Toward CO2 and CH4 measurements by ground-based observations of surface-scattered sunlight: Instrumentation and experiments

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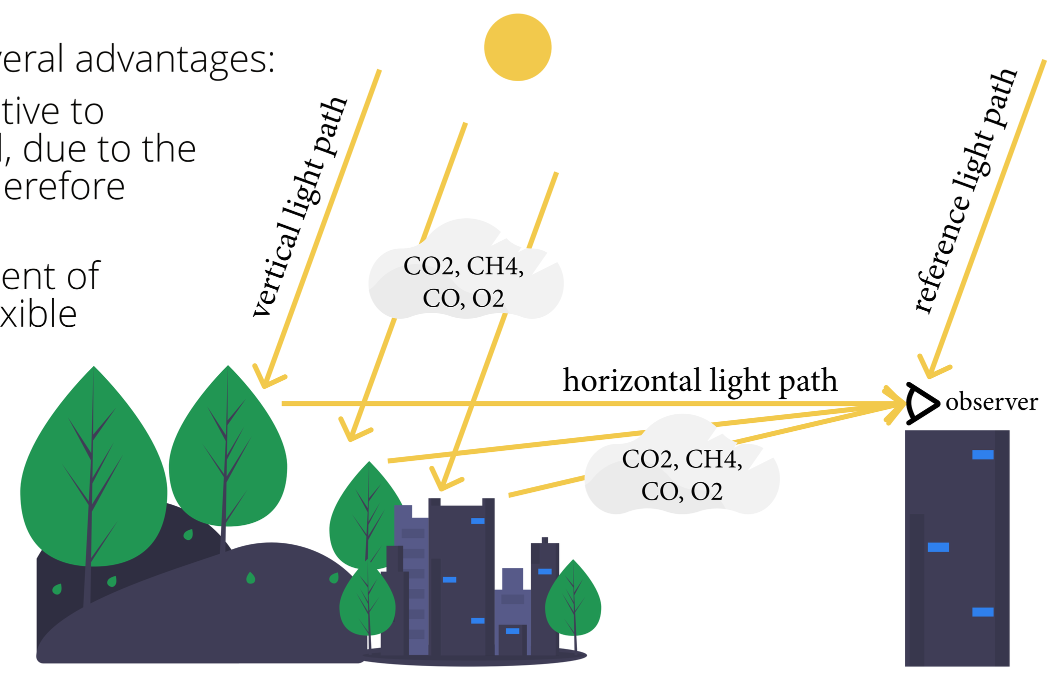
1 Introduction

We modify a mobile, commercially available Fourier transform spectrometer (FTS) for the ground-based measurement of surface scattered sunlight spectra. Spectra in the range of 4000 – 14000 cm^{-1} are recorded. In this region there are absorption bands for CO_2 (6300 cm^{-1}), CH_4 (6000 cm^{-1}), CO (4250 cm^{-1}) and O_2 (7900 and 13100 cm^{-1}), enabling the retrieval of the column density of the respective gasses.

The use of scattered sunlight has several advantages:

- The measurements are more sensitive to concentrations close to the ground, due to the horizontal path component, and therefore ideal for e.g., emission monitoring.
- Spectra can be recorded independent of the sun's position, allowing for a flexible choice of observation targets.
- Together with atmospheric scattered sunlight spectra insights into atmospheric scattering processes can be gained.



2 Instrument

EM27/SUN

We start from the EM27/SUN Fourier transform spectrometer for the measurement of CO_2 and CH_4 column densities.

- Reliable, mobile and commercially available
- Measures absorption spectra from direct solar radiation
- Equipped with a motorized automatic pointing system

Modifications

To record spectra of ground scattered solar radiation a more sensitive detector is used, and the optical throughput of the instrument is increased.

New Sensor:

- A custom designed sensor and amplifier replaces the standard sensor.
- InGaAs PIN photodiode (G12183-203K from Hamamatsu) with a cutoff wavelength of 2.57 µm.
- Detector operating temperature of T_{op} = -20 °C by a two-stage thermoelectric cooling. Dark signal noise reduced by a factor of 4.

