Reference: https://doi.org/10.5194/gi-2024-14

“The Harwell TCCON observatory”

Damien Weidmann, Richard Brownsword, Stamatia Doniki

Responses to Reviewers’ Comments

The authors would like to thank the reviewers for their detailed, thorough and constructive comments. The manuscript has been revised with most of their suggestions, and we think it has been improved thanks to the reviewers’ comments.

Further down this document, the reviewers’ comments are stated in blue font, and our respective answers in black font. Please note that all lines appearing in the answer sections refer to the revised manuscript.

Additionally, a manuscript file with tracked changes reflecting the modifications will be provided.

Reviewer 1 – David Griffith

This well-written paper provides a detailed description of the Harwell TCCON site and its operation since its inception in ~2020. TCCON sites are encouraged to publish such a detailed description, in particular to demonstrate their compliance with the standard operational procedures (both hardware and software) set out by TCCON. This paper is valuable reading for any TCCON practitioner (I learned a few things) and is well suited to publication in this journal. I recommend acceptance for publication after addressing several general and specific comments listed below.

General comments:

The technical descriptions are comprehensive with one glaring omission – there is no description of the path from fitted level 1 spectra to level 2 column averaged mole fractions (Xgas). This should be added, at least in summary, for completeness and readability of the paper, even if it is described elsewhere. The essential steps to describe are:

1. Retrieval of total column amounts of each gas from spectra, including the profile scaling approach (as distinct from profile retrieval). The is already partly covered.
2. Definition and calculation of total column dry air mole fractions as the ratio of total column of gas to O2 times O2 mole fraction, corrected for water vapour
3. Definition of XLuft and why it is useful

This description belongs in section 5.1.

We have addressed this comment by editing paragraphs 2, 3, and 4 of Section 5.1. The reviewer’s three comments above are now reflected in paragraphs 2-4 of the revised manuscript. We have clarified the profile scaling approach, we have added the derivation of Xgas, and added a paragraph defining Xluft and its value.

The full section is reproduced below. We also supplied a marked change manuscript that shows more clearly the changes made.

> Within the GGG software, a non-linear least-squared fitting of the level 1 data with an atmospheric transmission model is iteratively performed, by scaling an a-priori vertical profile of volume mixing ratios (VMRs) of atmospheric gases, until convergence between calculated and measured spectra is achieved and the residual is minimal, as seen in Fig. 9. The profiles are only scaled, so the vertical distribution of the a priori profile is preserved. The output of this fitting consists of total column abundances V_{Gas} of each gas in molecules.cm^{-2}, being the vertical integral over each profile. The a priori gas VMR, pressure and temperature profiles originate from the near real time GEOS meteorological dataset referred as FP-IT (Forward Processing for Instrument Team), switched to GEOS-IT since the 1st of April 2024. For the TCCON, Caltech produces the extrapolated dataset relevant to each site, made available with a timeliness of 24 to 48 hours (Laughner 2023).
>
> The processor then uses correction factors for each retrieved species: one to account for mostly spectroscopy errors producing a bias on column abundances dependent on the solar zenith angle (air mass dependent correction factor ADCF) (Wunch 2011), and one which is a global scaling (air mass independent scaling factor - AICF) designed to anchor the retrieved X_{Gas} product to WMO-traceable in-situ measurements (Wunch 2010). Currently, in the absence of an independent site scale-factor determination, the Harwell site processor uses the values that have been updated for the GGG2020 software release and are given in (Laughner 2023a). We are planning to attempt a site-specific update to the correction factors (Pollard 2021) using the travelling standard methodology (Herkommer 2024). The level 2 output data produced are column-averaged DMF of gases, X_{Gas}, defined by Eq. 3, where f_{O2} is the known mean DMF of O_2 in the well-mixed atmosphere. The calculation of X_{Gas} by reference to the column abundance of O_2 reduces the sensitivity to errors in Sun pointing and surface pressure measurements, among other advantages, as outlined in (Wunch 2011a) and (Laughner 2023a).
>
> $$X_{Gas} = \frac{V_{Gas}}{V_{O_2}} \cdot f_{O_2} \quad (3)$$
>
> A diagnostic quantity, X_{Luft}, is also calculated. It represents the ratio of two distinct means of calculating the air DMF: one from surface pressure, and the other from the O_2 column retrieval (Laughner 2023a). X_{Luft} should ideally be unity, with deviations of even a few per mil being indicative of retrieval bias.

