

The point-by-point overview to the referees comments taken from the author response in the manuscript discussion. Notes on what exactly was changed have been added. The page and line numbers refer to the attached LaTeX-Diff file.

Point-by-point response to Anonymous Review #1

General Comment:

One potential source of systematic bias is due to the fact that the solar spectrum observed on the moon is a solar disc-averaged spectrum, while TCCON observes disc-centered spectra (and this is assumed in the GFIT analysis also, therefore the lunar spectra need to be processed with different settings).

Response:

Sunlight reflected at the lunar surface will have a (solar-)disc-averaged spectrum, i.e. the solar lines will be broadened as a result of the different Doppler-shifted contributions from different parts of the solar disc. GFIT includes a setting, that switches to a calculation of a disc-averaged spectrum, when the moon is selected as the source, therefore no bias is expected. This will be clarified in the revised manuscript.

Changes:

Paragraph added on page 4 line 33-36.

General Comment:

A discussion of the two crucial items (accuracy budget and of the target accuracy and precision) should be discussed in the final version of the paper.

Response:

The referee is correct that the accuracy and precision of the lunar retrievals are crucial. Within the frame of this study the solar TCCON measurements are considered to be correct. Section 4 of the manuscript shows the validation with the TCCON data. Daytime TCCON data has been compared with the nighttime lunar measurements and it is assumed that diurnal variation can be neglected. We assume this is valid given that the model outputs for that time period show small variabilities in the order of 0.2 ppm (1.0 ppb) for xCO₂ (xCH₄) (see Tab. 2). The accuracy of the lunar measurements can be determined via the bias of the lunar compared to the solar measurements and can be deduced from Tab. 4 as well. In March 2013 the difference between solar and lunar measurements is 0.66 ± 4.56 ppm for xCO₂ and -1.94 ± 20.63 ppb for xCH₄. In the September 2013 campaign a bias of 1.01 ± 8.52 ppm for xCO₂ and -3.36 ± 41.13 ppb for xCH₄ can be observed. The diurnal variability of the lunar measurements is used to define the precision. As the later measurements have a higher precision, a typical value achieved in the 2014/2015 winter is used. This is given in the Conclusions as a standard deviation of the daily mean of 2 ppm (10 ppb) for xCO₂ (xCH₄), in both cases corresponding to about 0.5 %. This discussion will be added to Section 4 in the final manuscript and the values for the bias added to the abstract to further emphasize their importance.

The target accuracy on the other hand is more difficult to determine. As suggested by the reviewer, the detrended year-to-year wintertime variability in the models can be used as a proxy. In the smoothed, detrended MACC CO₂ and CH₄ model the arithmetic mean of the first week of January differs by 0.55 ppm in xCO₂ and 9.84 ppb in xCH₄ between 2012 and 2014. At the same time, the standard deviation of all values for the first week of January between 2012 and 2014 is about 1.8 ppm for xCO₂ and 18.8 ppb for xCH₄. However, these estimates are potentially subject to unknown biases in the model, i.e. the model could be biased similarly every year. Additionally, the seasonal variability surely is an upper limit for the target precision. Here the seasonal cycle amplitude measured by solar FTS is about 15 ppm for xCO₂ and about 40 ppb for xCH₄.

This will also be added in the revised manuscript.

Changes:

An extended discussion of the accuracy budget has been added on page 14 lines 4-10. A discussion of target accuracy added to pg. 14 lines 11-17.

Comment:

Abstract: also provide an estimate for the accuracy (bias with respect to solar TCCON measurements) of the lunar measurements.

Response:

An estimate of the accuracy as addressed in the answer to the previous comment will be added to the abstract.

Changes:

The abstract has been extended to include the estimates on page 1 lines 7-9.

Comment:

Page 2, line 28: "The extension of the bandgap . . . reduces the quantum efficiency" - is this true?

Response:

No, it should have stated "the extension of the detector sensitivity" and will be corrected in the revised manuscript. The manufacturer reports values for the noise equivalent power (NEP), that is the power required to achieve a signal-to-noise ratio of 1, for the 1.7 μm cut-off model of $1.8\text{E-}15 \frac{\text{W}}{\sqrt{\text{Hz}}}$ and $9.0\text{E-}15 \frac{\text{W}}{\sqrt{\text{Hz}}}$ for the 1.9 μm cut-off model respectively. For an uncooled diode with 2.6 μm cut-off, the reported NEP is $2.1\text{E-}12 \frac{\text{W}}{\sqrt{\text{Hz}}}$ (see http://www.teledynejudson.com/prods/Documents/InGaAs_shortform_Sept2003.pdf).

Changes:

The statement has been corrected on page 2 line 31 and an extended explanation added to page 3 in lines 4-5.

Comment:

Figure 1: It would be instructive to show a lamp spectrum recorded with the standard TCCON detector element also (and to provide some information concerning the noise level achieved with the selected lunar InGaAs diode (cooled and uncooled) and with the standard extended TCCON detector element for the same input signal level, e.g. for the 6000 . . . 6400 cm^{-1} region, where the CH_4 and CO_2 bands reside).

Response:

As mentioned above, the sensitivities of the standard TCCON diode and the TE cooled differ by about 3 orders of magnitude. Therefore, given the vast difference, we do not feel that adding a graphical representation of this is necessary.

Changes:

No changes to the manuscript.

Comment:

Page 4, line 10 ff: not a sentence.

Response:

Sentence will be reworded to: The differences between the solar and lunar measurements include the detector, the spectral resolution, the integration time and the size of the entrance aperture.

Changes:

Changed on page 4 line 22.

Comment:

Section 3.2: The fact that the noise level is too high in the lunar observations for using the spectroscopically observed oxygen column should be regarded as a severe drawback of the suggested approach.

Response:

This is a misunderstanding. The DMFs used in this study use the O₂-ratio approach, the noise here is not 'too high' but merely higher than using the surface pressure approach to calculate the DMFs. However, the error cancellation properties of the O₂-ratio approach outweigh the potential

lower noise achieved by using the surface pressure. We have attempted to clarify this in the paper to avoid confusion.

Changes:

No changes to the manuscript.

Comment:

Page 5, line 11: “. . .for the analysis in section 4.” It would be instructive to explain here to the reader which topic is covered in Section 4.

Response:

The sentence will be changed to: In the following, the approach described in equation 1 was used to retrieve xCO₂ and xCH₄. The second approach, in equation 2, was only used to retrieve xO₂ in Section 4, which covers the validation with solar measurements.

Changes:

Extension of the respective sentence on page 5 line 26-27.

Comment:

Page 7, line 10 ff: “. . .one option to decrease measurement time . . . is to increase the velocity . . .”. No, this is not the case in the context of optimizing the spectral signal-to-noise-ratio (SNR). Here, only the spectral resolution and the throughput matter. I would have expected (for a given allowed integration time) to see a more pronounced reduction of error bars on the retrieved columns until a further reduction of resolution starts to decrease the contrast between the lines and the adjacent continuum. Has the spectral SNR been adjusted as function of resolution in this manner (assumption of a certain amount of available integration time)? When comparing different resolutions, one might also take into account that a larger fieldstop can be applied when resolution is reduced (increasing the signal level, favoring shorter scans even more).

Response:

While the increase in velocity does increase the number of scans possible in the same time frame, this has no effect on the spectral S/N. The wording will be adjusted to clarify. Assuming the second comment refers to Figure 6, no, the data set has not been adjusted for the possible number of spectra within a certain time frame. The aim was to understand what impact the spectral noise has as a function of resolution on the retrieval. A figure highlighting the increase in signal-to-noise ratio (SNR) is included below in Figure 1 and will be integrated in the revised manuscript together with the following explanation: Here the increase in SNR was measured as a function of spectral resolution with a Bruker 125 HR, normalized to the SNR at 0.02 cm⁻¹, i.e. a spectrum recorded with 1.0 cm⁻¹ resolution has a 10 times larger SNR (see blue line). Additionally, the shorter scan length allows to record more spectra in the same time frame. Averaging leads to an increase in SNR by a \sqrt{N} with N measurements (red line). The resulting black line shows the potential increase in SNR with resolution for a fixed integration time. At lower resolutions the size of the entrance aperture is limited by the size of the image of the lunar disk, rather than the resolution. Figure and discussion will be added to Section 3.4 of the final manuscript.

Changes:

Figure 6 was added and the paragraph page 8 line 21 - page 9 line 6 included.

Comment:

Page 8, line 4: “. . .white noise were added.”. How has this operation been performed technically? In the interferogram domain before the FFT? Note that this section does not specify (nor treats) the choice of the numerical apodization function, which seems a further important choice in addition to the scan length if reaching the best possible precision of the retrieved column is so crucial.

Response:

Here, the interferogram was cut to obtain lower spectral resolution and then Fourier-transformed. Then the noise was added to the spectrum. In all retrievals, from solar and lunar spectra, a boxcar apodization function was applied. Note that the retrieval adjusts for the resulting sinc-shaped distortion of the spectral lines in the spectral domain. Using a different apodization function would result in information loss in the spectrum. The usage of the Boxcar apodization is mentioned in

the revised manuscript.

Changes:

Explanation added on page 4 line 17-18.

Comment:

Page 12, line 10: “air-glow emissions”. This study would be especially interesting if lunar spectra taken during twilight would be treated separately (spelling: airglow).

Response:

Unfortunately, all lunar spectra recorded at a time when the sun is higher than -5° elevation have to be filtered out (see sec. 2.3). In twilight, backscatter in the atmosphere leads to a light path through the atmosphere that is not well-defined. This leads to higher retrieved DMFs, this behaviour is only partly compensated by the O₂-ratio approach.

Changes:

Spelling corrected on page 14 line 19. No further changes to the manuscript.

Comment:

Figure 12: Despite the fact that no biases were discovered in the September 2013 measurements, one is left with the impression that the lunar CH₄ measurements in 2015 are biased high in comparison to the solar observations.

Response:

Yes, we do not see any mechanism that would explain the apparent bias between solar and lunar xCH₄ measurements in 2015 and 2016.

Changes:

Added brief discussion on biases on page 18 line 2 - page 19 line 6. See also the corresponding answer to review #2.

Response to review #2 by Debra Wunch

Comment:

The language needs tightening - some technical concepts that are specific to TCCON or Bruker 125HR instruments that may not be familiar to the wide AMT audience are glossed over and should be written in a clearer, more general way.

