Comment on acp-2021-307

Anonymous Referee #3

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-RC2, 2021

Response to Referee#3

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).

"Twenty years of ground-based NDACC FTIR spectrometry at Izana Observatory - overview and long-term comparison to other techniques" Omaria Garcia et al., 2021

The paper meticulously describes the methodology of FTIR retrievals and draws on previous work in the field. The writing style is clear and descriptive. It gives a very detailed description of the 20-years FTIR-related measurements from 120/5 HR at Izana. Such kind of overview paper is valuable for the global users (modelers, satellite developers, atmospheric scientists...) to use their data. Izana is located in the subtropical region, which is crucial to understand the change of atmospheric compositions. The 20-years FTIR measurements have already been used in many scientific studies, leading to more than 100 peer-reviewed papers. Overall, I recommend this paper to publish in ACP, and I only have a few minor comments:

P8 line 182: WACCM model used in NDACC-IRWG is v4 instead of v6

As far as the authors know, the NDACC-IRWG guideline recommends the versions 5 or 6 of the climatological WACCM model, depending on the target gas, to be used as a priori information (IRWG, 2014). In this respect, the authors would appreciate it if the Referee could provide us another reference supporting the usage of WACCM version 4.

P8 line 188: why only use the temperature and pressure at 12:00 UT? How about the H2O? The temperature and H2O variation can be very large even on one day. Would you like to address such uncertainty on your retrievals?

As shown in Figure 1 below for O_3, the FTIR measurements at Izaña observatory (IZO) are mostly taken around noon. In particular, more than 90% of the total observations during the 1999-2018 are concentrated in the interval 8:00-16:00 UT, i.e., ±4 hours around the NCEP temperature and pressure profiles used as reference in the NDACC FTIR retrievals (12 UT). Therefore, the 12 UT NCEP profiles can be considered a reliable proxy of the atmospheric state at IZO for the radiative transfer calculations. Nevertheless, as the Referee suggests, greater frequent NCEP profiles might improve the overall quality of FTIR retrievals, and it will be taken into consideration in the next re-evaluation of the NDACC IZO database that is expected to be carried out in 2021/2022. In this sense, a previous work analysing the effect of the intra-day variability of the pressure and temperature profiles (3-hourly profiles) on different FTIR products at IZO found that the differences among FTIR products did not show significant dependence on the altitude (±0.5%), except for H_2O, for which the differences are overall within ±2% (García et al., 2014a). These values are overall within the estimated theoretical uncertainty of the FTIR products.

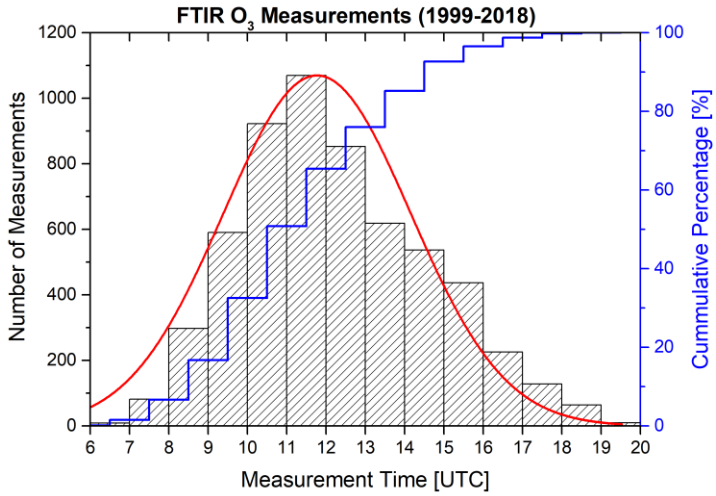

Figure 1. Hourly distribution of the FTIR O_3 measurements taken at IZO in the period 1999-2018. The number of measurements (left axis) and the cumulative percentage (right axis) are shown.