Mole fraction vs VMR:

The quantities Xgas such as XCO2 are mole fractions, not volume mixing ratios – they are not the same thing:

Mole fraction(A) = (moles(A)/total moles in mixture)

Volume mixing ratio(A) = volume(A)/(total volume – volume(A))

For TCCON purposes and in in situ measurements we wish to work with mole fractions. VMR introduces two errors:

1. For trace gases at the ppm level vmr and mole fraction may be insignificantly different (eg 0.04% for CO2 in air) but for O2 the difference is large: mole fraction = 0.21, VMR = .21/.79 = 0.26.
2. VMR assumes an ideal gas and that volume is proportional to amount (moles)

Unfortunately, incorrect use of VMR is entrenched in some TCCON literature and more widely, but its use should be discouraged. For example using VMR(O2) rather than mol frac (O2) in calculating Xgas (point 2 above) would lead to a >20% error in Xgas! It is not used in metrology literature, which also prefers "amount fraction" to mole fraction".

My recommendation is to change all usage of VMR to mole fraction throughout the paper, and at first use point out that although historical terminology often refers to VMR, it is actually mole fractions that are calculated and used.

Indeed, this is a point of confusion, even wider than the TCCON literature as the VMR is often defined as a molar ratio without stating the restricted validity domain of the approximation, and the two terms have been used interchangeably, including by us. We agree with the reviewer clarification which remove any ambiguity and have revised the manuscript accordingly:

L11-12: *dry volume mixing ratio (VMR)* replaced by *dry mole fractions (DMF)*

L203: *the retrieved gas VMRs* replaced by *the retrieved gas DMF*

L346: *some other gas column VMR* replaced by *some other gas column DMF*

L372: *for averaged-column dry VMR of* CO_2 replaced by *for averaged-column DMF of* CO_2

L381: *comprising column-averaged dry volume mixing ratios of* replaced by *comprising column-averaged DMF of*

Note that we have retained the term volume mixing ratio (VMR) when referring to the a-priori vertical profiles, which are provided as such.

Technical corrections

L4: The TCCON geometry is not solar occultation, it is (direct) solar absorption. Occultation refers to something like an eclipse, or the earth's atmosphere moving directly between the observer (eg satellite or high altitude balloon) and the sun during sunrise/sunset.

Agreed. Following the reviewer's suggestion, and to ensure consistency with prior TCCON documentation (eg. Laughner et al., 2024), we have corrected the statement as follows:

… high-resolution spectroscopy of the atmosphere's transmission in direct-sun viewing geometry, following the TCCON methodology.

L10: First use of VMR – see general comment above

Please see reply to second general comment regarding the use of VMR and mole fraction, listing all subsequent changes.

L13: “zenith solar occultation mode” => direct solar absorption mode”.

As explained previously in L4, the manuscript has been revised as:

... *in the direct-solar absorption mode*.

L15: it would be good to reference here the early TCCON papers led by Debra Wunch (2011, 2010) which provided the first description of the TCCON (2011) and its “calibration” (2010).