Response:

The final manuscript will be revised with emphasis on readability. We have tried to define TCCON and instrumental specific references to be more approachable by a general AMT audience.

Changes:

See various changes corresponding to other review comments.

Comment:

Night time validation with aircraft or AirCore profiles would be best, but appear to be unavailable (at least, they are not mentioned in the manuscript). Perhaps this should be mentioned in the discussion or conclusions section.

Response:

Correct, so far no aircraft campaigns above Ny-Ålesund are available. Aircore measurements are difficult. Ny-Ålesund is a coastal town surrounded by mountains and glaciers and the retrieval of the probe has to be ensured. One obvious solution to this is to deploy a guided descent, but as far as we know a secure retrieval glider is still under development. This will be mentioned in the Conclusions of the revised manuscript.

Changes:

Added to paragraph on page 19 line 12.

Comment:

In Figure 14, you compare the XCH₄ seasonal cycle from your lunar and solar measurements to the MACC model. It shows significant disagreement in summer, but not in winter, showing that the model isn't able to properly reproduce the Arctic methane seasonal cycle amplitude. Do you have any idea why? This, to me, is one of the most interesting figures/results of the paper.

Response:

This is indeed very interesting, and something we hope to examine further. It appears that there is a general bias between the model and the solar FTS measurements with specific events in spring, where the FTS measurements show sudden decreases of the xCH₄. Our current understanding is, that the model is not capable of addressing vertical transport very well. Specifically stratospheric intrusions during the breakdown of the polar vortex in spring lead to large, short-term decreases in xCH₄. This is currently being investigated by using a stratospheric species as a tracer to separate the xCH₄ column in a tropospheric and stratospheric part but exceeds the scope of this paper, however we will add the above explanation to Section 5 of the final manuscript.

Changes:

Paragraph added to page 18 line 2 to page 19 line 6.

Comment:

P1L4: The moon isn't a light source - it's reflected sunlight off the moon.

Response:

Yes, in the NIR, reflected sunlight is the main component of the lunar irradiance. This will be reworded for clarity.

Changes:

Wording changed on page 1 line 4.

Comment:

P1L5: I don't think you mean "parallel".

Response:

Yes, the measurements are not actually 'parallel', but happen on consecutive days and nights. Wording has been adjusted.

Changes:

Wording adjusted on page 1 line 6.

Comment:

P1L23: You don't need extended InGaAs detectors to measure above 5000 cm⁻¹.

Response:

Correct, reworded for clarity.

Changes:

Changed wavenumber range on page 4 line 2.

Comment:

P2L18: Do you use the solar brightness fluctuation corrections for high cirrus typically employed by TCCON (embedded in I2S for DC-recorded interferograms)?

Response:

Yes, however the effect of the correction is minimal, because in case of lunar spectra there is not enough signal with strong cirrus present. Additionally, due to the low resolution of the spectra, thin cirrus clouds typically lead to brightness fluctuations between consecutive scans and less to fluctuations within one interferogram record. This will be added to the description of the postprocessing of the spectra in Section 3.1.

Changes:

Explanation added to page 4 lines 19-21.

Comment:

P2L22: 0.04 what units? mrad?

Response:

Here: 0.04 radians. The units have been added.

Changes:

Units added on page 2 line 24.

Comment:

P2L22: This sentence may be too technical for this audience. Explain that this ME and phase error are consistent with a well aligned instrument.

Response:

The fact that these values are indicative of a well-aligned instrument has been included in the revised manuscript.

Changes:

Added to page 2 lines 25-26.

Comment:

L25-35: This is too technical - please explain further.

Response:

Assuming this comment refers to section 2.2, this will be reworded, see also the answer to the comments in review #1.

Changes:

Clarification added to page 3 lines 4-6.

Comment:

P4L11: Rework sentence beginning with "Generally speaking, . . ."

Response:

The sentence has been reworded to: Decreasing the resolution leads to a shorter measurement time and therefore allows for integration of more interferograms in the same time frame. Increasing the entrance aperture allows for more incident light on the detector which increases the signal-to-noise-ratio.

Changes:

Changed paragraph on page 4 lines 23-26.

Comment:

P4L14: The entrance aperture wasn't always 3.15 mm? Please explain.

Response:

At full moon, the entrance aperture could be set to 3.15 mm. If the moon is not full, its image on the aperture wheel requires a smaller aperture to still ensure that the aperture is uniformly lit. Additionally, the four-quadrant diode used in the tracking system, sometimes has difficulties centering the non-full lunar image, using a smaller aperture in this case, again ensures full illumination of the entrance aperture. The respective paragraph in Section 3.1 has been reworded to clarify this.

Changes:

Paragraph changed on page 4 lines 27-32.

Comment:

P7L1: Please note that the large deviations are at very high SZA that would be filtered out in a typical TCCON filter. Could you make this plot for days with lower SZAs? Does it look the same?

Response:

This has been only done for the Ny-Ålesund site, here lower SZAs are only possible in summer

and due to the midnight sun conditions, the differences between the day and night atmospheric models are smaller. However this approach can easily be adapted to other TCCON sites. It will be noted in Section 3.3 of the revised manuscript that higher SZA are generally filtered out in standard TCCON.

Changes:

Sentence added to page 6 lines 18-19.

Comment:

P7L11: This worry no longer holds, given that Bruker has provided two solutions to the ghost problem (the laser sampling board potentiometer and the new M16 controllers with the XSM option), and TCCON provides a ghost removal procedure with I2S, as long as you measure simultaneously on another detector with a spectral range that is entirely within a single alias. In fact, I believe Bruker recommends 20kHz as their preferred scanner speed.

Response:

Yes, the paragraph will be adjusted. See also the answer to review #1 regarding this issue.

Changes:

Sentence changed on page 8 lines 1-5. And further explanation added corresponding to answer to review #1 on page 8 lines 21 to page 9 line 6.

Comment:

Fig 5: I see what you're trying to do with this figure, but I find it very difficult to read and interpret quantitatively. Perhaps you also need to show example slices through the 3D figures showing XCH₄ vs resolution and XCH₄ vs SNR.

Response:

Figure 5 was intended to present the qualitative behaviour of different SNRs as a function of resolution. The quantitative information, e.g. xCH₄ vs. resolution - for two extrem cases of SNR - is shown in Fig. 6. An additional plot will be added as described in the answer to review #1 showing the improvement of the S/N with decreasing resolution.

Changes:

Figure 6 and explanation on page 9 lines 1-6 have been added, as discussed in corresponding answer to review #1.

Comment:

Fig 6: Would the x-axis scale work better as a log₁₀ scale? Also, with the low SNR error bars as large as they are, it's difficult to see what the mean value is as a function of resolution. Perhaps you need to reduce the y-axis limits and show a representative error bar.

Response:

The errorbars have been removed and representative errors added to the caption.

Changes:

Estimates added to page 9 lines 8-9 and further explanation to page 10 lines 2-6.

Comment:

P10L10: The averaging kernel also depends on the retrieval methodology.

Response:

Yes, this detail has been included.

Changes:

Included on page 11 lines 9-11.

Comment:

P12L1: Can you assume that the total columns do not change significantly during the 24-hour period? What about drawdown from the terrestrial biosphere throughout the day and respiration at night? Is night time respiration a feature of the carbon cycle you can hope to measure with your

lunar measurements given the precision of your measurements? The y-axis scale is too large in Figure 9 to see whether there is any diurnal cycle in your data and models. Ditto for Figure 10.

Response:

The standard deviation of all models are in the order of 0.2 - 0.3 ppm for xCO₂ and 1.0 - 1.6 ppb for xCH₄, which is an argument for the stability of the columns during the validation time period. Unfortunately the errors are too large to investigate night-time to day-time differences, e.g. due to respiration and carbon uptake, with the lunar observation presented here. The y-axis scales cannot easily be adjusted without losing information on the lunar data points. However corresponding values for mean and standard deviation are given in Table 2.

Changes:

The axes in the corresponding Figure 10 and 11 have been rescaled (page 13).

Comment:

P14L25: Remove the comma after “both”.

Response:

Done.

Changes:

Comma removed.

Comment:

P14L25: The models don’t capture the secular trends in XCO₂ and XCH₄? Why not?

Response:

This is a misunderstanding. The models do capture the secular trends. In order to directly compare one year with another, the time series has to be detrended. This will be rephrased in the final version.

Changes:

Sentences on page 16 lines 20-21 have been reworded.

Comment:

Fig 13, 14: I don’t see any green dots.

Response:

The color reference will be updated.

Changes:

Color reference updated in Fig. 14 & 15 and on page 16 line 35.

A markup of the changes made to the previously submitted manuscript is attached.

The arctic seasonal cycle of total column CO₂ and CH₄ from ground-based solar and lunar FTIR absorption spectrometry

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Abstract. Solar absorption spectroscopy in the near infrared has been performed in Ny-Ålesund (78.9°N, 11.9°E) since 2002; however, due to the high latitude of the site, the sun is below the horizon from October to March (Polar Night) and no solar absorption measurements are possible. Here we present a novel method of retrieving the total column dry-air mole fractions (DMF) of CO₂ and CH₄ using ~~the moon as a light source~~ [moonlight](#) in winter. Measurements have been taken during the Polar Nights from 2012 to 2016 and are validated with TCCON (Total Carbon Column Observing Network) measurements by ~~parallel~~ solar and lunar absorption measurements [on consecutive days and nights](#) during spring and autumn. The complete seasonal cycle of the DMFs of CO₂ and CH₄ is presented and a precision of up to 0.5 % is achieved. [A comparison of solar and lunar measurements on consecutive days during day and night in March 2013 yields non-significant biases of \$0.66 \pm 4.56\$ ppm for xCO₂ and \$-1.94 \pm 20.63\$ ppb for xCH₄.](#) Additionally a model comparison has been performed with data from various reanalysis models.

1 Introduction

Since 1992 a Fourier-Transform Infrared (FTIR) Spectrometer in Ny-Ålesund (78.9°N, 11.9°E) has been used for the ground-based observation of total column trace gas abundances in the Arctic via solar absorption spectroscopy (Notholt and Schrems, 1994). The measurements are taken within the Infrared Working Group (IRWG) of the Network for the Detection of Atmospheric Composition Change (NDACC). Since 2002, measurements in the near infrared (NIR) spectral region have been performed to retrieve the dry-air mole fractions (DMFs) of CO₂ and CH₄ (denoted here as xCO₂ and xCH₄) and other gases (Warneke et al., 2005, 2006). These are, since 2005, part of the Total Carbon Column Observing Network (TCCON). Today, these measurements are widely used as validation for satellite products, in model comparisons and studies of sources and sinks.