Increased Optical Throughput:

- All apertures are removed from the light path inside the instrument.
- A larger parabolic mirror focuses more light on the photodiode.
- The field of view (FOV) is increased from 0.17° to 0.5° (full angle).

Imaging Camera:

- Specific targets by imaging camera boresighted with the optical axis of the FTS.
- Camera FOV of 7.3° is larger then the instrument FOV.
- Calibration of the camera's FOV to the instrument's FOV by a small but bright thermal light source.
- Well known targets make it possible to determine the viewing geometry, especially the horizontal path component.

Lambertian Reflector:

- Spectrally calibrated Lambertian reflector target allows measurements of the reference light path

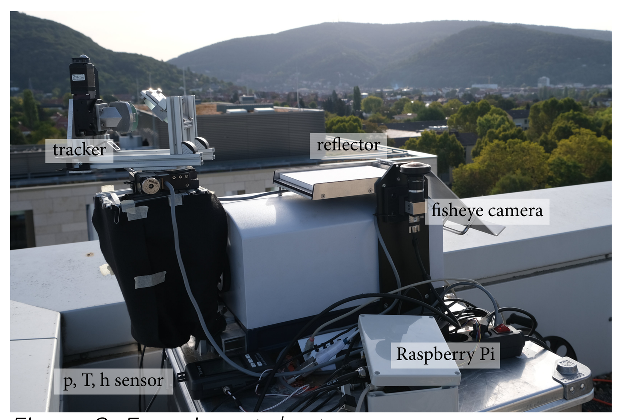


Figure 2: Experimental setup

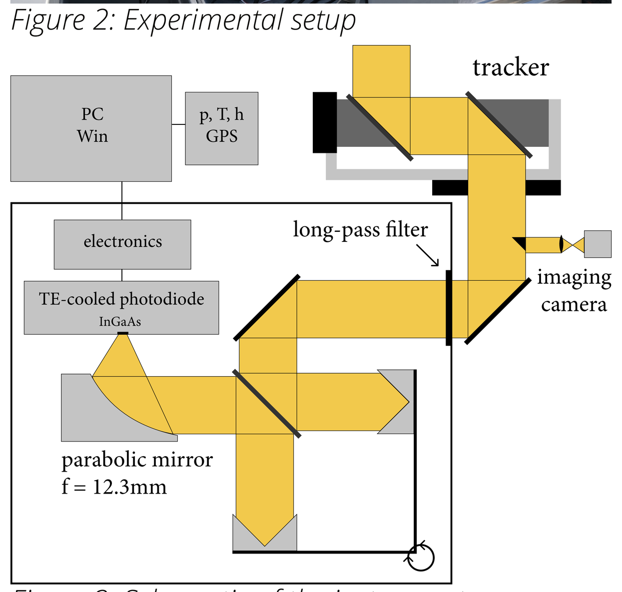


Figure 3: Schematic of the instrument

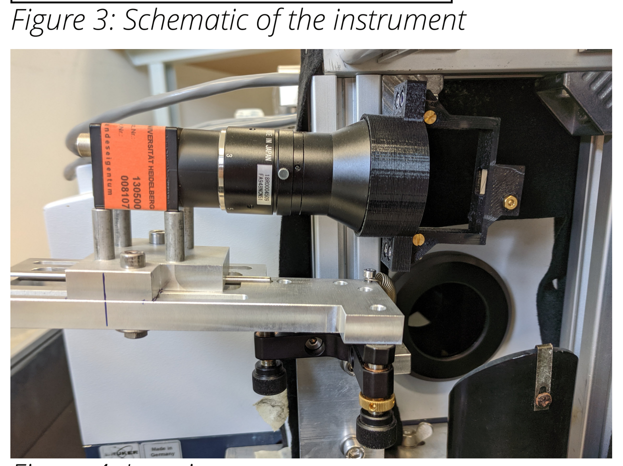


Figure 4: Imaging camera

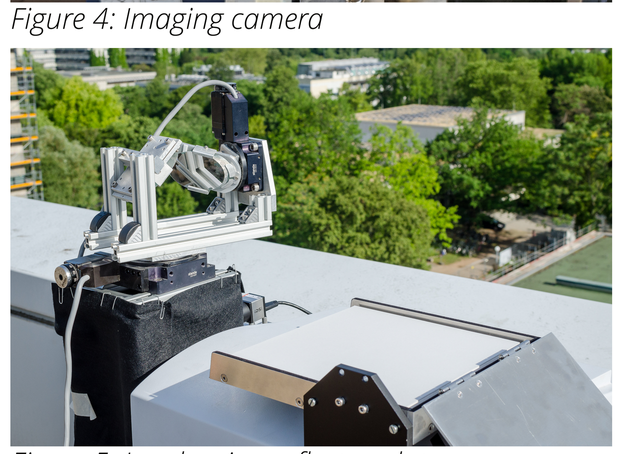


Figure 5: Lambertian reflector plate

3 Signal to Noise Ratio (SNR)

- To verify the SNR, we took alternate spectra of a surface-scattering target and the reflector plate throughout a full day. Each spectrum is generated from 10 double sides interferogram scans, resulting in an exposure time of one minute.
- The SNR is calculated as the ratio of the mean radiance in an interval close to the respective absorption band and the standard deviation in the region between 2000 and 3500 cm^{-1}, beyond the photodiode's cutoff wavelength.
- Dependence of the SNR on the solar zenith angle (SZA) depends on reflection properties of the scattering target. For CO_2, CH_4 and $O_2\Delta$: SNR > 200