Agreed. References have been added. The manuscript now reads:

L16

... *and associated sciences (Wunch et al., 2010, 2011a). TCCON has become essential for satellite data product calibration and validation (Wunch2011b),...*

Table 1: If possible please provide a reference (eg wiki page) for these criteria and requirements. Eg from the wiki

https://tccon-wiki.caltech.edu/pub/Main/TCCONCharter/ETL_TCCON_application.pdf

This is not a public page, maybe it could be referenced and copied to an appendix (without the ETL responses)

We agree with the reviewer that a reference is needed. The TCCON wiki page with the main requirements is publicly accessible, therefore we have updated the manuscript as follows:

L53

... *and framed by the TCCON measurement system requirements (TCCON wiki, 2021)*.

And added the full citation as:

TCCON wiki: TCCON Requirements,

https://tccon-wiki.caltech.edu/Main/TCCONRequirements, last accessed: 2025-02-12, 2021.

Table 1: “ME variation over OPD” is not clear – perhaps substitute “ME variation from 1.0”

Agreed. We have adopted the proposed correction for Table 1:

ME variation from 1.0

Table 1: Lineshape analysis “In beam” => “HCl cell in beam”

Agreed, though the HCl cell refers to the lineshape monitoring, as the analysis is done by the LINEFIT software, therefore Table 1 is now updated as:

Lineshape monitoring In-beam HCl cell

L57: “input FTS aperture (field stop)”

Proposed change adopted, as:

L61

… onto the input FTS aperture (field stop).

L57: This input accepts an f/6.5 beam (full angle of view 8.7 deg)

Proposed changed was adopted as:

L62

This input accepts an f /6.5 beam (full angle of view of 8.7◦)

L59: the exit aperture of the interferometer (replace “beamsplitter”)

Proposed changed was adopted as:

L63

The exit aperture of the interferometer compartment…

L61: one side of the dichroic is in nm and the other in cm-1. For FTIR, better to use cm-1, replace 625-825 nm with 12000-16000 cm-1 (or quote both nm and cm-1) in both places

For consistency with the FTIR community, we have changed the units to wavenumber. The manuscript now writes:

L65

… between 12000 to 16000 cm^{-1}

L66: an InGaAs detector (not “a”)

Correction adopted as:

L70

… onto an InGaAs detector

L76: Much but not all of the “development” was done at NIWA. Perhaps a clearer sentence would be “A high precision alt-azimuth solar tracker was built in house based on the design of Robinson et al (2020) with some adaptions described below to capture…”

Suggestion adopted. Manuscript now reads:

L82

A high-precision alt-azimuth solar tracker was built in-house based on the design of (Robinson et al., 2020), with some adaptations described below, to capture the radiation input to be directed into the FTS.

L78: Specify the manufacturer (Physik Instrumente) for the rotators. “PI” is not clear

Indeed, a possible point of confusion. The manuscript has been revised as follows:

L84

(Physik Instrumente part No PRS200 6449921111)

L86-87

(Physik Instrumente part No L-611.9ASD)

L94 “magnification of 0.25” Is this correct? Is the image of the 1 mm input aperture just 0.25 mm on the camera sensor?

Yes, the statements in the original manuscript are correct as they stand.

L98: remove “occultation”

As mentioned previously, we have now adopted the term “direct sun” or “solar”. The manuscript now reads:

L103

… start direct solar measurements …

L102: NI => National Instruments

Adopted.

L107

National Instruments Vision library

L111: pure HCl at ~ 5 hPa pressure

Manuscript corrected as:

L116

… with pure HCl at 4.865 hPa pressure

L114: Is your HCl cell really 25mm diameter? Every one I've seen is 50 mm.

Yes, the original manuscript is correct as it stands.

L183: O2 A-band (760 nm, 13000 cm-1)

Corrected to add the band in wavenumber. The manuscript now reads:

L188

... of the O2 A-band (760 nm, 13000 cm^{-1}).