A large limitation of the availability of these measurements is the absence of sunlight in the polar winter. At Ny-Ålesund, between October and March, the Sun is permanently below the horizon. However, during this period the moon is permanently above the horizon around full moon.

Moonlight has already successfully been used as a light source in retrievals of various trace gas concentrations via the FTIR spectrometer (FTS) in Ny-Ålesund in the middle infrared spectral region (Notholt et al., 1993, 1997; Notholt and Lehmann,

2003; Palm et al., 2010) and in Antarctica (Wood et al., 2004). Here the employment of liquid nitrogen cooled InSb and MCT detectors ensures low instrumental noise, even under low light conditions. In the NIR, i.e. $> 5000\text{ cm}^{-1}$ $> 4000\text{ cm}^{-1}$, typically extended range InGaAs diodes are used. Recently Fu et al. (2014) and Wong et al. (2015) showed the application of a thermo-electrically cooled InGaAs detector for the measurement of reflected sunlight spectra from the Los Angeles basin on a mountaintop site. The thermo-electrical (TE) cooling reduces the detector noise and allows for higher signal-to-noise ratios in the measured spectrum.

After initial tests at the Bremen TCCON site (Buschmann et al., 2015), a TE cooled InGaAs diode detector was implemented in the Ny-Ålesund FTS and a time series of $x\text{CO}_2$ and $x\text{CH}_4$, the total column dry air mole fraction, was obtained from spectra measured during polar night between 2012 and 2016. The resulting product is compared to TCCON solar measurements as well as model simulations from the MACC reanalysis model for CO_2 (v14r2 MACCCO2 (2016)) and for CH_4 (v10 MACCCH4 (2016)), the Jena CO_2 inversion CarboScope s04_v3.7 (JenaCO2, 2005) and the Carbontracker 2015 model (CT2015, 2016). Together with the summer TCCON data from Ny-Ålesund, for the first time the whole seasonal cycle of $x\text{CO}_2$ and $x\text{CH}_4$ is presented.

In Sections 2 and 3, this paper describes the measurement setup and the methods used to retrieve the dry air mole fractions. Section 4 describes the newly obtained time series and the comparison to TCCON. Finally we compare our results with model data in Section 5.

2 Setup

2.1 Measurement site

The instrument, a Bruker IFS 120-5HR, is located at the AWIPEV research station in Ny-Ålesund (78.92°N, 11.92°E). Measurements are taken under cloud-free conditions for both the NDACC and TCCON networks during summer, and lunar absorption and atmospheric emission measurements are performed in winter. In 2014/2015 the measurement setup was gradually changed to a semi-automated system. The new system is able to automatically start a set of measurements without the need of an operator, which considerably increased the number of measured spectra. The performance of the instrument is monitored by reference cell measurements on a monthly basis and it is ensured that the phase error is smaller than $\pm 0.04 \pm 0.04\text{ rad}$ and the modulation efficiency is $\pm 2\%$ of 1.0 up to a maximum optical path difference of 180 cm. These values are indicative of a well-aligned instrument.

2.2 Thermoelectrically cooled InGaAs diode

The sensitivity of the extended InGaAs diode used as a detector in standard TCCON near-infrared measurements is too small to obtain a sufficient signal-to-noise ratio from lunar irradiance. The introduction of a two-stage Peltier element cooling system attached to the back of the diode can reduce the dark current noise and thereby minimise overall detector noise. Generally the extension of the band-gap in the diode production process detectors spectral sensitivity range reduces the quantum efficiency.

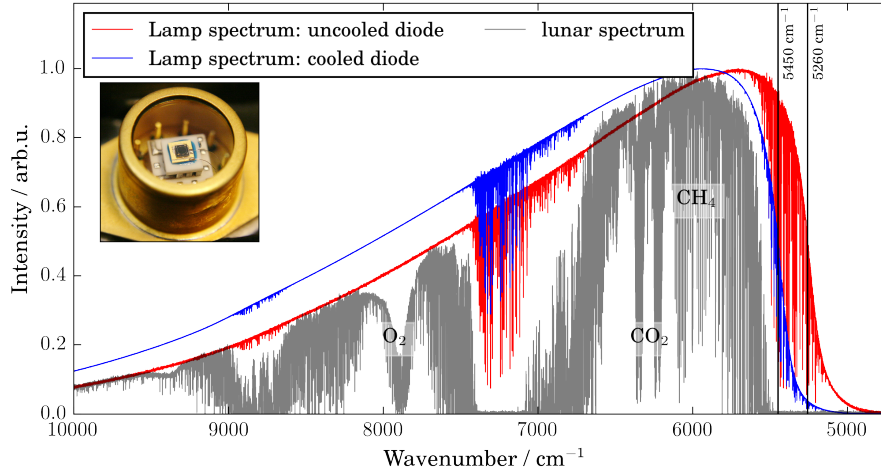


Figure 1. Example measurements of the InGaAs diode: Cooled (blue) and uncooled (red) lamp spectra. Note the indication of the cut-on wavenumbers. An averaged lunar spectrum is shown in gray and a picture of the diode was added.

Therefore, a non-extended diode improves the signal-to-noise ratio; however, cooling the InGaAs diode affects its crystal structure and therefore widens the band-gap, which leads to a shift of the diode's sensitivity range. The commercially available diode used here has a cut-on frequency of about 5260 cm^{-1} in the uncooled and about 5450 cm^{-1} in the cooled state. The noise equivalent power (NEP) of the cooled diode, i.e. the power of the incident light to achieve a signal-to-noise ratio of 1, is about 3 orders of magnitude smaller than that of a standard TCCON diode. The shift in sensitivity due to cooling, an example of an averaged lunar spectrum, and a picture of the diode are shown in Fig. 1.

2.3 Availability of moonlight

The total number of potential lunar measurement hours can be calculated by excluding all times where the lunar elevation is below the terrain height. Additionally, lunar phases with insufficient illumination (lunar phase $< 85\%$) and times where the solar zenith angle is smaller than 95° have to be excluded. Depending on lunar orbital parameters, the maximum number of measurement hours ranges from about 886 h in 2012 to 634 h in 2016. This is much less than the potential yearly solar measurement time of 3883 h. The minimum lunar zenith angle is 57.13° (2012) and 60.84° (2016) compared to a minimum solar zenith angle of 55.47° . The actual possible time available for near-infrared measurements, of course, further depends on clear sky conditions and other scheduled FTS experiments. The number of measurements was increased by switching to a semi-automated measurement setup that required less operator intervention in autumn 2015, as described above.

3 Method

3.1 Measurement setup

5 The measurements follow the TCCON standard settings wherever possible. A solar (lunar) tracker is mounted on the roof of the AWIPEV observatory and the light is reflected into the laboratory underneath and into the FTIR spectrometer. Accurate tracking is ensured by usage of a four-quadrant diode with feedback to the solar tracker motor controller. The incident light is focused on an entrance aperture and afterwards parallelised to enter a Michelson interferometer arrangement of the Bruker IFS 120-5 HR. The movable retro-reflective mirror is mounted on a sledge on steel rods. Accurate tracking of the movable
10 mirror's position is provided by a stabilized internal HeNe laser reference. The light path arrives in the detector compartment of the instrument, where it is focused through a HeNe laser filter onto the InGaAs detector. The resulting signal is amplified and recorded together with the internal laser reference.

In a post processing step the spectra are calculated via a Fast Fourier Transform (FFT) routine by the instrument operating software OPUS (by Bruker). After changing the measurement routine in 2015 to a semi-automated setup, less intervention
15 from the operator is required. At the same time, the interferograms are read directly from the instrument, resulting in raw data slices that are processed to spectra via the i2s program shipped with the GGG2014 software suite used within TCCON.

~~Differences to the solar, TCCON measurements and post-processing setup are: the detector itself~~ All interferograms have been transformed using Boxcar apodization and the retrieval code adjusts for the resulting sinc-shaped distortion of the spectral lines. Using i2s, the DC interferograms have been corrected for brightness fluctuations. However, the effect of the correction is
20 expected to be minimal; because of the low resolution, thin cirrus clouds for example typically lead to brightness fluctuations between consecutive scans and to a lesser degree to fluctuations within one interferogram record.

The differences between the solar and lunar measurements include the detector, the spectral resolution, the integration time and the size of the entrance aperture. ~~Generally speaking, decreasing the resolution~~ Decreasing the resolution leads to a shorter measurement time and therefore allows for integration of more interferograms in the same time frame ~~and the larger.~~ Increasing
25 the entrance aperture , the more light is incident allows for more incident light on the detector , i.e. the signal which increases the signal-to-noise-ratio.. The impact of spectral resolution is further discussed in section 3.4. ~~The~~

At full moon, the entrance aperture was set as large as possible (typically a diameter of to 3.15 mm), given full illumination to ensure a uniform light path in the instrument. The actual setting depends on lunar phase, as a smaller lunar cross-section requires a smaller aperture. Occasionally, a smaller entrance aperture is required, because if the moon is not full, its image on
30 the aperture wheel requires a smaller aperture to still ensure that the aperture is uniformly lit. Additionally, the four-quadrant diode used in the tracking system sometimes has difficulty centering the non-full lunar image, using a smaller aperture in this case, again, ensures full illumination of the entrance aperture.

In the TCCON the small entrance aperture samples the center of the solar disk and the corresponding solar lines are narrow. Sunlight reflected at the lunar surface will have a (solar-)disk-averaged spectrum, i.e. the solar lines will be broadened as a result of the different Doppler-shifted contributions from different parts of the solar disk. GFIT includes a setting that switches to a calculation of a disc-averaged spectrum when the moon is selected as the source, therefore no bias is expected.

3.2 Calculation of dry-air mole fractions

For this analysis the current TCCON standard processing code GGG2014 was used for both solar and lunar retrievals. The retrieval code returns vertical columns (VC_{gas}), that have to be converted to dry-air mole fractions. There are two possibilities to do this. The standard TCCON processing uses the simultaneously retrieved vertical O_2 column to scale the target gas' vertical column via:

$$x_{\text{Gas}} = \frac{VC_{\text{gas}}}{VC_{O_2}} 0.2095 \quad (1)$$

The dry-air mole fraction of O_2 is well known and assumed constant; therefore systematic errors common to both vertical column retrievals cancel out using this approach.