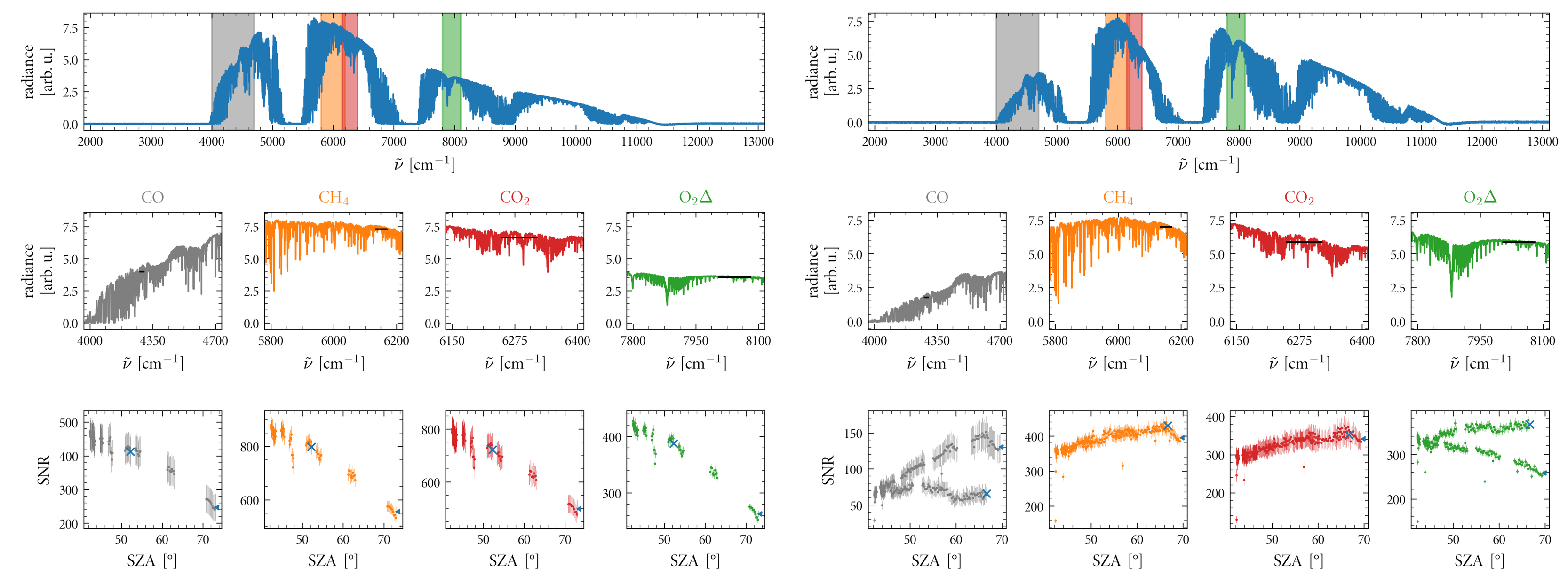


Figure 6: SNR for the measurements on 04.09.2019. The upper two panels display a full, arbitrarily chosen, spectrum from that day and the regions with the respective absorption bands. The triangle marks the start of the measurement series, the x marks the end.

4 Instrument Line Shape (ILS)

- The ILS is retrieved with the procedure by Frey et al. (2015).
- After thermalizing, 30 interferograms of an external light source are recorded with one minute exposure time. The water absorption lines in the range of 7000 – 7400 cm^{-1}, caused by humidity, are used to retrieve the ILS.
- Difficulties arise from the enhanced optical throughput and the increased field of view.
