Long open path Fourier Transform Spectroscopy measurements of atmospheric greenhouse gases

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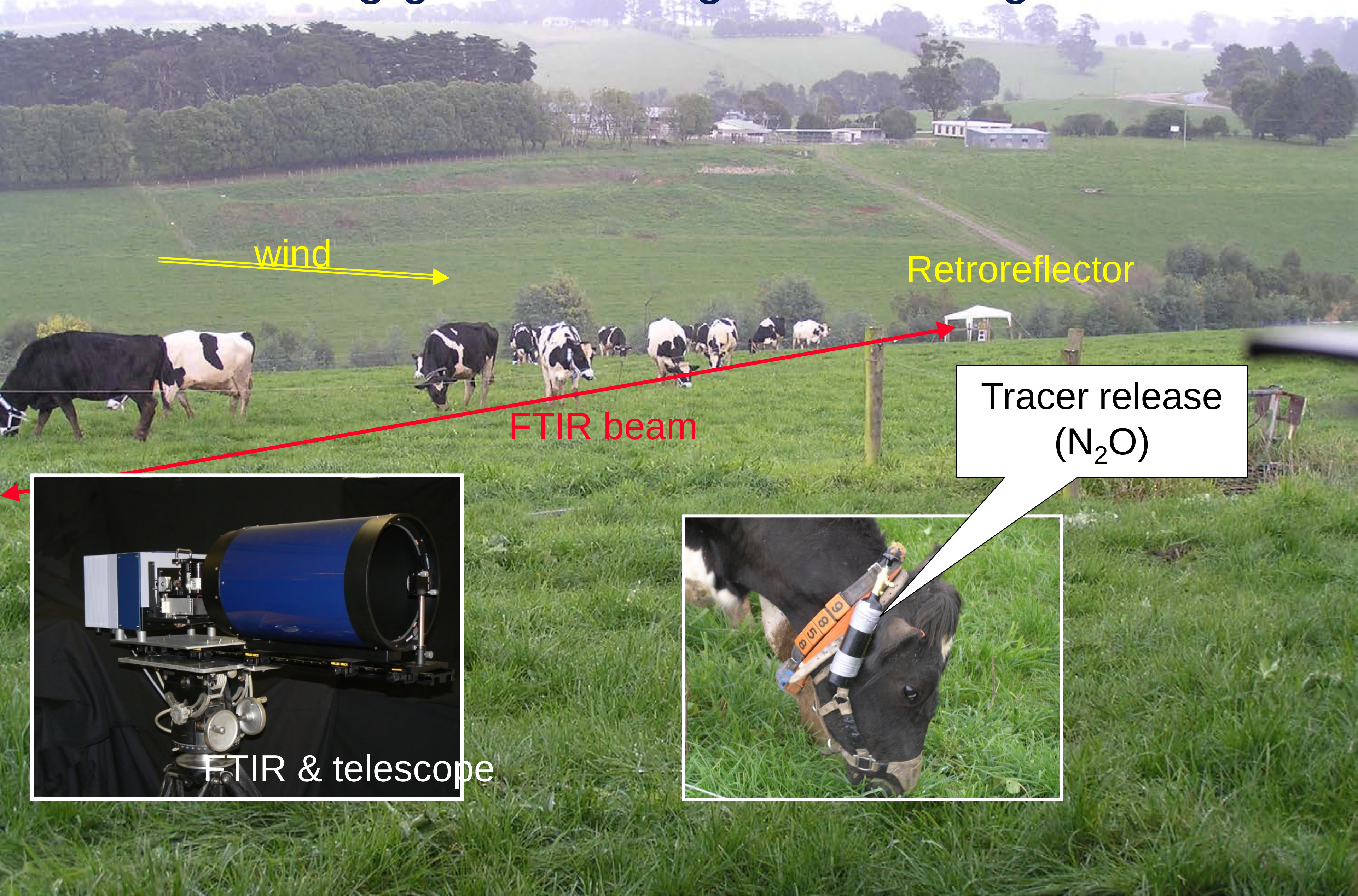
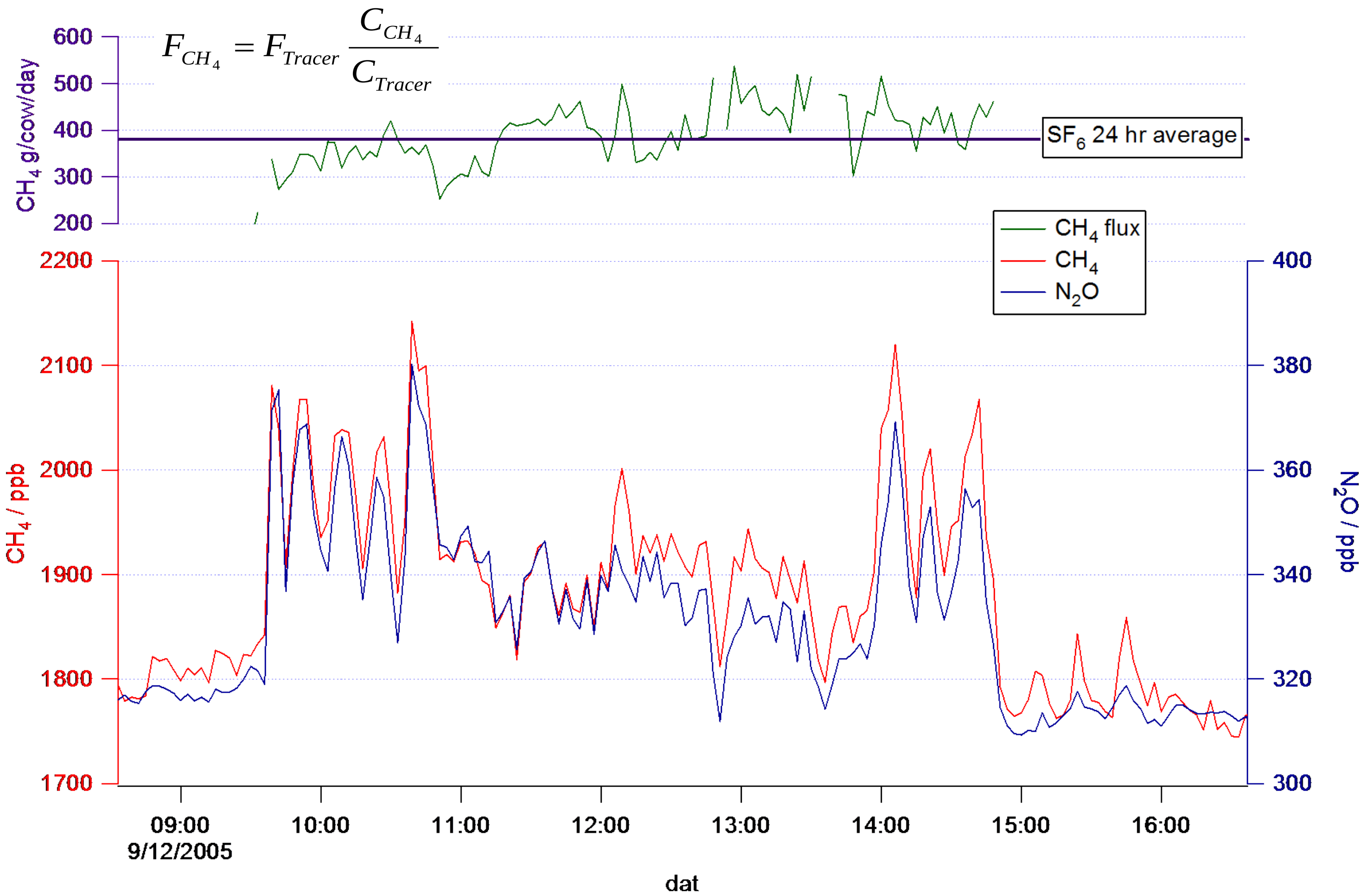
2nd ICOS Science Conference Helsinki, Sept 2016