Regarding water vapour, its treatment in the operational NDACC FTIR retrievals strongly depends on its interference on the NDACC target gas and, therefore, it varies from gas to gas. For example, the NDACC CH_4 products are retrieved following Sepúlveda et at. (2012, 2014) that proposes six micro-windows, which contain strong, not saturated, and well-isolated CH_4 absorption lines as well as H_2O lines, in order to better account for the H_2O interferences. The H_2O profiles are simultaneously retrieved with CH_4 using a dedicated profiling retrieval. As documented by Hase et al. (2011), the Sepúlveda approach could be less dependent on humidity conditions as it minimises perturbing H_2O absorptions and it handles the problematic interference species H_2O/HDO in a rigorous manner. For other gases for which the H_2O interference shows a minor impact, such as N_2O, the H_2O profile is simultaneously scaled from the WACCM climatological profiles during the inversion procedure. Finally, for other gases, if possible, the spectral regions used for retrievals are selected so that H_2O absorption lines are not presented like for O_3 (García et al., 2021).

P9 table 2. do you want to add N2 also here?

Given that Table 2 only lists those FTIR products available at the NDACC archive, the authors prefer not to include N_2 in this table and keep it as an auxiliary product to test the long-term performance of FTIR observations in Section 4.

P14 line 261: "the total column-averaged amount of dry air (Xair) " is not appropriate. Xair is the ratio of o2 or n2 derived dry air to DPC, please use a better definition here.

Effectively, as stated by the Referee, the Xair parameter is computed as the ratio of the total column of O_2 or N_2 to DPC. Nevertheless, the authors have adopted the definition of Xair widely used within the TCCON community, in which Xair is defined as the column-averaged amount of dry air (e.g. Wunch et al., 2011, 2015; Pollard et al., 2017; Frey et al., 2019). The authors would appreciate it if the Referee could provide us another valid definition or reference for Xair parameter.

P22 Figure 8. in the bottom panel, are you sure the colors are correct? because the CO is increasing, but you mention that in P23 line 464 that the CO is decreasing.

Effectively, the coloured line assigned to the CO time series is wrong in Figure 8. The CO and OCS lines were exchanged. The figure will be corrected accordingly in the revised manuscript.

P22 Figure 8 in the middle panel and Figure 11, I see that the CH4 and N2O long-term trends are similar. However, other in-situ measurements show that N2O is increasing continuously while the annual growth of CH4 is variable: 1999-2007 stable, and reincreased after 2007 (https://gml.noaa.gov/ccgg/trends_ch4/). Any explanation here? Why we get a different CH4 tend from Izana FTIR CH4 measurements, especially between 1999 and 2007, compared to other surface measurements?

Figure 2 below shows the time series of the NDACC FTIR CH_4 products (total columns and tropospheric VMR means) along with the corresponding annual anomalies relative to the 1999-2018 background. Note that the anomalies are the same as those displayed in Figure 8 and Figure 11 of the preprint. The GAW ground-level CH_4 records have also been included for a better comparison with the FTIR tropospheric observations. These plots show more clearly the variable CH_4 annual growth remarked by the Referee: constant until about 2005-2007 and increasing after 2005-2007. As reported in the preprint, the NDACC IZO CH_4 data confirms a speed-up in the emission rates for CH_4 in the last decade, which is likely caused by the increase in anthropogenic emissions. This acceleration is found to be of +0.13±0.03%yr^{-1} and +0.43±0.03%yr^{-1} for the periods 1999-2008 and 2009-2019, respectively. In addition, as documented in Figure 2 below and in the detailed comparison results included in the current work (Section 8.3), there is excellent agreement between GAW and NDACC FTIR tropospheric data (excluding the well-known bias of about 2% in the FTIR products). In fact, as further illustrated in Section 8.4 of the preprint, the uneven sampling of the FTIR system results in a negligible effect on CH_4 trend estimations.

These findings are in total agreement with the results presented by Bader et al. (2017), who evaluated changes of CH_4 total columns since 2005 using FTIR observations carried out at ten globally-distributed NDACC sites (IZO among them). Particularly for IZO, they documented a close to statistical agreement with a mean annual increase of 0.33±0.01 and 0.28±0.02 % $year^{-1}$ for the NDACC FTIR total columns and GAW ground-level records, respectively, for the period 2005-2014. These values were close to those reported considering the ten NDACC sites, 0.31±0.03 % $year^{-1}$, and the GEOS-Chem CH_4 simulations included in Bader's work as well.

Nevertheless, the authors should admit that the change of FTIR instrument in 2005 at IZO could slightly affect the change point (2005-2007) between the well-known steady and increasing evolution of CH_4. As illustrated in the bottom panel of Figure 2, there is a small jump in 2005 preceded and followed by a flat period between 1999-2005 and 2005-2007, respectively. Although this potential instrumental issue does not influence the trend estimations presented in the current work, as mentioned above, it will be analysed in detail in future studies.