- A large and homogeneous, yet dim light source is needed. We currently use the Lambertian reflector plate for this.
- FWHM of the ILS is 0.54 ± 0.03 cm^{-1}

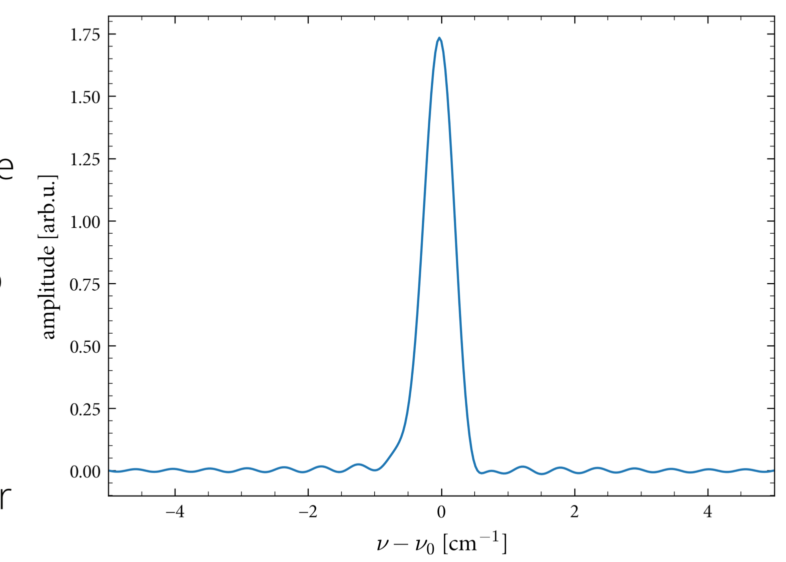


Figure 7: ILS of the modified EM27/Sun

5 Retrieval of Vertical Column Densities (VCD)

- We use the RemoTeC algorithm to retrieve VCDs of CO_2, CH_4 and O_2, neglecting atmospheric scattering.
- To obtain VCDs in surface-scattering geometry, the air mass factor in the lowest atmospheric retrieval layer is enhanced according to the length of the geometric horizontal light path.
- Column averaged mole fractions XGHG are calculated as GHG / O_2 · 0.2095
- The lower XCO_2 for the surface-scattered spectra is most likely caused by spectral differences of in-scattering on the horizontal light path.