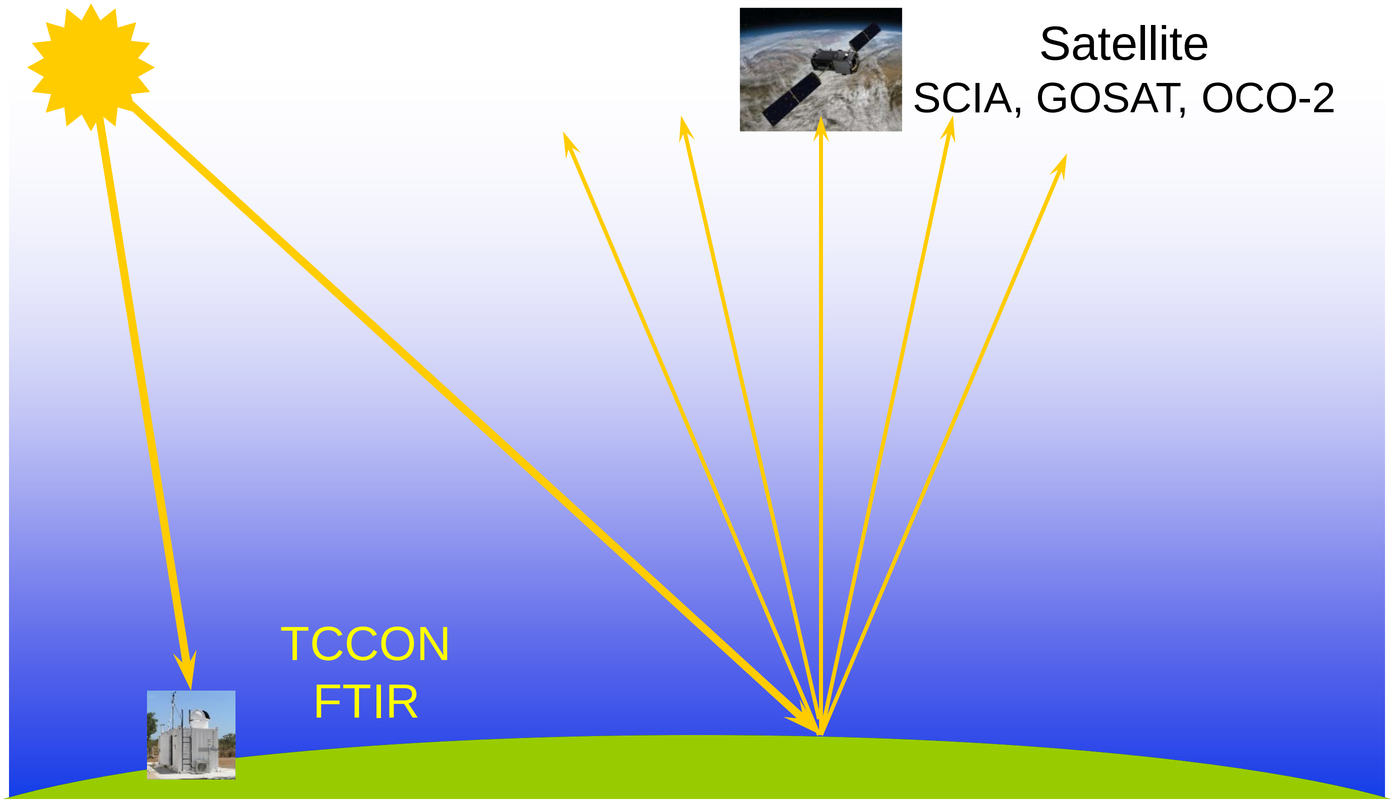
History 1: open path FTIR in the mid IR
Measuring greenhouse gases from agriculture



OP-FTIR-tracer: results



History 2: TCCON, GOSAT and OCO-2: Near IR remote sensing of CO_2, CH_4, N_2O, CO, H_2O