However, for the retrieval of O_2 the spectral band at $1.27 \mu\text{m}$ (7880 cm^{-1}) is used and the detector is much less sensitive in that region compared to the CO_2 and CH_4 windows between 5800 cm^{-1} and 6400 cm^{-1} (compare Fig. 1). This results in a noisier O_2 retrieval especially under low signal-to-noise conditions (see Fig. 2).

The second option to calculate the dry-air mole fraction involves the scaling to atmospheric surface pressure and a correction for the water contained in the column:

$$x_{\text{Gas}} = \frac{VC_{\text{gas}}}{\frac{p_0 N_A}{m_{\text{dry}}^{\text{air}} \bar{g}} - VC_{H_2O} \frac{m_{H_2O}}{m_{\text{dry}}^{\text{air}}}} \quad (2)$$

Here, x_{Gas} denotes the target species' dry-air mole fraction, VC_{gas} the vertical column and p_0 the surface pressure. N_A is Avogadro's number and the molecular masses of water, $m_{H_2O} = 18.01534 \text{ g mol}^{-1}$, and dry air, $m_{\text{dry}}^{\text{air}} = 28.9644 \text{ g mol}^{-1}$, are given. \bar{g} denotes the column averaged gravitational acceleration at the measurement site and is assumed to be $\bar{g} = 9.81 \text{ m s}^{-2}$.

This approach requires accurate knowledge of the surface pressure p_0 . Additionally systematic errors, e.g. pointing errors can affect the retrieval, as they are not cancelled out via ratio with O_2 . The surface pressure measurement is performed at the Ny-Ålesund station of the Baseline Surface Radiation Network (BSRN), located adjacent to the AWIPEV observatory and thus the FTIR spectrometer. The raw pressure measurements are then scaled to compensate for the height difference to the FTS. The meteorological data is provided by AWIPEV and publicly available at doi:10.1594/PANGAEA.150000 for years until 2013, with corresponding updates for more recent years.

In the following, the approach described in equation 1 was used to retrieve x_{CO_2} and x_{CH_4} . The second approach, in equation 2, was only used to derive x_{O_2} ~~for the analysis in section 4~~ [in Section 4, which covers the validation with solar measurements](#). The main retrieval windows and the fit residuals of an example spectrum are shown in Fig. 2. The vertical column of H_2O used for the water correction in equation 2 is retrieved simultaneously in several micro-windows in the same spectral region as the target species.

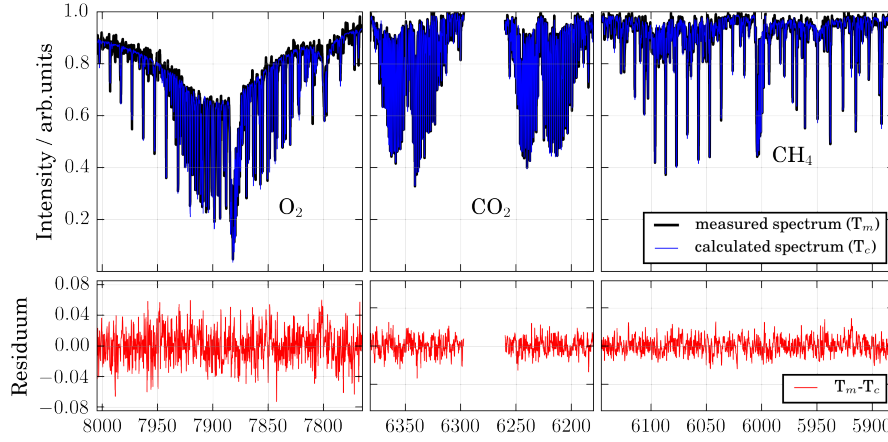


Figure 2. Example fit of a measured spectrum (black line) on October 25 2015, the corresponding calculated spectrum (blue line) and their residuum (red line) for the retrieved windows of O₂, CO₂ and CH₄.

3.3 Atmospheric model

Information on the target gas is retrieved from the processed spectra by the least-square fitting algorithm GFIT (see Sec. 3.2).

- 5 The software assumes an a priori profile of the target gas and calculates an artificial spectrum given additional information on the atmospheric profile. In TCCON the interpolation of the NCEP/NCAR reanalysis data (NCEPNCAR, 2016) to the sites latitude, longitude and local noon is used as an atmospheric model, resulting in one model profile per day. The NCEP/NCAR reanalysis data is publicly available and was provided via <http://www.esrl.noaa.gov/psd/> (NCEPNCAR, 2016). In case of
- 10 atmospheric models, i.e. the one interpolated to local noon.

Given that the reanalysis data are available in six hour time intervals, we use the model profile interpolated to the site coordinates and the time of measurement, resulting in specific model profiles for each measurement. These profiles presumably better reflect the atmospheric conditions, especially at night. The increased computational effort for this per-spectrum-model approach is affordable for this comparatively small time series.

- 15 A comparison of the differences in retrieved xCO₂ and xCH₄ between the daily and spectrum-specific model profiles is shown in Fig. 3 for the lunar time series and for selected days in the TCCON time series in Fig. 4. The two retrievals show minimal differences at local noon (as they should), but differences of about ± 0.5 ppm (CO₂) and about ± 2 ppb (CH₄) can occur later in the day, under quickly varying atmospheric conditions distant in time from local noon. Note that the measurements showing potentially large deviations are typically filtered out within TCCON as they occur at high solar zenith angles.

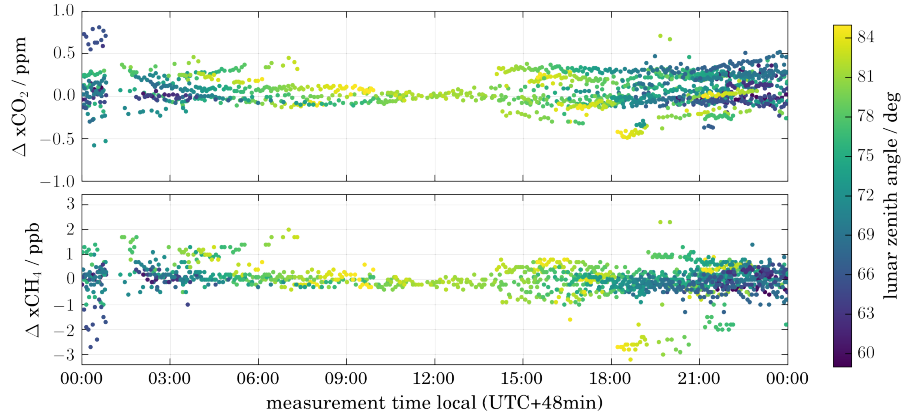


Figure 3. Differences in the lunar absorption retrieval results (2012 – 2015) using the site and time of measurement interpolated atmospheric model compared to using the model interpolated to site and local noon for both target species dependent on the lunar zenith angle.

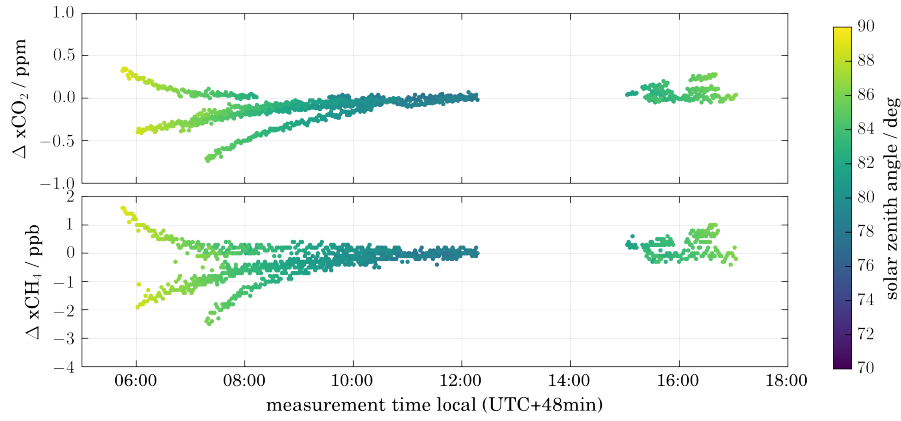


Figure 4. Same as Fig. 3 but for TCCON solar absorption measurements for the time between 2013-09-19 and 2013-09-24. Note the generally higher differences at high zenith angles. Between 12:30 and 15:00 local time the sun moves behind a mountain at lower zenith angles.

3.4 Analysis of optimal resolution

The resolution used in the TCCON is better than 0.02 cm^{-1} , corresponding to a maximum optical path difference (OPD) of 45 cm. Initial tests showed that even with the cooled detector, the spectral signal-to-noise ratio did not allow for a robust retrieval unless a lot of spectra were averaged; however, the path of moonlight through the atmosphere changes rapidly with time. Although this is more prominent in lower latitudes, it still must be considered here, especially at large lunar zenith angles. To avoid bias from inaccurate knowledge of the viewing geometry, the integration time per measurement must be as small as possible.

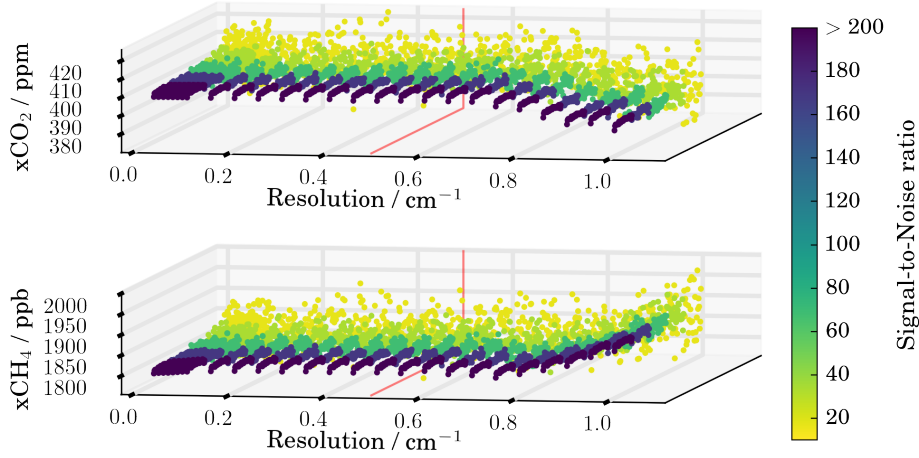


Figure 5. Retrieved $x\text{CO}_2$ and $x\text{CH}_4$ from cropped interferograms with different resolutions and different levels of white noise (z-axis and colorbar) added to the spectra.

