO suggest that water vapour total column amounts have been significantly slowing down over the last two decades. This finding is consistent with the downward trends obtained from coincident CIMEL and GPS measurement techniques. This progressive loss of humidity in the subtropical free troposphere might be associated with the expansion of the tropical belt, meaning that the descending limb of the Hadley cells is shifting towards the poles in both hemispheres (Heffernan, 2016, and references therein). As a consequence of this poleward movement, their associated subtropical dry zones are expected to move as well (Seidel et al., 2008). In addition, dynamical variability of atmospheric transport circulation may affect atmospheric composition of subtropical regions and influence trend assessments (Li et al., 2009; Strahan et al. 2020). Although dedicated studies would be of great use in better understanding these drivers and connections on short-term and long-term scales, they are beyond the scope of this review work.

Finally, the authors should also admit that, although the estimated water vapour trends are found to be statistically significant, the period analysed is relatively short to draw robust conclusions for a trace gas, like water vapour, with a so large spatial and temporal variability. These results will be revisited as the water vapour records at IZO extend over time.

Line 823: Do the authors mean to say that the NDACC FTIR product is able to capture "only" a part of its tropospheric variations?

The timescale analysis presented in the current work is a very powerful tool to examine what temporal signals are captured by the FTIR observations, and to what extent. For example, as illustrated in Figure 14 of the preprint, the agreement observed between ground-level GAW records and tropospheric NDACC products is mainly the result of seasonal and long-term signals. Therefore, the NDACC FTIR products are able to properly monitor the tropospheric variations at those time scales. On a daily basis, the correlation found is almost nil for long-lived gases, such as CH_4 and N_2O , likely due to their daily variations being smaller than the FTIR precision. However, for CO, the agreement obtained significantly increases, leading to the NDACC FTIR CO product also being able to capture significant information of tropospheric CO signals on a daily basis.

Line 842: What is meant by "pure free conditions"?

This statement was included to point out that the comparison between radiosonde humidity and FTIR profiles is carried out for those layers where the subtropical free tropospheric conditions are reached. As discussed in detail in Schneider et al. (2016), a fair comparison of in-situ profile measurements (free troposphere) with ground-based FTIR measurements made at IZO (on a mountain ridge) is difficult due to the local thermal circulation that starts on the island during the morning hours. To ensure that both measurement techniques are detecting air masses with the same atmospheric characteristics and representative of the free tropospheric signals, these authors found the optimal coincidences correspond to those FTIR observations taken at low solar elevation angles (between 25° and 45°). For that reason, this criterion was applied in the current work.

However, we agree with the Referee in that this short statement as it is in the preprint is vague and confusing. Therefore, it will be modified as follows: "To examine this effect and ensure the comparison is carried out for free tropospheric conditions, the FTIR observations have also been

restricted to low solar elevation angles (between 25° and 45°) (Schneider et al., 2016), with the resulting difference profile also included in Figure 15 (in red)."

Line 845: I suggest the wording be: Despite a considerable decrease in the number of coincidences, ...

This statement will be modified following the Referee's suggestion.

Line 849: I suggest the wording: ...which makes the comparison of the remote sensing and in-situ profiles difficult.

This statement will be modified following the Referee's suggestion.

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