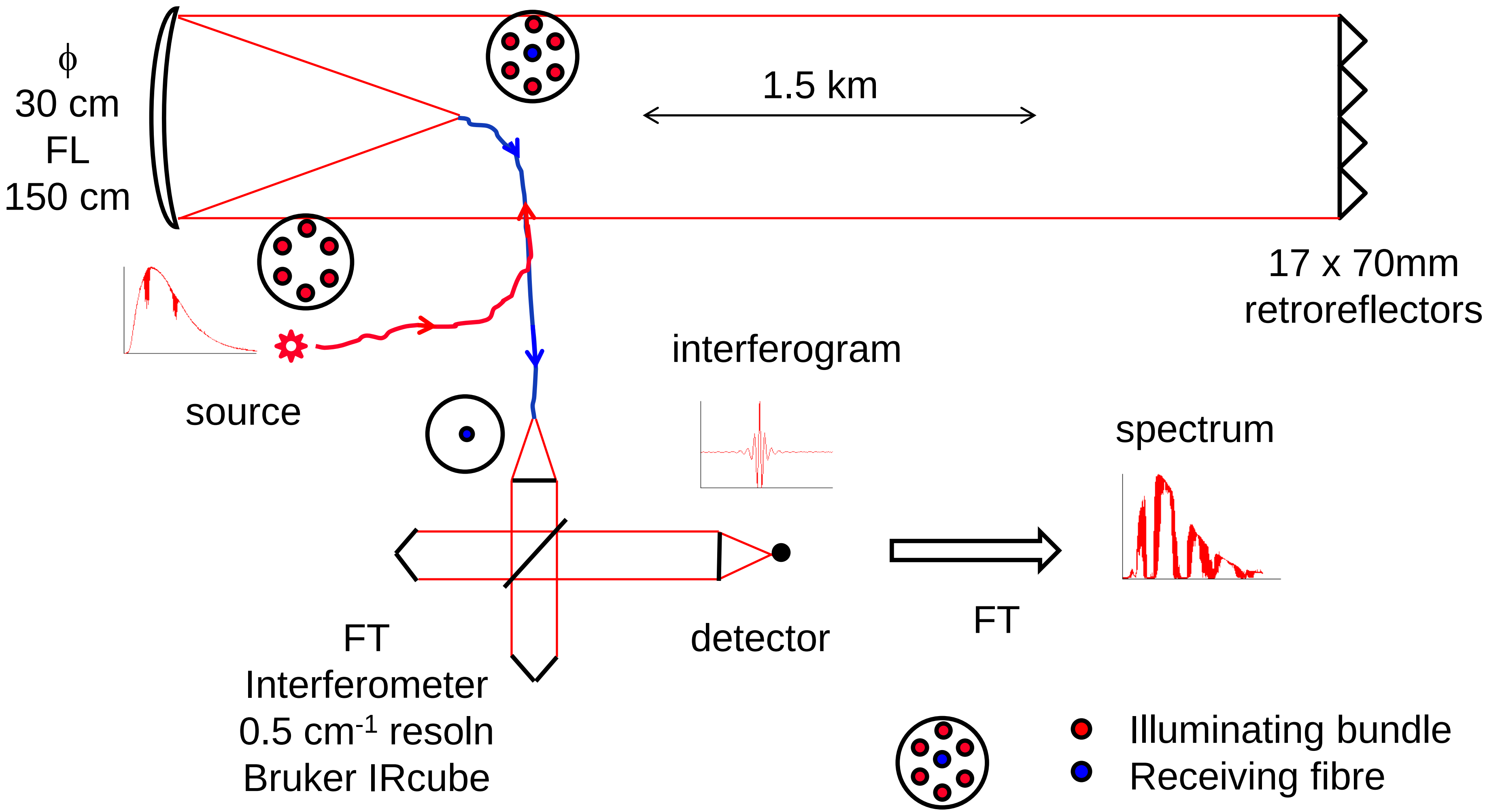


- Long path absorption spectroscopy in the near IR
- Sun as source, ~8-40 km absorption path
- TCCON precision/accuracy for CO_2 0.1 – 0.2% (0.4 – 0.8 ppm)

Open path FTIR meets DOAS in the NIR

- The most accurate greenhouse gas measurements are point-based
- Open path spectroscopy has some advantages
  - Spatial averaging
  - Better match to regional-scale model resolution
  - How spatially representative are point measurements?
  - How accurate are open path measurements?
- TCCON provides precise measurements of GHGs
  - Solar NIR absorption spectroscopy, 8-20 km-atm atmospheric path, high spectral resolution
  - Precision/accuracy 0.1 – 0.2% (0.4 – 0.8 ppm for CO_2)
- How well can we measure GHGs at the ground in an open path?
  - Low resolution portable FTS
  - Weaker source than the sun (50W quartz lamp)
  - 2-6 km path

Long path FTS setup




IUP
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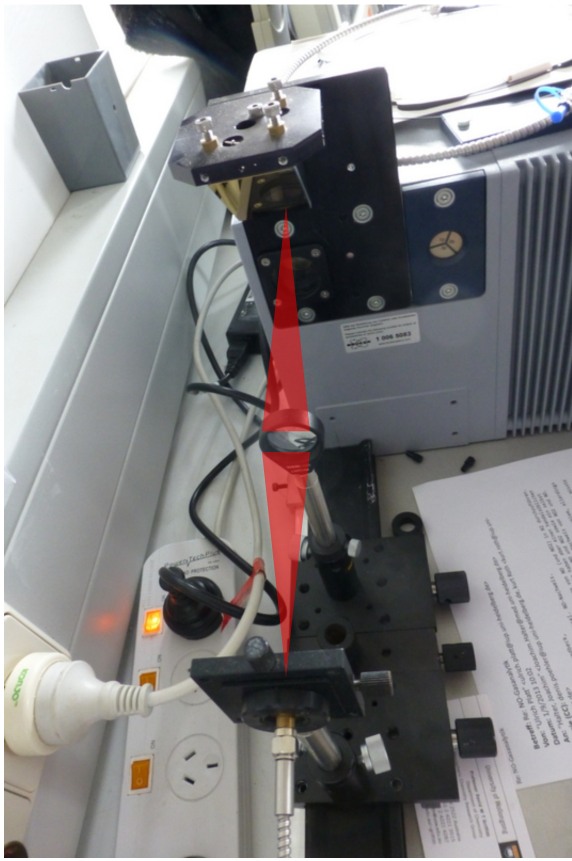
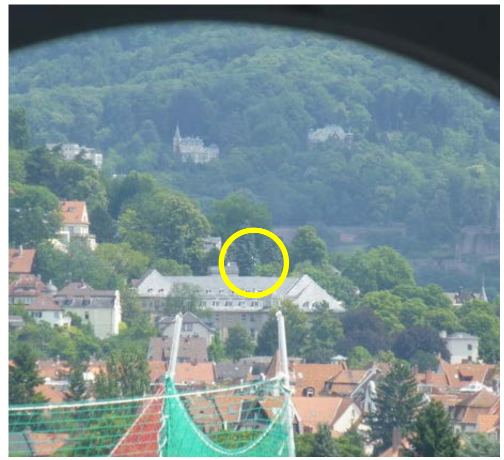
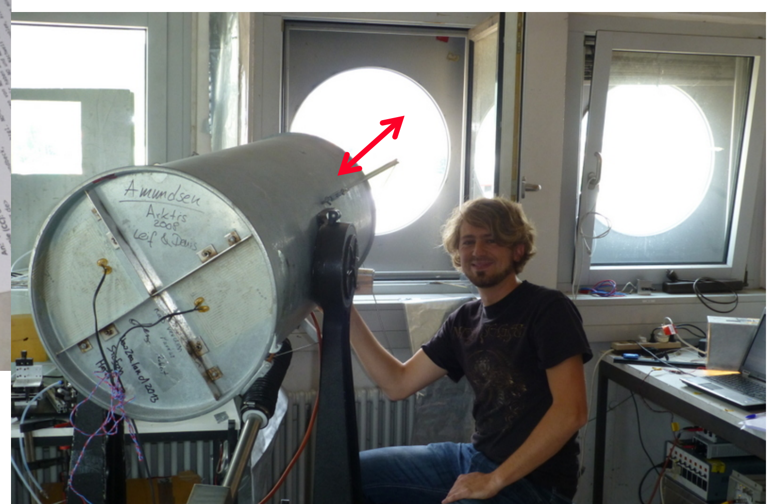
FTIR-DOAS setup