One option to decrease the measurement time is to increase the velocity of the instrument's scanning mirror; however [previous studies \(e.g. Messerschmidt et al. \(2010\)\) have linked an increase in scanner velocity to the increase of laser sampling errors,](#)

5 [this has no effect on the spectral signal-to-noise ratio.](#) The scanner velocity was therefore not changed and kept at 10 kHz to minimise potential differences from the solar absorption measurements. The second option is to decrease the spectral resolution, which increases the spectral signal-to-noise ratio. Additionally, it allows for shorter measurement times and thus for more spectra to be averaged within the same time, resulting again in an increased signal-to-noise ratio.

10 The influence of resolution on the retrieval can be analysed in further detail and to circumvent differences arising from a varying atmospheric state. Previously, Petri et al. (2012) investigated this for the TCCON standard retrieval windows. Here the analysis was repeated with emphasis on lower resolutions (down to 1.0 cm^{-1}) and additionally spectra with different signal-to-noise ratios were used.

15 A set of 60 consecutive solar spectra has been selected and the interferograms cropped at lengths corresponding to a range of maximum optical path differences between 45 cm (0.02 cm^{-1}) and 0.9 cm (1.0 cm^{-1}). The interferograms were reprocessed and the spectra calculated with the i2s program within the GGG2014 program suite.

In addition to this series of spectra, different magnitudes of white noise were added to the created spectra to simulate the effect of the lower signal-to-noise ratio expected in lunar spectra. The signal-to-noise-ratios are calculated from the reprocessed spectra by dividing the maximum mean signal between absorption lines at about 6000 cm^{-1} by the root mean square of a blacked out region of the spectrum. Figure 5 shows the results of the standard retrieval of $x\text{CO}_2$ and $x\text{CH}_4$ for the various

20 combinations of resolution and signal-to-noise ratio of the series.

[The decrease in resolution leads to an increase in S/N. Fig. 6 shows the increase in S/N measured as a function of spectral resolution with a Bruker 125 HR, normalized to the SNR at \$0.02 \text{ cm}^{-1}\$, i.e. a spectrum recorded with \$1.0 \text{ cm}^{-1}\$ resolution has a 10 times larger S/N \(see blue line\). Additionally, the shorter scan length allows to record more spectra in the same time](#)

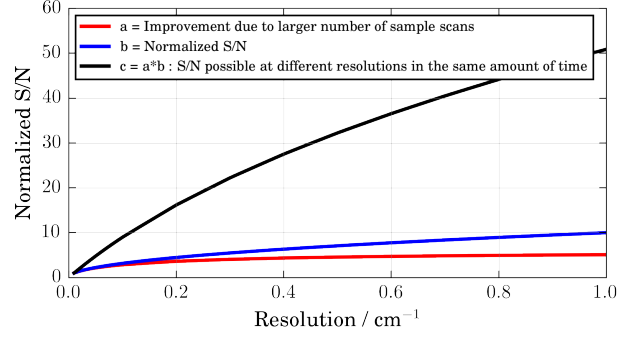


Figure 6. Spectral signal-to-noise ratio as a function of resolution. The improvement due to lower resolution (blue line) and averaging over larger number of spectra in the same time frame (red line) and the resulting relative S/N from both effects (black line), normalised to the S/N at 0.02 cm^{-1} .

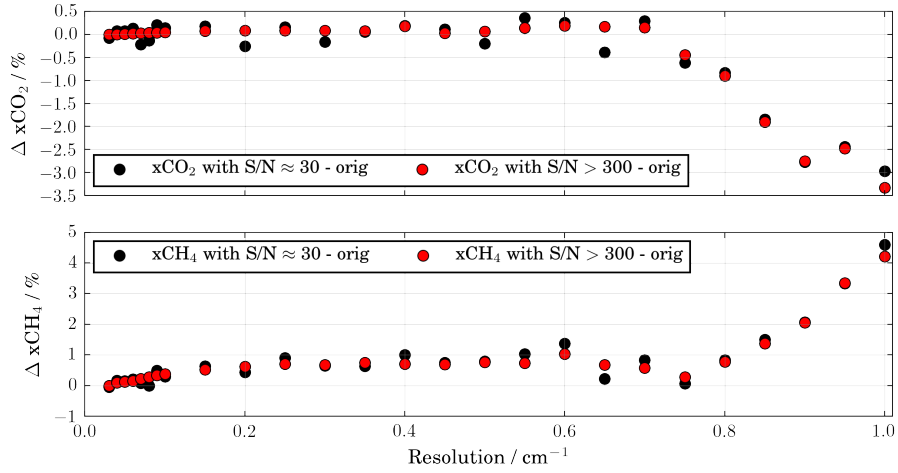


Figure 7. Mean of the retrieved $x\text{CO}_2$ and $x\text{CH}_4$ from cropped interferograms at different resolutions with low and high signal-to-noise ratio. Shown is the relative difference to the highest signal-to-noise ratio and highest resolution. Errorbars show the 1σ standard deviation of the mean.

Averaging leads to an increase in S/N by a factor of \sqrt{N} with N measurements (red line). The combination of both effects (black line) shows the potential increase in S/N with resolution for a fixed integration time. A lower resolution would potentially also allow for a larger entrance aperture. However, at lower resolutions the size of the entrance aperture is limited by the size of the image of the lunar disk, rather than the resolution.

For better visibility, Fig. 7 shows a subset of the data from Fig. 5, showing the mean retrieved $x\text{CO}_2$ and $x\text{CH}_4$ DMFs at a given resolution. The errorbars give Two series have been selected, with high (red) and low (black) signal-to-noise ratios. The associated errors can be estimated by the standard deviation (1σ) of the arithmetic mean. Two series have been selected, with

Table 1. Comparison of the biases, introduced by lower resolution measurements and low signal-to-noise ratio. Subset of data points from Fig. 7

S/N	Resolution [cm^{-1}]	$\Delta x\text{CO}_2$ [%]	$\Delta x\text{CH}_4$ [%]
> 300	0.08	0.03 ± 0.57	0.28 ± 2.61
	0.5	0.07 ± 0.65	0.76 ± 3.03
≈ 30	0.08	-0.13 ± 4.12	0.00 ± 15.03
	0.5	-0.20 ± 4.50	0.79 ± 22.89

high (red) and low (black) signal-to-noise ratios, and do not change much with resolution for a given S/N. The mean errors and their standard deviation for $x\text{CO}_2$ are 4.0 ± 0.6 ppm for the low S/N case (black dots in Fig. 7) compared to 0.6 ± 0.05 ppm for the high S/N case (red dots). Similarly the errors for CH_4 are 18.5 ± 3.2 ppb (low S/N, black dots) and 2.9 ± 0.3 ppb (high S/N, red dots).

A distinct cut-off above 0.7 cm^{-1} can be identified in the $x\text{CO}_2$. For higher resolutions, i.e. $0.02 - 0.7 \text{ cm}^{-1}$, no significant difference is visible in high signal-to-noise conditions. In general, a lower signal-to-noise ratio of the spectra leads to increased scatter of the retrieved DMFs, but to no significant bias. Table 1 shows the bias in the retrieved DMFs of high and low signal-to-noise ratio spectra for the two resolutions used in the measurement setup later.

Gisi et al. (2012) showed that lower resolution solar spectra can be used to retrieve DMFs with a low resolution FTS (Bruker EM27/SUN). Recently Hedelius et al. (2016) investigated errors and biases from a 0.5 cm^{-1} FTS (Bruker EM27) for TCCON relevant species. The three studies (Petri et al., 2012; Gisi et al., 2012; Hedelius et al., 2016) report different biases in $x\text{CO}_2$ when changing the resolution to 0.5 cm^{-1} in the range from -0.12% to 0.13% . For $x\text{CH}_4$, Hedelius et al. (2016) reported an increase of 0.28% when decreasing the resolution to 0.49 cm^{-1} . In our analysis (see Tab. 1) a consistent decrease in mean $\Delta x\text{CO}_2$ and $\Delta x\text{CH}_4$, i.e. the difference between DMFs from low and high resolution spectra, is observed when moving to lower resolutions. However, when considering the assigned errors (1σ standard deviation) this is not significant, especially under lower signal-to-noise conditions.

For the final decision on the best resolution for low S/N conditions the possible number of recorded spectra per time interval has to be considered. This number does not increase linearly, due to instrumental effects, i.e. the deceleration of the moving mirror and the time needed for data acquisition and storage. The first measurements were taken at a reasonably high spectral resolution of 0.08 cm^{-1} (OPD = 11.25 cm). The measurement setup was adjusted after further tests. The benefit of a better signal-to-noise ratio on the measurement precision lead to finally decreasing the resolution to 0.5 cm^{-1} (OPD = 1.8 cm) and all measurements from 2015 onwards were taken with a resolution of 0.5 cm^{-1} .

The effect of different resolutions on the retrieved columns can also be investigated by comparing different measurements taken consecutively with different resolutions. Figure 8 shows lunar absorption measurements of the target species on October

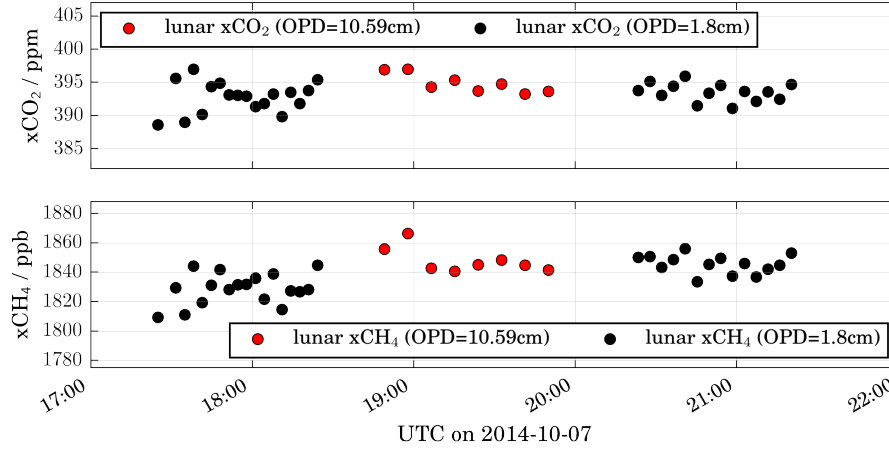


Figure 8. Comparison of retrieved $x\text{CO}_2$ and $x\text{CH}_4$ for different resolutions from low (~~$\text{OPD} = 1.8 \text{ cm} \hat{=} 0.5 \text{ cm}^{-1}$~~ $\text{OPD} = 1.8 \text{ cm} \hat{=} 0.5 \text{ cm}^{-1}$, black) and higher (~~$\text{OPD} = 10.59 \text{ cm} \hat{=} 0.085 \text{ cm}^{-1}$~~ $\text{OPD} = 10.59 \text{ cm} \hat{=} 0.085 \text{ cm}^{-1}$, red) resolution measurements on 2014-10-07.