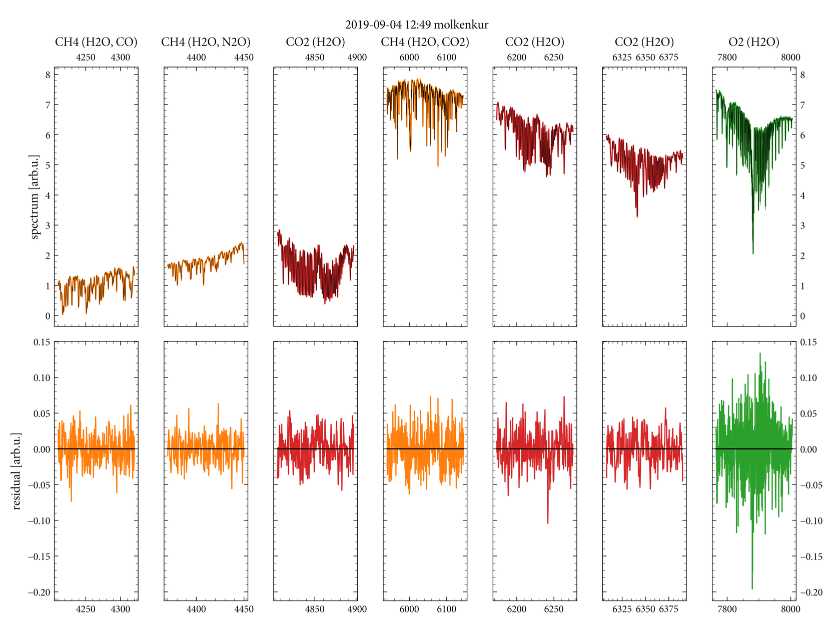


Figure 8: The upper panel shows the measured (colored) and modeled (black) spectrum of surface-scattered light. The lower panel shows the residual between both.

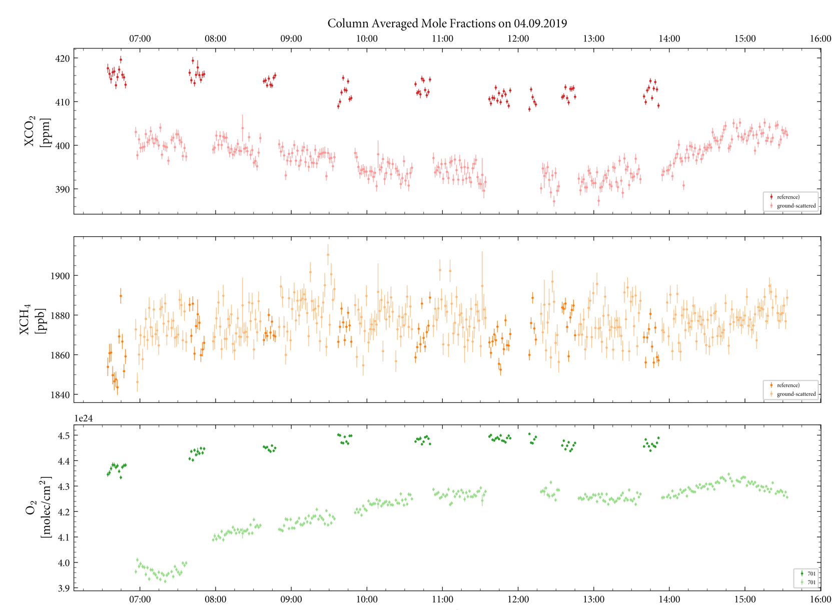


Figure 9: XCO_2 and XCH_4 calculated from the 6000 and 6300 cm^{-1} windows over the course of 04.09.2019 for surface-scattering (light) and reference light path (dark).

6 Precision

- From the retrievals of multiple days, we calculate 5 min moving averages. Averages, which contain less then 3 data points, are excluded.
- The deviation from this moving average (ΔXGHG) is shown in Figure 9 and as histogram in Figure 10.
- Precision of 1 min spectra is given by the standard deviation of the difference to the moving average.
- XCO_2: 1.3ppm for reference and 1.8ppm for surface-scattered measurements
- XCH_4: 6ppb for reference and 9ppb for surface-scattered measurements