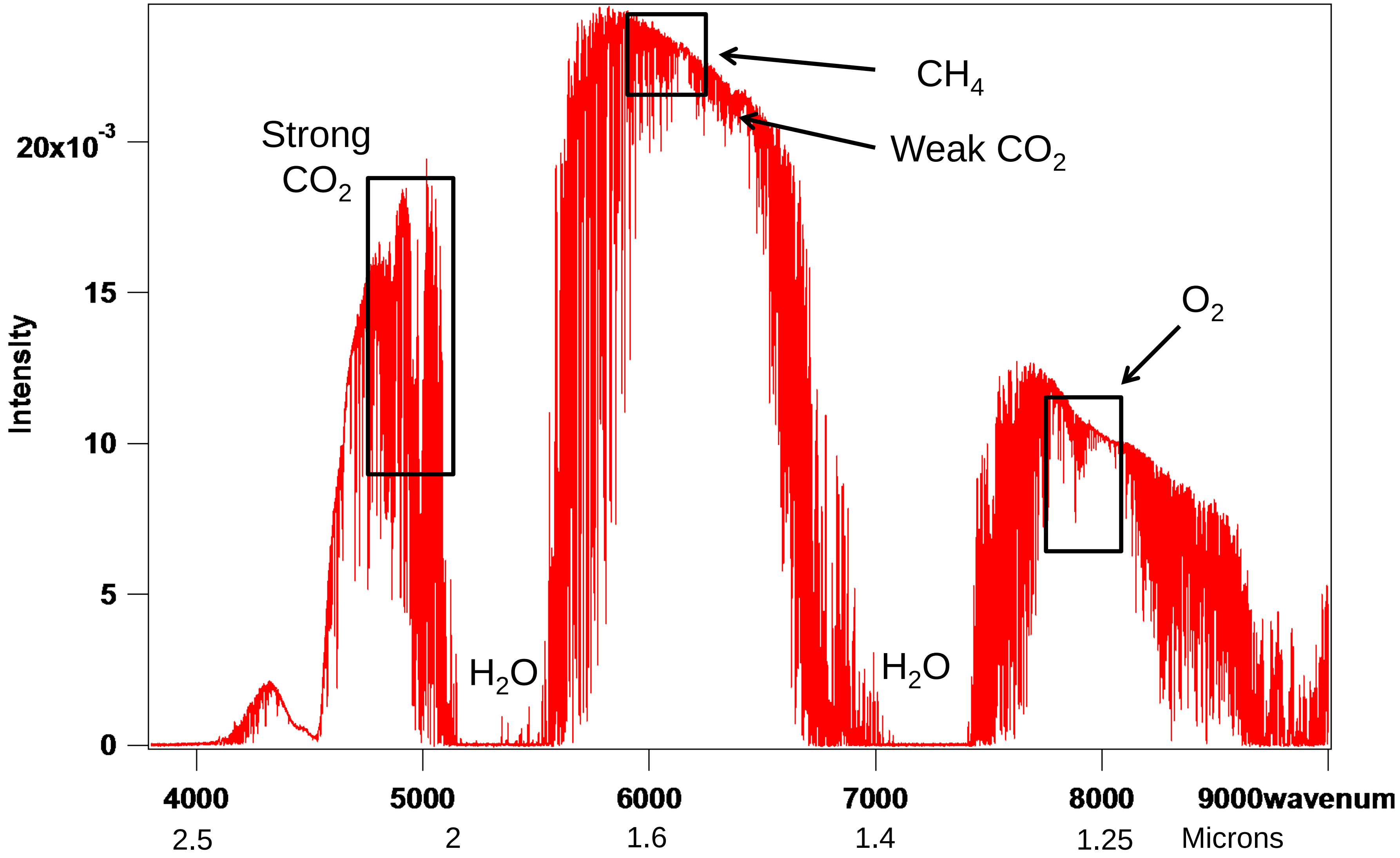




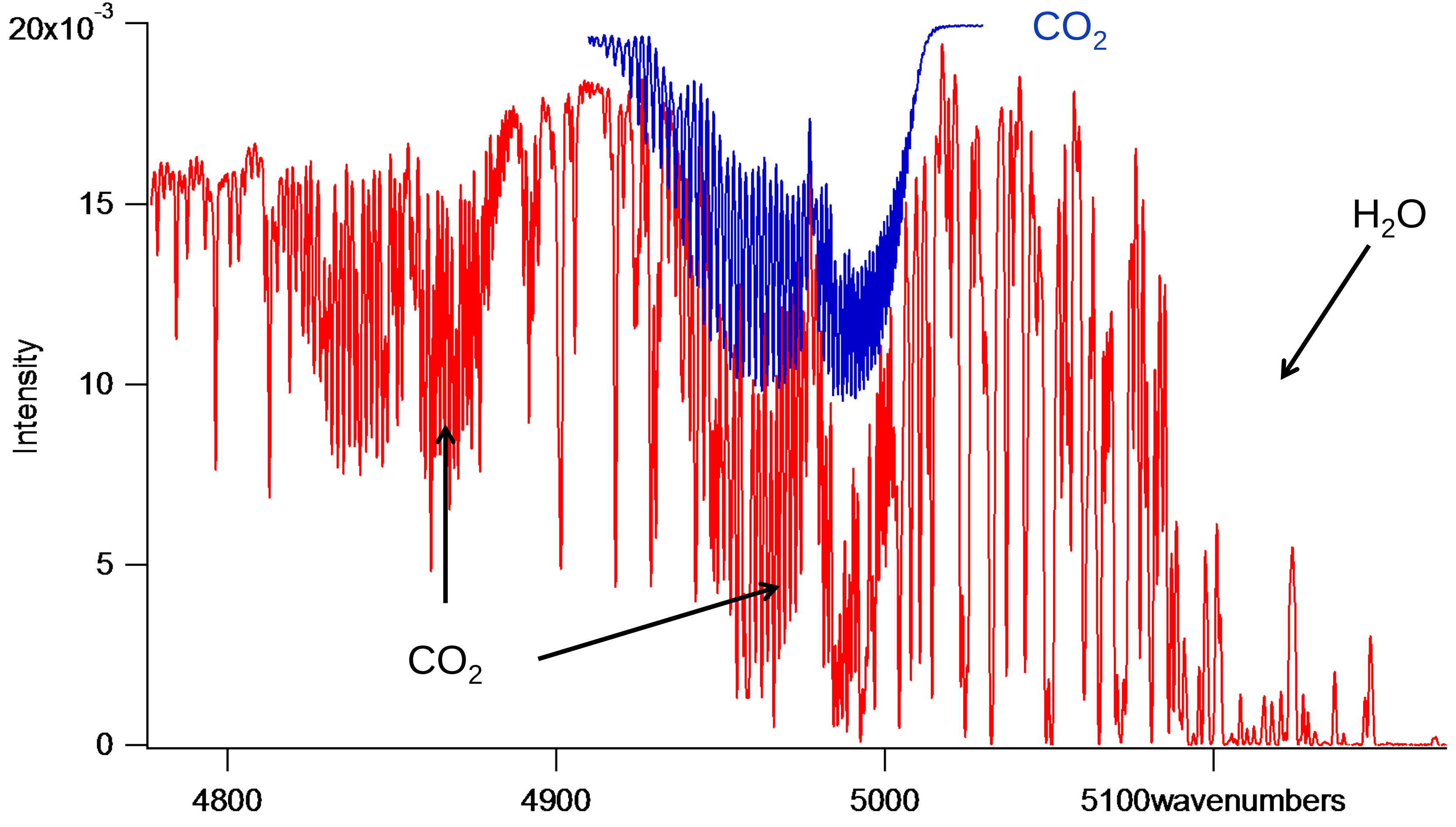