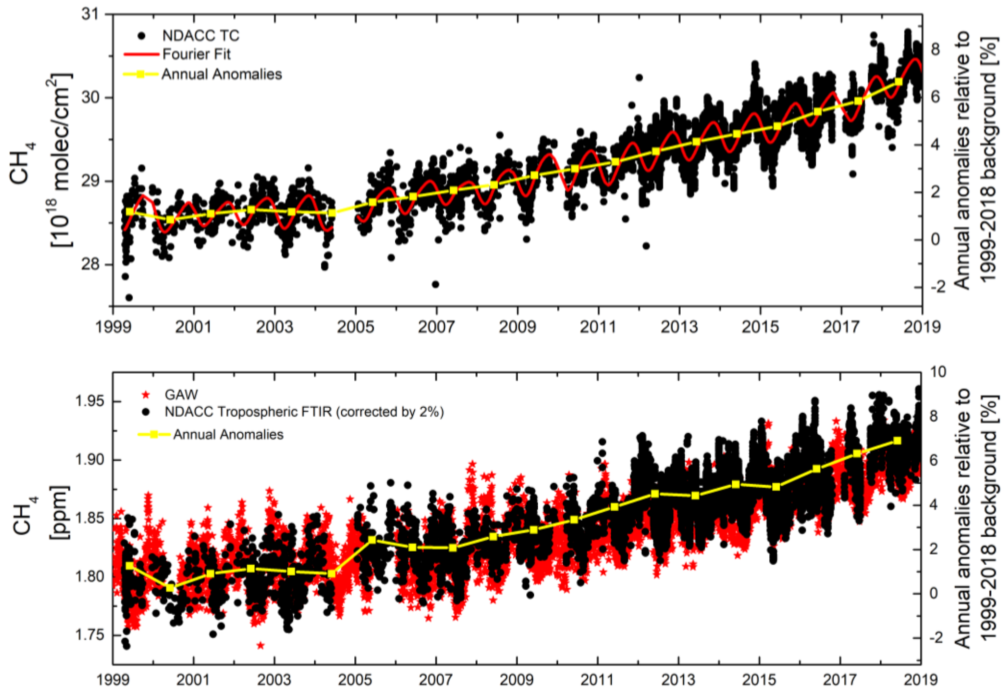

Figure 2. Upper panel: time series of NDACC FTIR CH_4 total columns (left axis) and of annual anomalies relative to the 1999-2018 background (right axis). Bottom panel: time series of NDACC FTIR tropospheric CH_4 VMR and GAW ground-level records (left axis) and of annual anomalies relative to the 1999-2018 background (right axis).

P39 line 811, the tropospheric XCH4 is compared to the surface measurements to found a bias of ~2.6%. I do not support such direct comparison, as they are sampling different vertical ranges still.

Effectively, as stated by the Referee, when comparing different measurement techniques, there are multiple factors affecting the comparison, namely, approaches, sampled air masses, observing geometries, spectral ranges, spectral resolution, retrieval strategies, vertical sensitivity, etc. All these aspects can introduce significant differences and must be considered when interpreting the comparison results. This was briefly discussed in the current work in Section 8.2.

The authors assume that the Referee is referring to the comparison between the NDACC tropospheric CH_4 product and GAW ground-level records, albeit the Referee mentioned XCH_4. The latter is the term used to refer to the total column-averaged CH_4 mole fraction.