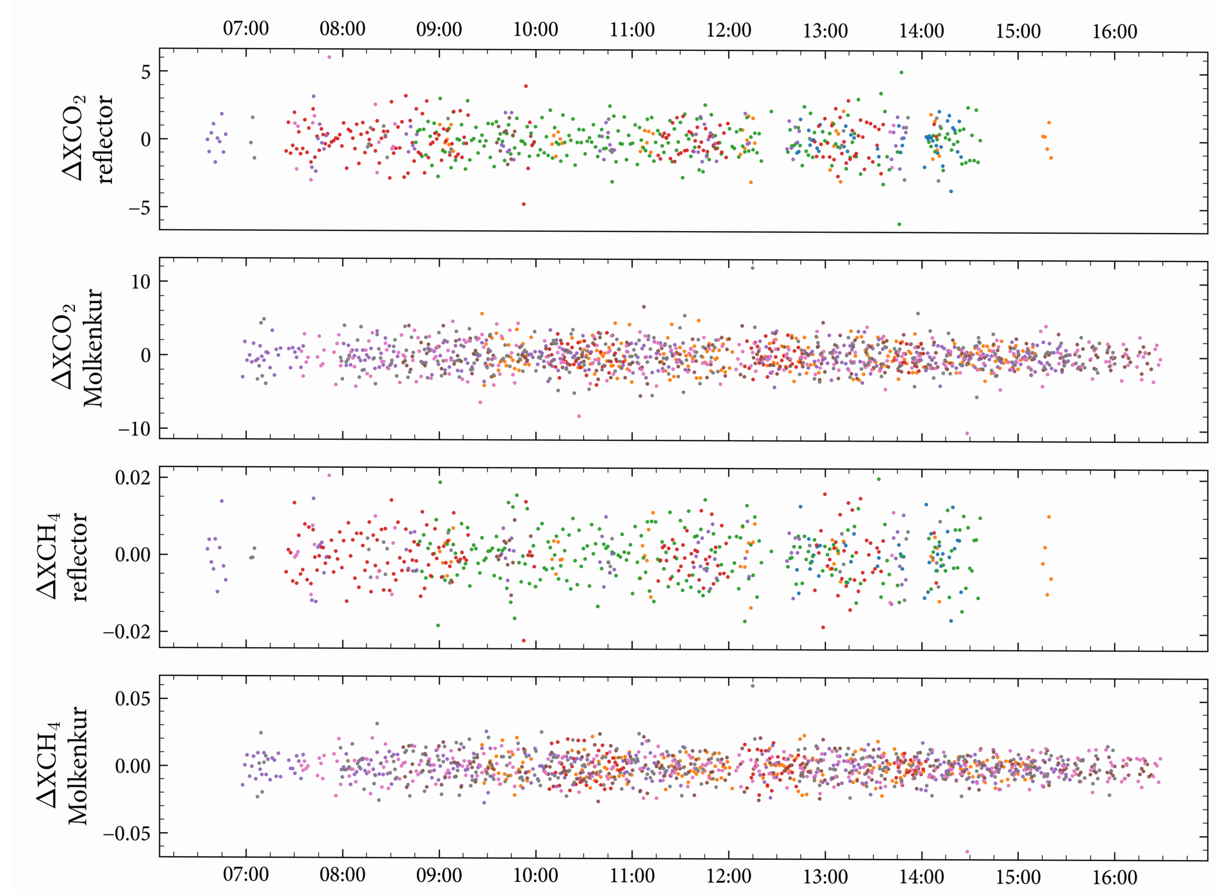


Figure 9: Time series of the XCO_2 and XCH_4 deviation from the 5 minute moving average.