NIR long path spectrum

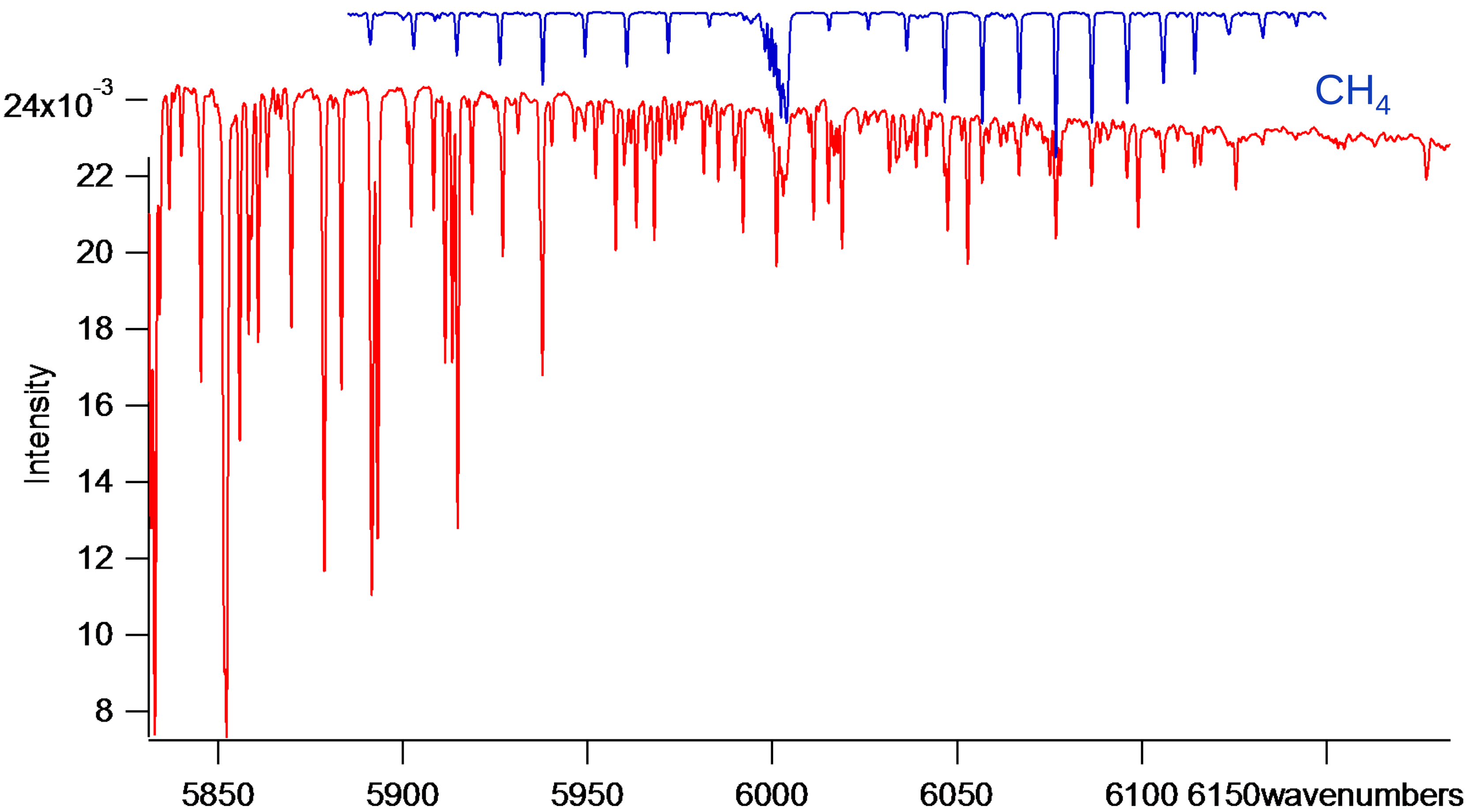
3.1 km return path IUP - Philosophen Weg