L185: remove "occultation"

Removed and revised as:

L190

... direct solar measurements

L200: suggest rephrase as ...the requirement of ME = 1.0 +/- 0.05 or 5% over the range of OPD

Suggestion adopted. We have also rephrased the sentence for greater clarity, which now reads:

L205

The variation of spectrometer modulation efficiency (ME) is consistently well within the requirement 0.95 < ME < 1.05 over the full range of OPD, as seen in Fig. 5 which shows the extreme values of ME picked up from each of the ME vs OPD output analyses.

L265: Debra Wunch has very recently prepared a detailed paper on GGG2020 vs GGG2014 that has just been published on the Caltech server, it would be useful to reference that alongside Laughner 2023b

The Total Carbon Column Observing Network's GGG2020 Data Version: Data Quality, Comparison with GGG2014, and Future Outlook (GGG2020.R0). CaltechDATA. https://doi.org/10.14291/TCCON.GGG2020.DOCUMENTATION.R0

Accepted and added as:

L270-271

... GGG software (Laughner et al., 2024; Wunch et al., 2025)

L271 and following: VMR => mole fraction

Accepted. Please refer to the answer of the second general comment.

L279: No definition of Xgas etc, see general comments.

Accepted. Addressed in our response to 'General comments'.

L281: best to avoid "calibration" as it does not have the same meaning as commonly used in the in situ community (and has raised ire…) Suggest replace with " in the absence of independent site scale factor determination."

Proposed correction adopted. The manuscript now reads:

L287-288

… *in the absence of independent site scale-factor determination* …

L294: This ZPD location error has now been corrected using a different, simpler approach and will be implemented in I2S in the next GGG2020 version.

Proposed correction adopted. The manuscript has been updated as:

L312-314

As more TCCON facilities encountered the same issue, the TCCON partners agreed on a unified approach for the correction to be part of the main algorithm, to be implemented in the next GGG2020 version (Griffith and Laughner, 2024).

And the following reference was added:

Griffith, D. and Laughner, J.: Bug #349: I2S fix for HR125 firmware bug, TCCON software development platform, https://gggbugs.gps.caltech.edu/, only accessible to the TCCON partners, Last accessed: 2025-02-13, 2024.

L296 it is not "incompatible with I2S", it is "incorrect". The GGG2020.next fix identifies and corrects it in I2S. (see GGGBUGS)

Corrected as:

L310

… *introduces an incorrect value to I2S* …

L314, table 3: Please provide the Lindqvist equation that is L-M-fitted, it would be much easier to follow the meanings of the fitted parameters in Table 3.

Accepted. Equation (4) added as L331.

L329: Explain Xluft, it is not obvious to the reader. It is important that the 0.2095 factor for O2 is described as a mole fraction not a VMR (see general comments).

We have addressed this in the third paragraph of text added in Section 5.1 as our response to ‘General comments’.

L373: In acknowledging the TCCON community, it would also be appropriate to mention the QAQC teams that especially provide a lot of time and feedback to all sites.

Absolutely, this is a clear oversight from us. The authors would like to apologise for the omission. The acknowledgement part has been updated as:

L394-396

Furthermore, the quality of the TCCON data is heavily dependent on reviewers who assess site data submission as part of the TCCON quality assurance and control (QA/QC) process. We acknowledge the effort, advice, and feedback from the QA/QC teams, particularly David Griffith, Debra Wunch and Thorsten Warneke, who regularly review the Harwell site data submission.

Reviewer 2 – Anonymous

"The Harwell TCCON Observatory" by Weidmann, Brownsword and Doniki, seeks to describe the implementation of a TCCON site at the Rutherford Appleton Laboratory in the UK and demonstrate that the data is of high quality and compliant with TCCON (Total Carbon Column Observing Network) protocols and then goes on to describe a few of the interesting and notable features of the dataset.

Site and dataset descriptions such as this provide an invaluable resource to the user community as well as those seeking to start their own measurement system or even to diagnose issues with existing systems, and their creation and publication should be encouraged.