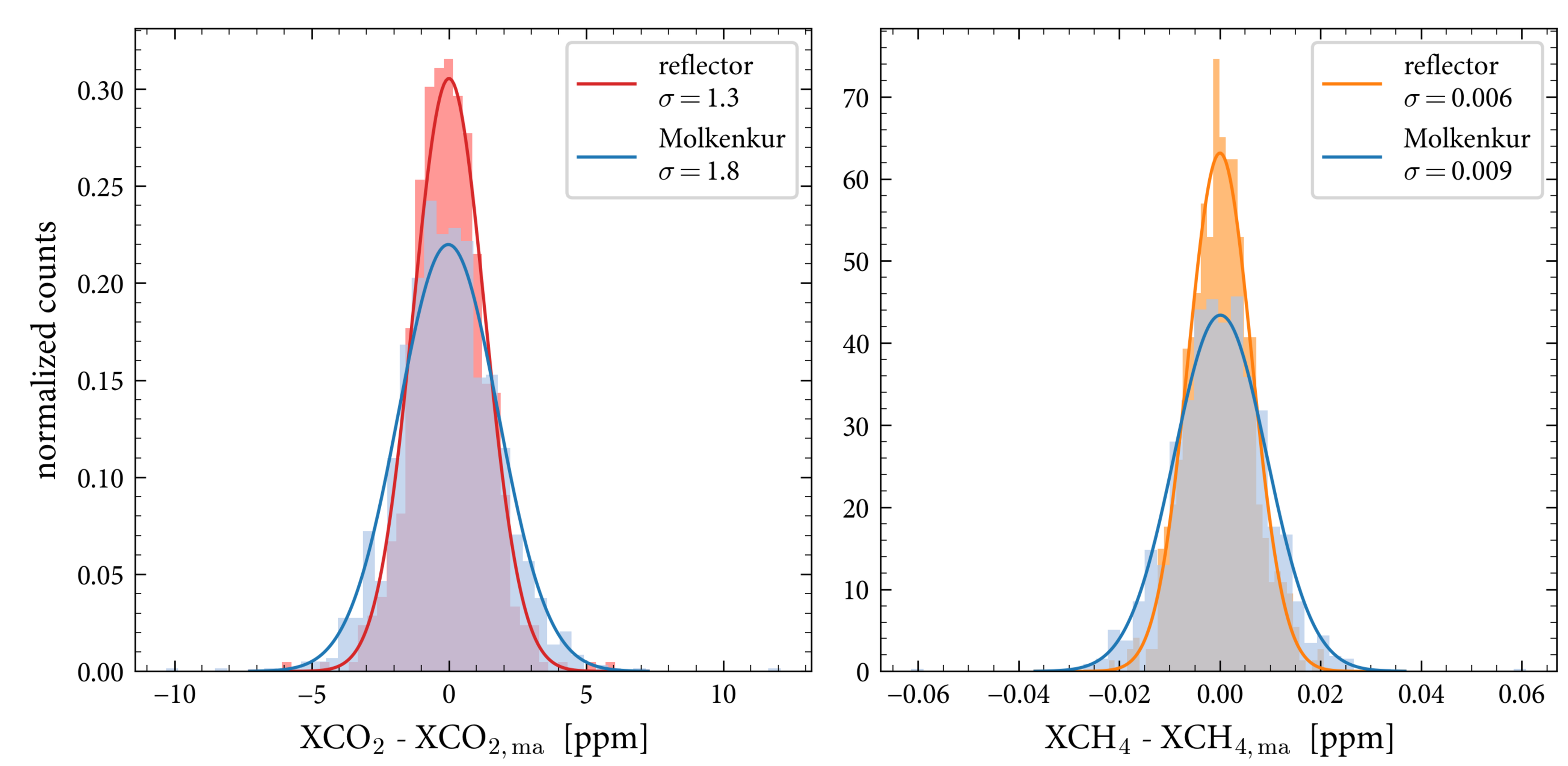


Figure 10: Histogram of the differences to the 5 min moving averages in XCO_2 and XCH_4.

7 Outlook

- Reliable automation of the tracking process as well as an automated recording of the viewing geometry. This will enable regional scans, with multiple devices even two dimensional.
- Radiometric calibration to absolute radiance to quantify atmospheric scattering properties and include scattering process in the trace gas retrievals.

References

- Hemmer, 2019: Toward ground-scattered sunlight measurements of carbon dioxide and methane, Master's thesis
- Kostinek, 2015: Enhancing optical throughput and detector sensitivity of the EM27/FTS IR spectrometer, Master's Thesis
- Frey et al., 2015: Calibration and instrumental line shape characterization of a set of portable FTIR spectrometers for detecting greenhouse gas emissions, Atmos. Meas. Tech., 8, 3047-3057, https://doi.org/10.5194/amt-8-3047-2015