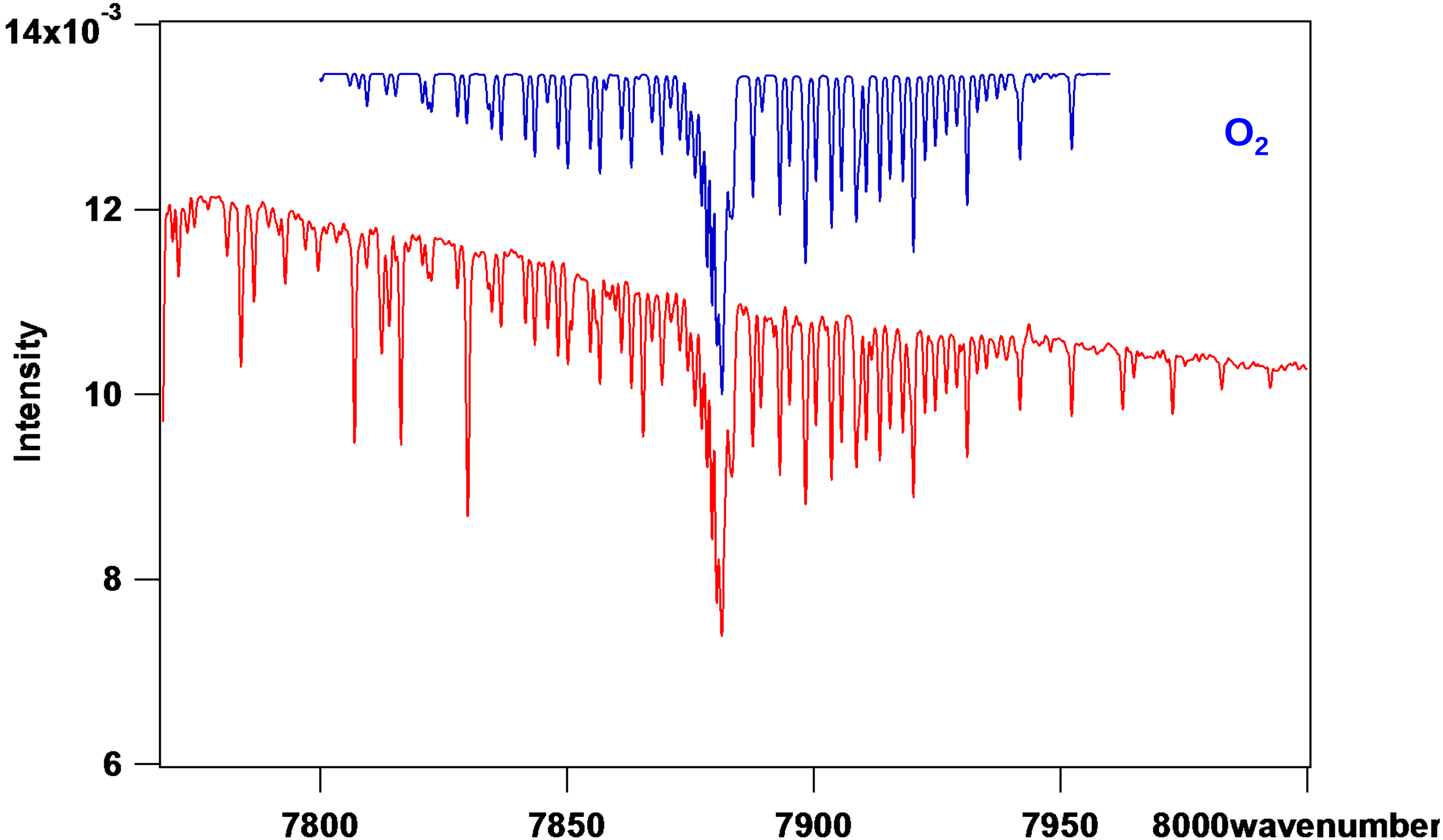
"Strong" CO_2 band 4900-5000 cm^{-1}



CH_4 is weaker…

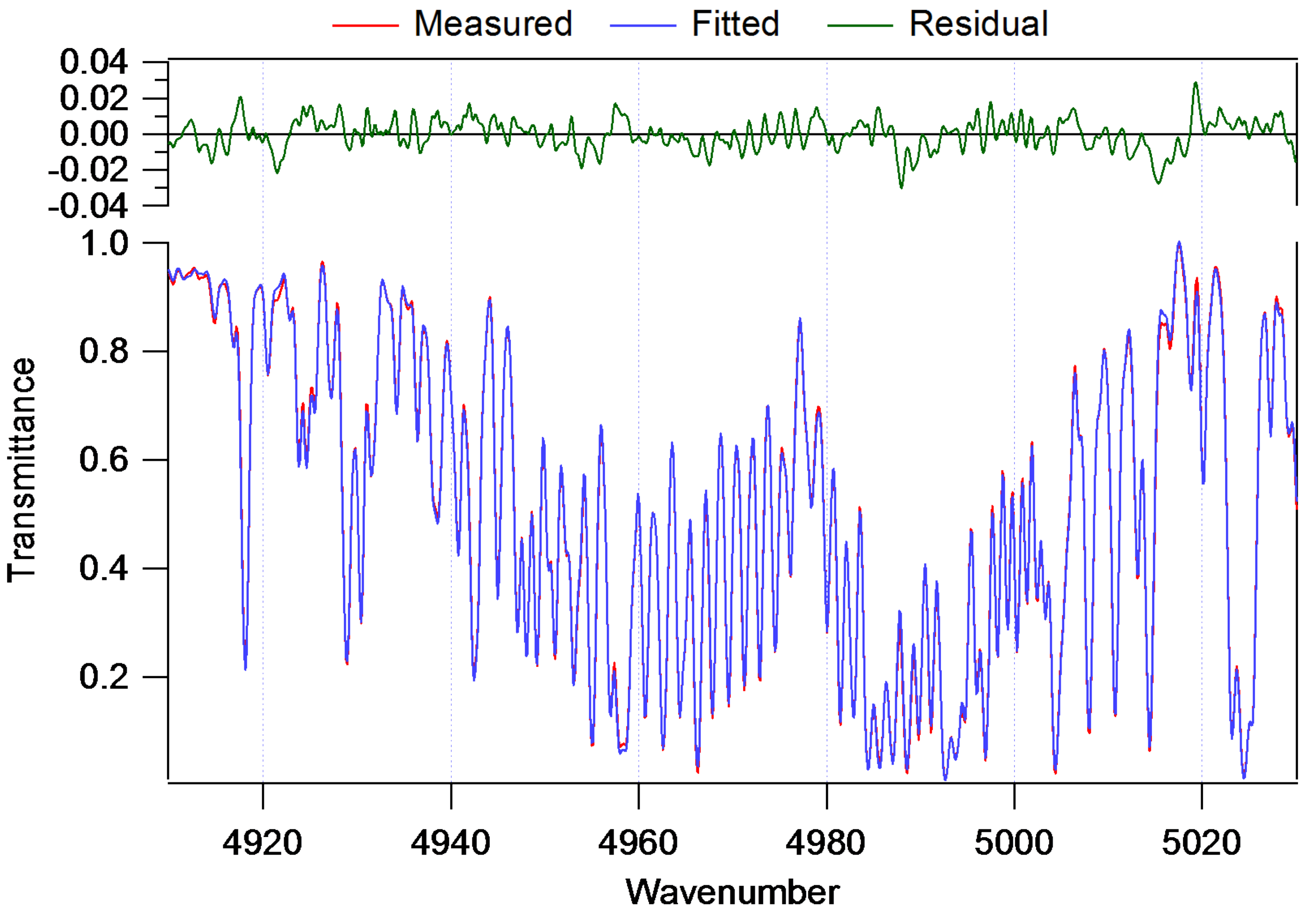


O_2