7 2014. The first and third batch of measurements were taken with a resolution of 0.085 cm^{-1} ($\text{OPD} = 10.59 \text{ cm}$), the second batch was measured with 0.5 cm^{-1} ($\text{OPD} = 1.8 \text{ cm}$) resolution. No significant bias is observed.

Decreasing the spectral resolution also changes the information content of the recorded spectral lines. This results in a
5 change in shape of the measurements averaging kernels and is discussed below.

3.5 Averaging Kernels

The sensitivity of the retrieved dry-air mole fraction of the target gas depends on the a priori information and the measurement's altitude dependent sensitivity, i.e. the averaging kernels. The a priori profiles used are the default TCCON ones. The averaging kernel of a measurement strongly depends on the retrieval methodology and the information content of the corresponding spectrum. As such it depends on the viewing geometry as well as the resolution, the absorption strength and the signal-to-noise ratio. The ~~averaging kernels strongly depend on the information content of the spectrum. The~~ weight different altitude levels have in the retrieval can be parameterized as a function of the zenith angle. As the instrument faces the light source at a certain zenith angle, the measurement samples different contributions from the various atmospheric layers. The pressure broadening of the absorption features shows a specific altitude dependent sensitivity and this information depends on the chosen resolution
15 and the signal-to-noise ratio of the measurement.

The setup of the lunar measurements is similar to that of TCCON measurements, therefore the averaging kernels are quite similar, aside from effects of resolution and noise for a given zenith angle.

The top panel in Fig. 9 shows the averaging kernels for the lunar measurements. The middle panel shows the difference from the standard TCCON ones from Ny-Ålesund, interpolated to the corresponding zenith angles. The lines are color gradient coded with their respective zenith angles and different color schemes reflect different resolutions.

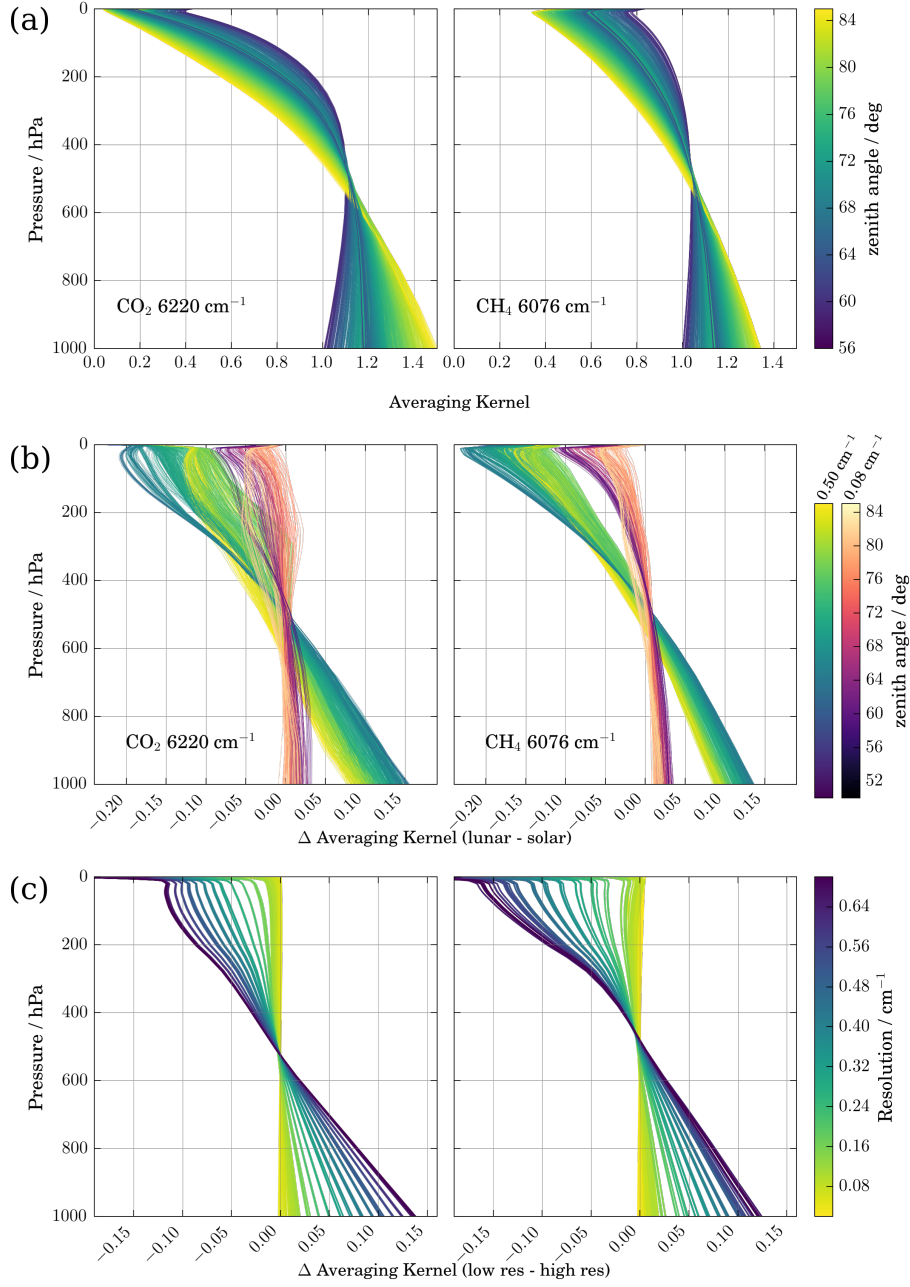


Figure 9. (a) Averaging kernels of the lunar measurements. (b) Difference between lunar and solar averaging kernels color coded for different spectral resolutions. (c) Differences between low resolution and TCCON spectra averaging kernels as a function of resolution.

Pressure broadening leads to spectral lines originating from gases at low pressure being narrower than those at higher pressure. The narrow part of a spectral line sampled with fewer points therefore cannot give as much information as one with

Table 2. Comparison of the retrieved solar, lunar and model DMFs for the two comparison time periods. Note that xO_2 was calculated using the surface pressure and the offset to the true atmospheric value of 20.95% is caused by spectroscopic errors.

		xCO_2 [ppm]	xCH_4 [ppb]	xO_2 [%]
March 2013	solar	397.47 ± 0.67	1773.78 ± 2.99	21.33 ± 0.08
	lunar	396.81 ± 3.89	1775.72 ± 17.64	21.34 ± 0.36
	Jena CO_2	398.01 ± 0.13	–	–
	CT15 CO_2	396.89 ± 0.22	–	–
	MACC CO_2	397.16 ± 0.18	–	–
	MACC CH_4	–	1784.09 ± 1.06	–
September 2013	solar	393.16 ± 0.49	1810.26 ± 3.11	21.38 ± 0.06
	lunar	392.15 ± 8.03	1813.62 ± 38.02	21.40 ± 0.60
	Jena CO_2	391.56 ± 0.26	–	–
	CT15 CO_2	391.29 ± 0.24	–	–
	MACC CO_2	392.07 ± 0.39	–	–
	MACC CH_4	–	1800.79 ± 1.58	–

higher resolution. This leads to averaging kernels from low resolution spectra being less sensitive to the stratosphere and more sensitive in the lower troposphere than their high resolution counterparts. This can be seen in the lower panel of Fig. 9, where the difference between standard TCCON averaging kernels and their lower-resolution counterparts at the same zenith angle is shown. As expected, decreasing the spectral resolution leads to greater differences between the averaging kernels.

4 Validation with solar absorption spectroscopy

The validation of the measurements performed during the polar night is difficult. In the absence of other options, here we compare to solar absorption measurements taken within TCCON. In spring and autumn there are a few consecutive days around the full moon where solar absorption measurements during the day and lunar absorption measurements during the night are possible. Such comparison measurements were performed in March and September 2013. Here the DMFs of xCO_2 and xCH_4 for both solar and lunar measurements were retrieved using equation 1. For the comparison of xO_2 equation 2 was used, respectively.

Assuming the total column values do not change significantly during that time period, the means of the two retrievals can be compared directly. Figure 10 shows the comparison results and the calculated means for a comparison in September 2013. Table 2 shows the corresponding values of the arithmetic mean and its standard deviation as an indication of the error for both comparison campaigns in March and September 2013. The same analysis was performed on the available smoothed model output. The calculated standard deviation of the models of about 0.2 ppm (March) and 0.3 ppm (September) for CO_2

and 1.0 ppb and 1.6 ppb respectively for CH₄ indicates that the assumption of stable DMFs for the observed time frame is reasonable. ~~During this brief comparison time period, no significant bias~~

5 The accuracy of the lunar measurements can be determined via the bias of the lunar compared to the solar measurements and can be deduced from Table 2 as well. In March 2013 the difference between solar and lunar measurements is 0.66 ± 4.56 ppm for xCO₂ and -1.94 ± 20.63 ppb for xCH₄. In the September 2013 campaign a bias of 1.01 ± 8.52 ppm for xCO₂ and -3.36 ± 41.13 ppb for xCH₄ can be observed~~when comparing the lunar with the solar DMF retrievals~~. The diurnal variability of the lunar measurements is used to define the precision. As the later measurements have a higher precision, a typical value achieved in the 2014/2015 winter is used. Here the standard deviations of the daily mean of 2 ppm for xCO₂ and 10 ppb for
10 (xCH₄), corresponding to 0.5 % in both cases.