The NDACC tropospheric FTIR and ground-level observations can indeed be representative of different air masses (i.e. different vertical ranges). However, the comparison methodology used in the current work was designed to minimise these potential impacts. On the one hand, the NDACC tropospheric concentrations are obtained as the mean of retrieved VMR profiles between IZO altitude (2.37 km a.s.l.) and middle troposphere (5.6 km a.s.l.). This layer has proved to represent well the tropospheric signal detectable by the FTIR system (Sepúlveda et al., 2012, 2014; García et al., 2014b). On the other hand, the daily nighttime means (20.00-08.00 UT) of the IZO ground-level records and the daily daytime means of the FTIR products are paired. As mentioned in the preprint, given the strategic location of IZO, diurnal insolation generates a slight up-slope flow of air originating from below the inversion layer that can disturb the free troposphere conditions at IZO. However, during nighttime, the subsidence regime typical for subtropical regions prevails and the atmospheric observations taken at IZO are well-representative of the subtropical North Atlantic free troposphere (Cuevas et al., 2019, and references therein). Under these conditions, the ground-level records are well comparable to the FTIR observations in the lower troposphere (Sepúlveda et al., 2012; 2014; García et al., 2014b, 2018). In particular, for CH_4, the bias of 2.6% of the NDACC FTIR products related to GAW ground-level data found at IZO is consistent with those reported at different NDACC FTIR stations ranging different latitudes and altitudes (Sepúlveda et al., 2014). Therefore, the authors presume that it is likely due to spectroscopic parameter issues in the mid-infrared spectral region, not being introduced by the comparison approach.

P39 line 814, the reference "Zhou et al., 2019" is wrong. It should be https://amt.copernicus.org/articles/12/5979/2019/

This reference will be changed following the Referee's comment.

References

Bader, W., Bovy, B., Conway, S., Strong, K., Smale, D., Turner, A. J., Blumenstock, T., Boone, C., Collaud Coen, M., Coulon, A., Garcia, O., Griffith, D. W. T., Hase, F., Hausmann, P., Jones, N., Krummel, P., Murata, I., Morino, I., Nakajima, H., O'Doherty, S., Paton-Walsh, C., Robinson, J., Sandrin, R., Schneider, M., Servais, C., Sussmann, R., and Mahieu, E.: The recent increase of atmospheric methane from 10 years of ground-based NDACC FTIR observations since 2005, Atmos. Chem. Phys., 17, 2255–2277, https://doi.org/10.5194/acp-17-2255-2017, 2017.

Cuevas, E., Milford, C., Bustos, J. J., R., García, O. E., García, R. D., Gómez-Peláez, A. J., Guirado-Fuentes, C., Marrero, C., Prats, N., Ramos, R., Redondas, A., Reyes, E., Rivas-Soriano, P. P., Rodríguez, S., Romero-Campos, P. M., Torres, C. J., Schneider, M., Yela, M., 1005 Belmonte, J., del Campo-Hernández, R., Almansa, F., Barreto, A., López-Solano, C., Basart, S., Terradellas, E., Werner, E., Afonso, S., Bayo, C., Berjón, A., Carreño, V., Castro, N. J., Chinea, N., Cruz, A. M., Damas, M., De Ory-Ajamil, F., García, M., Gómez-Trueba, V., Hernández, C., Hernández, Y., Hernández-Cruz, B., León-Luís, S. F., López-Fernández, R., López-Solano, J., Parra, F., Rodríguez, E., Rodríguez-Valido, M., Sálamo, C., Sanromá, E., Santana, D., Santo Tomás, F., Sepúlveda, E., and Sosa, E.: Izaña Atmospheric Research Center Activity Report 2017-2018, Eds. Cuevas, E., Milford, C. and Tarasova, O., State Meteorological Agency (AEMET), Madrid, Spain, 1010 and World Meteorological Organization (WMO), Geneva, Switzerland, WMO/GAW Report No. 247, 2019.

Frey, M., Sha, M. K., Hase, F., Kiel, M., Blumenstock, T., Harig, R., Surawicz, G., Deutscher, N. M., Shiomi, K., Franklin, J. E., Bösch, H., Chen, J., Grutter, M., Ohyama, H., Sun, Y., Butz, A., Mengistu Tsidu, G., Ene, D., Wunch, D., Cao, Z., Garcia, O., Ra1035 monet, M., Vogel, F., and Orphal, J.: Building the COllaborative Carbon Column Observing Network (COCCON): long-term stability and ensemble performance of the EM27/SUN Fourier transform spectrometer, Atmospheric Measurement Techniques, 12, 1513–1530, https://doi.org/10.5194/amt-12-1513-2019, 2019.

García, O.E., M. Schneider, F. Hase, T. Blumenstock, E. Sepúlveda, E. Cuevas, A. Redondas, A. Gómez-Peláez and J.J. Bustos, Effect of updates in LINEFTIT and PROFFIT at Izaña ground-based FTIR: improvement of the ILS characterisation and intra-day pressure and temperature variability, NDACC-IRWG/TCCON meeting 2014, 12-16 May, Bad Sulza (Germany), 2014a.