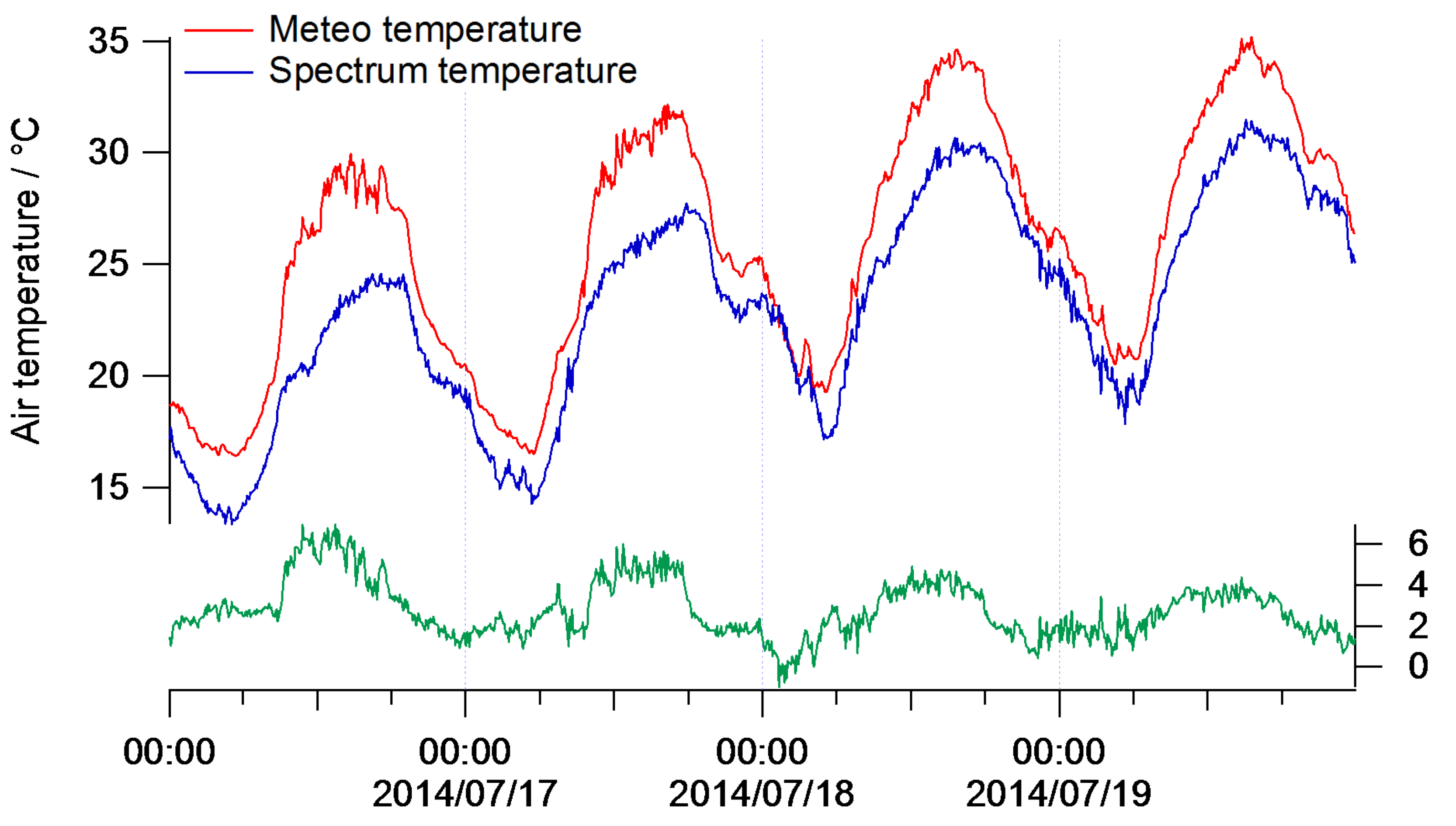

14x10^-3
12
10
8
6
Intensity
7800
7850
7900
7950
8000wavenumber
O2

Quantitative spectrum analysis