The target accuracy can be estimated via the detrended year-to-year wintertime variability. Here model output can be used as a proxy. In the smoothed, detrended MACC CO₂ and CH₄ model (see Sec. 5.1) the arithmetic mean of the first week of January differs by 0.55 ppm in xCO₂ and 9.84 ppb in xCH₄ between 2012 and 2014. At the same time, the standard deviation of all values for the first week of January between 2012 and 2014 is about 1.8 ppm for xCO₂ and 18.8 ppb for xCH₄.
15 However, these estimates are potentially subject to unknown biases in the models, i.e. the model could be biased similarly every year. Additionally, the seasonal variability surely is an upper limit for the target precision. Here the seasonal cycle amplitude measured by solar FTS is about 15 ppm for xCO₂ and about 40 ppb for xCH₄.

As described in section 3.2 (see equation 1), the dry-air column is calculated using the vertical column of O₂, retrieved from the 7885 cm⁻¹ spectral region. Here ~~air-glow~~ airglow emissions in the high atmosphere could potentially disturb the
20 O₂ spectra. This can typically be ignored in solar absorption spectra, as the magnitude of the emissions is negligible, when viewing directly into the sun. In case of lunar spectra, however, air-glow emissions could potentially fill in the spectral lines and influence the measurements. To test this, xO₂ was retrieved using the surface pressure to calculate the dry-air column as described in equation 2.

In both comparison periods, no significant difference between the solar and lunar retrievals of xO₂ can be observed. Note
25 that xO₂ retrieved via surface pressure shows an offset of 0.4 % in both cases (lunar and solar). This offset originates in the line parameters used for the O₂ retrieval and is compensated in the xCO₂ and xCH₄ retrieval with the TCCON in-situ correction. Washenfelder et al. (2006) reported values that are 2.27 ± 0.25 % larger if the surface pressure retrieved dry column was used. Here we find a mean difference of 1.96 ± 0.14 %, when calculating the mean and standard deviation of the solar and lunar mean xO₂ values shown in the sidebars in Fig. 11. Note that these retrievals were performed with updated spectroscopy available
30 within GGG2014 compared to that used by Washenfelder et al. (2006).

5 Seasonal cycle and model comparison

5.1 Method – model comparison

The rigorous comparison of ground-based column measurements of a trace gas to model simulations requires resampling the model profile as if it was measured by the instrument.

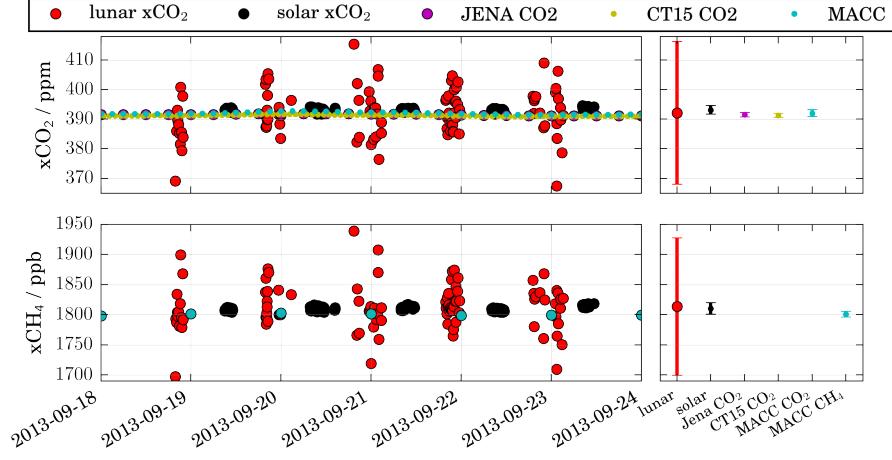


Figure 10. Comparison of the solar and lunar measurements of $x\text{CO}_2$ and $x\text{CH}_4$ in September 2013 (dots) and the corresponding arithmetic means (lines). Values are given in Table 2.

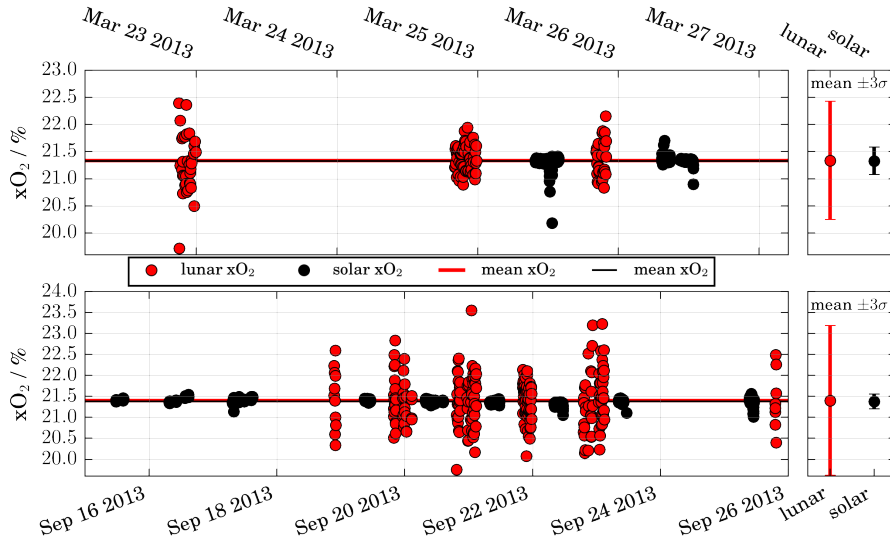


Figure 11. Comparison of the solar and lunar measurements of $x\text{O}_2$ in March and September 2013.

The smoothed column dry-air mole fraction \hat{c} can be calculated following Rodgers and Connor (2003); Connor et al. (2008); Wunch et al. (2010) by adding the column integrated a priori profile (c_a) to the difference between the model (\mathbf{x}) and the dry TCCON a priori profile (\mathbf{x}_a) weighted with the averaging kernel (\mathbf{a}):

$$\hat{c} = c_a + \mathbf{h}^T \mathbf{a}^T (\mathbf{x} - \mathbf{x}_a) \quad (3)$$

Here, h represents the pressure weighting function (see Connor et al. (2008)).

- 5 Given a vertical model profile, the measurement's averaging kernel and the vertical columns of water vapour and the a priori profile of the target gas, the smoothed dry-air mole fraction of the model output can be calculated. Due to the high random error of the lunar FTS measurements, daily means have been calculated for both the measurements and the model data, after the smoothing was applied.

5.2 Results – time series

- 10 In this section the FTIR time series is compared to CO₂ model results from three different models: the MACC CO₂ model version 14r2 (MACCCO₂, 2016), the CarbonTracker 2015 (CT2015, 2016) model and the Jena CO₂ inversion version s04_v3.7 (JenaCO₂, 2005). In case of the CH₄ time series, the MACC CH₄ v10 (MACCCH₄, 2016) is used. As described in Section 5.1 the models DMF profile has been smoothed with the corresponding a priori and averaging kernel of the lunar and solar measurement, respectively. For times where there are no FTS measurements available, an averaging kernel was calculated
15 using the solar zenith angle of the corresponding time. In winter the lunar zenith angle was used instead. For times where no FTS measurements were possible at all, e.g. sun and moon are below the horizon, a mean zenith angle of 65° was assumed.

The resulting model time series can now be compared directly to the FTS measurements. Figure 12 shows the comparison of the FTS and the smoothed model time series for CO₂. The CH₄ comparison is shown in Fig. 13.

5.3 Results – seasonal cycle

- 20 The detrended seasonal cycles of ~~the both~~ target species are similar from year to year, ~~except for a secular increase in both, and~~. In the following ~~the de-trended~~, the detrended seasonal cycles are compared to the models already discussed in section 5.2.

Figure 14 shows the seasonal cycle of xCO₂ as observed with the Ny-Ålesund FTS between 2012 and 2016, detrended with a linear increase of 2.6 ppm yr⁻¹, an offset of 380.0 ppm on 2012-01-01 and condensed to one year. The seasonal cycle of xCO₂ shows little difference between the three models, therefore the comparison can be performed with an model average.

- 25 The shaded area in Fig. 14 shows the 3 σ standard deviation around the daily mean of the combined model data points of all three models (MACC, CarbonTracker and Jena). The weighted average of all FTS measurements during one full moon period is shown (green dots) with errorbars corresponding to the standard error (σ/\sqrt{N}) of the daily mean calculated from N measurements. The weights are chosen to be the inverse squared residual of the spectral fits.

- The difference between the models and the TCCON measurements in summer is quite small, except for a phase shift in the
30 onset of the downward slope at the beginning of the growing season decline. In winter the models agree well with the FTIR lunar absorption measurements, within the given error margin.

In the case of CH₄ a similar comparison has been performed and the results can be seen in Fig. 15. Here the xCH₄ time series have been linearly detrended with an annual increase of 10.6 ppb yr⁻¹ and an offset of 1760.0 ppb on 2012-01-01. Figure 15 shows the 3 σ standard deviation around the daily means of the MACC ~~CH4~~ CH₄ model (shaded area) compared to the FTS measurements (~~green-red and blue~~ dots) averaged over one full moon measurement cycle. The errorbars correspond to the 1 σ standard deviation of the mean.