García, O. E., Schneider, M., Hase, F., Blumenstock, T., Sepúlveda, E., Gómez-Peláez, A., Barthlott, S., Dohe, S., González, Y., Meinhardt, F., and Steinbacher, M.: Monitoring of N2O by ground-based FTIR: optimisation of retrieval strategies and comparison to GAW insitu observations, in: NDACC-IRWG/TCCON meeting 2014, Bad Sulza, Germany, 2014b.

García, O. E., Schneider, M., Ertl, B., Sepúlveda, E., Borger, C., Diekmann, C., Wiegele, A., Hase, F., Barthlott, S., Blumenstock, T., Raffalski, U., Gómez-Peláez, A., Steinbacher, M., Ries, L., and de Frutos, A. M.: The MUSICA IASI CH4 and N2O products and their comparison to HIPPO, GAW and NDACC FTIR references, Atmospheric Measurement Techniques, 11, 4171–4215, https://doi.org/10.5194/amt11-4171-2018, 2018.

García, O. E., Sanromá, E., Schneider, M., Hase, F., León-Luis, S. F., Blumenstock, T., Sepúlveda, E., Redondas, A., Carreño, V., Torres, C., and Prats, N.: Improved ozone monitoring by ground-based FTIR spectrometry, Atmos. Meas. Tech. Discuss., https://doi.org/10.5194/amt-2021-67, in review, 2021.

Hase, F., Blumenstock, T., Schneider, M., and Sepúlveda, E. Interactive comment on “Strategy for high-accuracy-and-precision retrieval of atmospheric methane from the mid-infrared FTIR network” by R. Sussmann et al., Atmos. Meas. Tech. Discuss., 4, C1048–C1048, 2011.

IRWG: Infrared Working Group Uniform Retrieval Parameter Summary, Tech. rep., http://www.acom.ucar.edu/irwg/IRWG_Uniform_RP_Summary-3.pdf, 2014.

Pollard, D. F., Sherlock, V., Robinson, J., Deutscher, N. M., Connor, B., and Shiona, H.: The Total Carbon Column Observing Network site description for Lauder, New Zealand, Earth Syst. Sci. Data, 9, 977–992, https://doi.org/10.5194/essd-9-977-2017, 2017.

Sepúlveda, E., Schneider, M., Hase, F., García, O. E., Gómez-Peláez, A., Dohe, S., Blumenstock, T., and Guerra, J. C.: Long-term validation of tropospheric column-averaged CH4 mole fractions obtained by mid-infrared ground-based FTIR spectrometry, Atmospheric Measurement Techniques, 5, 1425–1441, https://doi.org/10.5194/amt-5-1425-2012, 2012.

Sepúlveda, E., Schneider, M., Hase, F., Barthlott, S., Dubravica, D., García, O. E., Gómez-Peláez, A., González, Y., Guerra, J. C., Gisi, M., Kohlhepp, R., Dohe, S., Blumenstock, T., Strong, K., Weaver, D., Palm, M., Sadeghi, A., Deutscher, N. M., Warneke, T., Notholt, J., Jones, N., Griffith, D. W. T., Smale, D., Brailsford, G. W., Robinson, J., Meinhardt, F., Steinbacher, M., Aalto, T., and Worthy, D.: Tropospheric CH4 signals as observed by NDACC FTIR at globally distributed sites and comparison to GAW surface in situ measurements, Atmospheric Measurement Techniques, 7, 2337–2360, https://doi.org/10.5194/amt-7-2337-2014, 2014.

Wunch, D., Toon, G. C., Blavier, J.-F. L., Washenfelder, R. A., Notholt, J., Connor, B. J., Griffith, D. W. T., Sherlock, V., and Wennberg, 1300 P. O.: The total carbon column observing network, Philosophical Transactions of the Royal Society - Series A: Mathematical, Physical and Engineering Sciences, https://doi.org/doi:10.1098/rsta.2010.0240, 2011.

Wunch, D., Toon, G. C., Sherlock, V., Deutscher, N. M., Liu, X., Feist, D. G., and Wennberg, P. O.: The Total Carbon Column Observing Network's GGG2014 Data Version, https://doi.org/10.14291/TCCON.GGG2014.DOCUMENTATION.R0, 2015.