The manuscript achieves nearly all its objectives in a thorough, technically competent and well written way, with a couple of minor exceptions described in the general comments below. Once these have been addressed, publication of this manuscript would constitute a valuable contribution to the literature.

General comments:

The introduction of the column averaged dry mole fractions of trace gases (denoted X_{gas}), which are the major outputs of the TCCON processing system, is vague to the point of being non-existent. In particular the fact that these quantities are the result of scaling by the co-retrieved oxygen column means that various instrumental/systematic errors are removed. The introduction of the scaling to produce the X_{gas} quantities would also lead naturally to the definition of the X_{luft} diagnostic, which is mentioned in the manuscript but not described. The correct units for X_{gas} should be mole fractions, e.g. nano or micro moles per mole, although ppb and ppm respectively seem to be used interchangeably in the literature.

This point concurs to one of the points of the Reviewer 1. We have addressed this comment by editing paragraphs 2, 3, and 4 of Section 5.1. The reviewer's three comments above are now reflected in paragraphs 2-4 of the revised manuscript. We have clarified the profile scaling approach, we have added the derivation of Xgas, and added a paragraph defining Xluft and its value.

We supplied a marked change manuscript that shows more clearly the changes made to address this point.

Throughout the manuscript the measurement principle is described a solar occultation. This is not correct; occultation refers to the process of the source becoming hidden or eclipsed over time, whereas these measurements are made nearly instantaneously whilst the source (the sun) is fully visible.

We acknowledge the usage of the incorrect term and have replaced all mentions of "occultation" with "direct-sun viewing geometry" or similar, which keeps the article in line with Laughner et al. (2024). The changes can be identified in the following lines of the revised manuscript: L4, L13, L103, and L190.

Specific comments:

Abstract: It would be nice to have more of the meta-data included in the abstract, such as the latitude and longitude of the site. As the data set is fundamentally associated with this paper it would be justified to include the full DOI either here or prominently in the introduction.

The facility coordinates have been added in L1 as follows:

The Harwell observatory, located in Oxfordshire, UK (51.57◦ N, 1.315◦ W), now part …

A hyperlink to the DOI for the Harwell TCCON data has been added in L8.

L9. At the first mention of the TCCON would be a logical place to cite Wunch et. al. 2011.

Agreed. References have been added as follows:

L10

The Total Carbon Column Observatory Network (TCCON) (Wunch et al., 2011b; Laughner et al., 2024) is an …

L10. (and subsequent mentions) column averaged dry mole fraction (or ratio). This is also a good opportunity to introduce the X_{gas} notation, even if it is defined later.

We have adopted the use of the correct unambiguous term "mole fraction" and have revised all instances in the manuscript as:

L11-12: *dry volume mixing ratio (VMR)* was replaced by *dry mole fraction*

L346: *some other gas column VMR* was replaced by *some other gas column mole fractions*

L354: *for averaged-column dry VMR of CO_2* was replaced by *for averaged-column dry mole fractions of CO_2*

L381: *comprising column-averaged dry volume mixing ratios of* was replaced by *comprising column-averaged dry mole fractions of*

L18. References for CO2M and MicroCarb.

Agreed. We have now updated the document with a reference for each mission, to the best of our knowledge. The manuscript now writes:

L19-20

… such as EUMETSAT CO2M (Sierk et al., 2019; Courrèges-Lacoste et al.,2024) and CNES MicroCarb (Bardoux et al., 2019; Cansot et al., 2023)) …

The references section has been updated accordingly.

L25. TCCON has already been introduced and so the acronym could be used here.

Agreed, the manuscript has been updates as:

L27

… in compliance to the TCCON requirements a …

L31. It is mentioned later in the manuscript, but the dates of the change of location would be useful here.

We have adopted the proposed changed. The manuscript now writes:

L33

… by 40 metres (from December 2021 to February 2022) about 15 month after starting operation (September 2020)..

L39. The description of the wind rose implies that this is only selected data. If this is so, it should be made explicit here, and in the caption of Fig. 1.