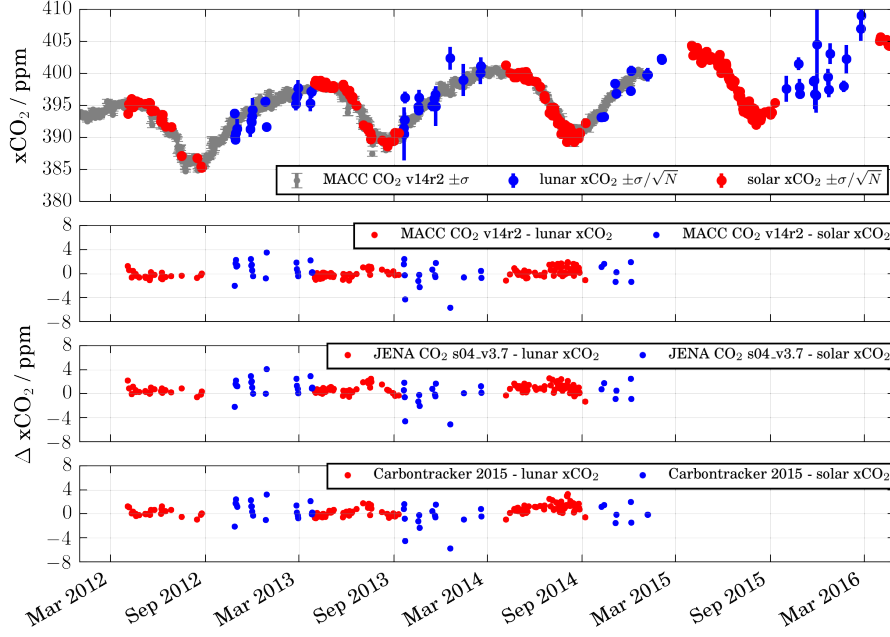


Figure 12. Comparison of the daily means of lunar (blue) and solar (red) $x\text{CO}_2$ FTIR measurements to the AK-smoothed MACC CO_2 model v14r2 (top panel, gray). Errorbars show the standard error (σ/\sqrt{N} , with N number of measurements). The lower panels show the difference model - measurement for all models.

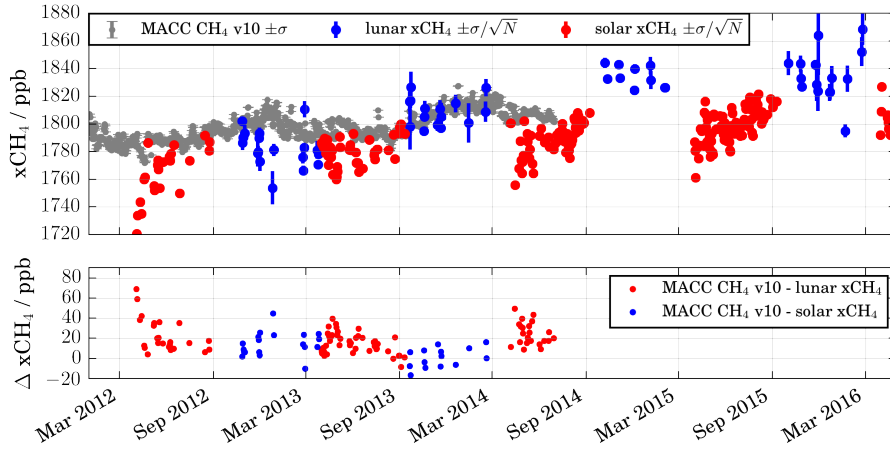


Figure 13. Comparison of the daily means of lunar (blue) and solar (red) $x\text{CH}_4$ FTIR measurements to the AK-smoothed MACC CH_4 model v10 (gray). Errorbars show the standard error (σ/\sqrt{N} , with N number of measurements). The lower panel shows the difference model - measurement.

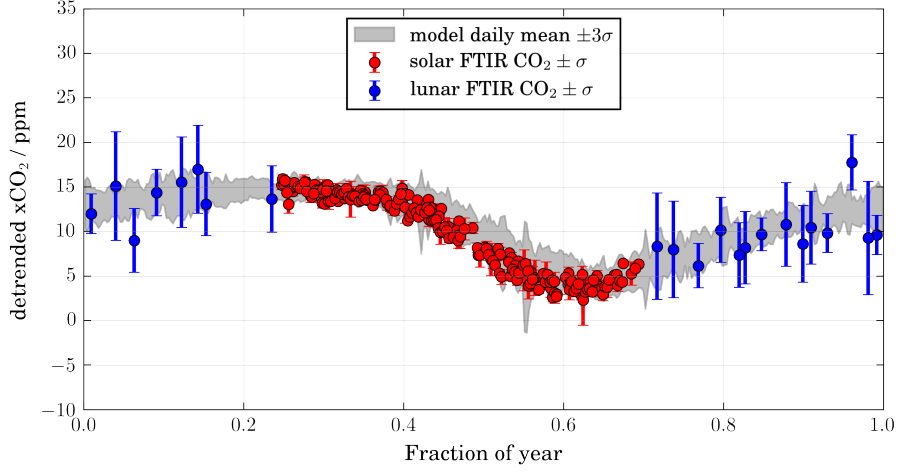


Figure 14. Comparison of solar (red) and lunar (blue) $x\text{CO}_2$ FTIR measurements (green dots). Errorbars show 1σ standard deviation of the daily mean. The lunar data points have been averaged over one full moon period each. The shaded gray area shows the 1σ standard deviation of the three model daily means (MACC, CarbonTracker and Jena) as shown in Fig. 12.

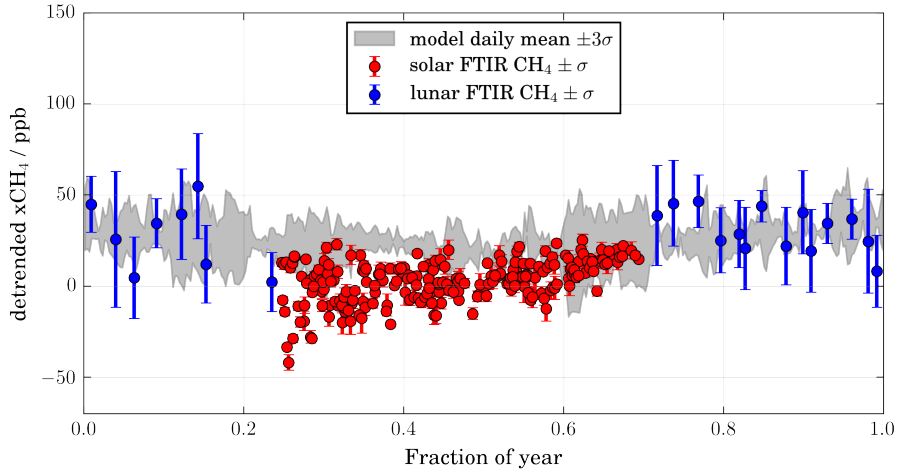


Figure 15. Comparison of solar (red) and lunar (blue) $x\text{CH}_4$ FTIR measurements (green dots). Errorbars show 1σ standard deviation of the daily mean. The lunar data points have been averaged over one full moon period each. The shaded gray area shows the 1σ standard deviation of the MACC CH_4 model daily means as shown in Fig. 13.

In spring/summer the FTS measurements show generally smaller values than the model and a larger spread. From late summer throughout the winter the measurements are in better agreement with the model. At specific events in spring, the FTS measurements show sudden decreases of $x\text{CH}_4$ (compare Fig. 13 and Fig. 15). This could be due to the model not being able to capture vertical transport very well, which has been shown previously by Ostler et al. (2016). Here, stratospheric intrusions

during the breakdown of the polar vortex in spring can lead to large, short-term decreases in $x\text{CH}_4$. This is currently being investigated by using a stratospheric species as a tracer to separate the $x\text{CH}_4$ column in a tropospheric and stratospheric part and exceeds the scope of this paper.

6 Conclusions

Measurements of the column averaged dry-air mole fractions of CO_2 and CH_4 have been performed in the polar night from 2012 to 2016 to complement the established solar absorption measurements within the TCCON. The newly employed thermoelectrically cooled InGaAs detector allows the usage of reflected sunlight on the full lunar disc to serve as a light source above the atmosphere to perform lunar absorption spectroscopy in the near-infrared spectral region.

Aircraft or AirCore profiles are not yet available for Ny-Ålesund. The lunar absorption measurements have therefore been validated with standard TCCON measurements in spring and autumn 2013 and the comparison shows no significant biases. The decrease of spectral resolution allows for an increase of the spectral signal-to-noise ratio, which in turn decreases the random error significantly. Under optimal conditions, lunar measurements with standard deviation of the daily mean (1σ) of about 2 ppm for $x\text{CO}_2$ and about 10 ppb for $x\text{CH}_4$ can be achieved using this approach. This corresponds to a precision of about 0.5 % for each gas.

The newly created time series has been compared to different model simulations. All three CO_2 models (MACC ~~CO2~~- CO_2 model v. 14r2), CarbonTracker 2015, Jena CO_2 inversion s04_v3.7) are generally in good agreement with the FTIR measurements. The $x\text{CH}_4$ time series shows large deviations in spring/summer and an overall good agreement in autumn/winter.

7 Data availability

The TCCON data are publicly available from the TCCON archive at <http://tccon.ornl.gov/>. The solar and lunar measurement data used in this study has been uploaded to the PANGAEA database and is available at <https://doi.pangaea.de/10.1594/PANGAEA.872007>. The MACC model data can be accessed via <http://www.gmes-atmosphere.eu/>. The Jena CO_2 inversion is provided via <http://www.bgc-jena.mpg.de/CarboScope> and the Carbontracker data can be found at <http://carbontracker.noaa.gov>. The NCEP/NCAR reanalysis is provided by NOAA via <http://www.esrl.noaa.gov/psd/>. The surface meteorology data from the Ny-Ålesund BSRN station used here is available from doi:10.1594/PANGAEA.150000.

Author contributions. The measurements were taken by M. Buschmann, the co-authors and the AWIPEV station staff. M. Buschmann performed the analysis and prepared the manuscript with contributions from all co-authors.

Competing interests. The authors declare that they have no conflict of interest.

Acknowledgements. This project was funded from the German Research Foundation (DFG) as part of the grant NO 404/17. M.Buschmann
5 received additional funding from the Helmholtz Earth System Science Research School (ESSReS), through the SFB/TR172 “Arctic Amplification: Climate Relevant Atmospheric and Surface Processes, and Feedback Mechanisms (AC)³” by the DFG and the GAIA-CLIM Horizon-2020 project of the European Union. The authors would like to thank the station personnel of the AWIPEV station, who operate the instrument on a routine basis. We further thank the AWI staff at Potsdam for providing the long term observations of surface meteorology and we thankfully acknowledge the provision of NCEP Reanalysis data by the NOAA/OAR/ESRL PSD, Boulder. The FTIR Observations in Ny-Ålesund are funded by the EU projects InGOS and ICOS-INWIRE, and by the Senate of Bremen. Nicholas Deutscher is supported by an ARC-DECRA, DE140100178. We thank Stanley Sander at JPL for initial discussions on the detector design. Additionally we would like to thank Frederic Chevallier at LSCE for providing the MACC model data and for helpful comments on an earlier version of the paper.

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