The origin of the data for the wind rose is already stated in L42: "The diurnal wind rose derived from the Harwell site meteorological station recorded between 14/09/2022 and 14/09/2023 is shown in an inset of Figure 1.". We have updated the caption of Figure 1 to read "The insert in the bottom left gives the diurnal wind rose from the site wind measurements for September 2022 to September 2023".

Fig. 1. The reproduction of this (and several other figures) is small and difficult to read, a larger font would be helpful.

We have updated the figures to increase its size by taking advantage of the full available page width. The fonts in Fig. 1 have also been enlarged, and the wind rose key parameters are now given in the caption.

The only figure that remains the same is Figure 4, which is there to give a sense of the different applications used for automating the solar measurements; it is a result of various screenshots compilation, and its resolution can only be as good as a basic screen resolution.

L50. Although reproduced fully here, the TCCON requirements are also documented on the TCCON wiki, a reference and last accessed date here would be informative.

We agree for the need of a reference. The TCCON wiki page with the main requirements is publicly accessible, therefore we have updated the manuscript as follows:

L53

… and framed by the TCCON measurement system requirements (TCCON wiki, 2021).

And added the full citation as:

TCCON wiki: TCCON Requirements,

https://tccon-wiki.caltech.edu/Main/TCCONRequirements, last accessed: 2025-02-12, 2021.

L61. Inconsistent use of wavelength and wave number.

For consistency with the FTIR community, we have changed the units to wavenumber. The manuscript now writes:

L65

... *between 12000 to 16000 cm^{-1}*

L73. Are solar measurements DC?

Indeed, they are. To avoid future misunderstandings, we have updated the manuscript as follows:

L75

For solar measurements, the detectors are operated in DC mode, unit gain is applied in a...

L73. (or Table 1) define HCl at first use.

We have now added the HCl definition in the caption of Table 1.

L78 and 81. It can be assumed that the parenthesised codes are part numbers, but this should be made more explicit.

Indeed, they are part numbers. The manuscript has been revised as follows:

L84

(Physik Instrumente part No. PRS200 6449921111)

L86-87

(Physik Instrumente part No. L-611.9ASD)

L110. The acronym OAPM has previously been defined, one acronym should be used consistently.

Agreed. We have removed the OAPM acronym in L67 and revised the manuscript as:

... *length off-axis paraboloid (OAP) mirror.*

It is now consistent with all other instances in the document.

L113. Define CaF_2

Definition has been now added:

L118

... 40 mm focal length calcium fluoride (CaF_2) lens.

L124. Enumerate the second item.

Enumeration has been corrected, as:

L129-130

... a rain gauge (Gill Instruments Maximet GMX240), 2) a sun pyranometer (Gill Instruments Maximet GMX101) ...

L154. It would be more normal to specify rainfall as a rate, mm hr^{-1}, or simply state "no rainfall".

The use of the term "rainfall" was inaccurate. The instrument measures precipitation, which includes snow, hail and others, and is measured in units of $mm.hr^{-1}$. Therefore, we have updated the manuscript as:

L159

Precipitation must be < 0.08 $mm.hr^{-1}$ (the sensor resolution).

Fig. 5. Sub figure labels are missing.

We have now updated the figure and added the missing labels.

L231. Remove second "to" or consider modifying the sentence to make it more understandable.

A typing mistake. We have now corrected it and revised the manuscript as:

L235-236

The non-linear distortion of the detector output relative to the photon flux can either be due to detector saturation, resulting in a ...

L313. "the associated modelled seasonal variation." Might be clearer.

Accepted. The sentence now reads:

L329

Figure 10 shows the X_{CO2} temporal series, together with a fit of the seasonal variation model developed by (Lindqvist et al., 2015) and given in Eq. 4

L318. Including a description of the model as an equation would help the reader.

Equation added as L331.