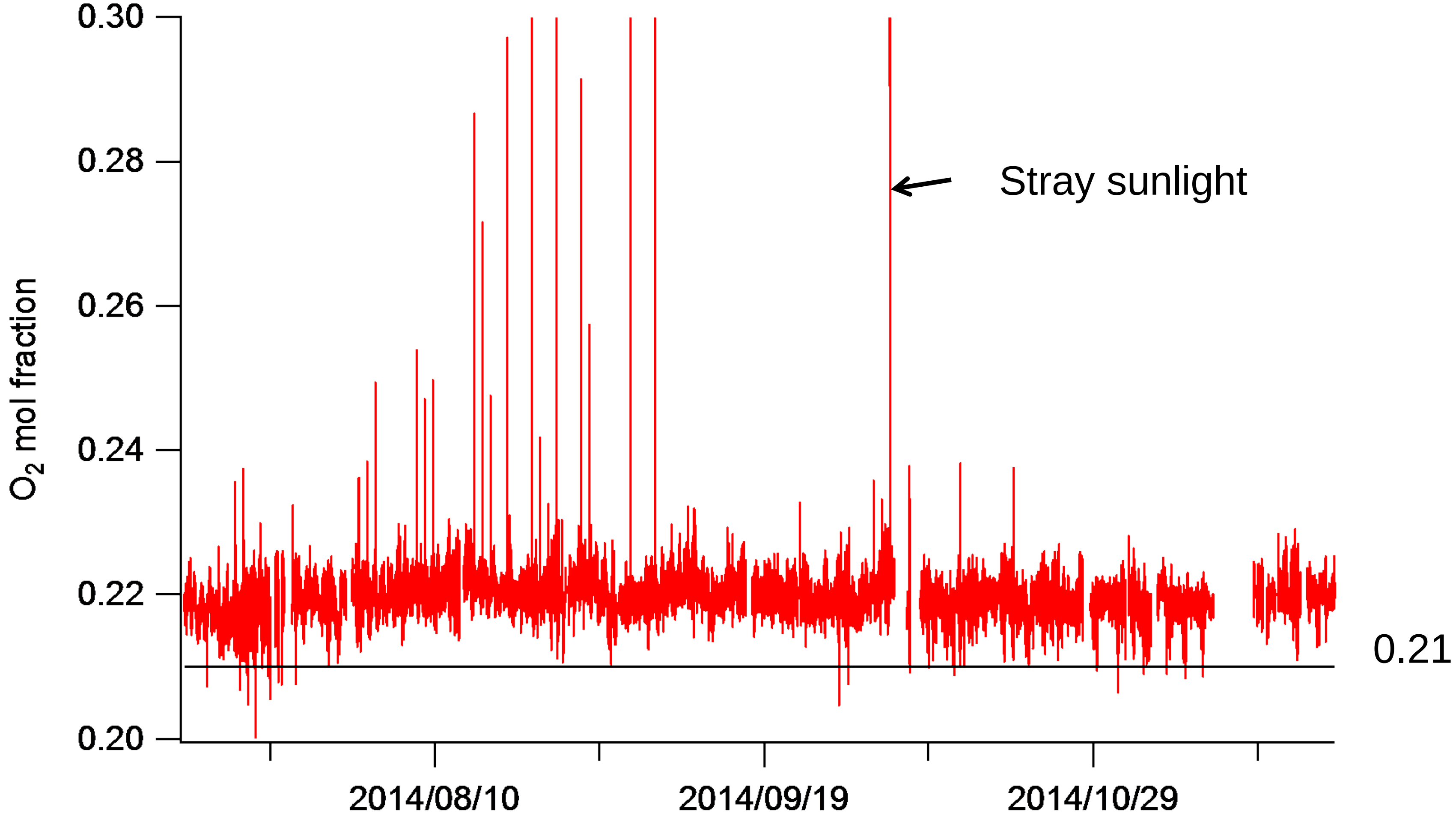
Least squares calculated fit: example CO_2



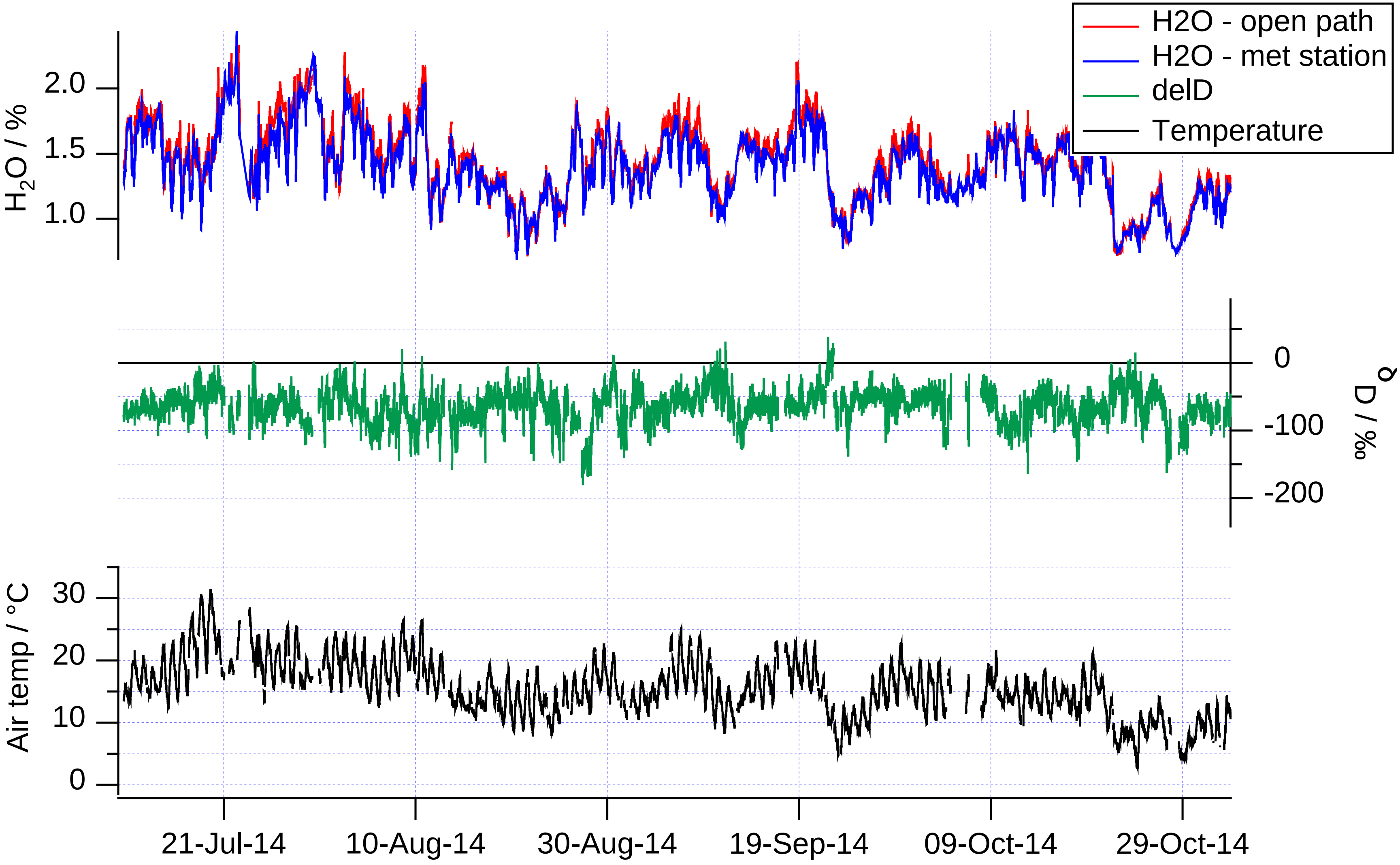
Temperatures
point vs path averaged



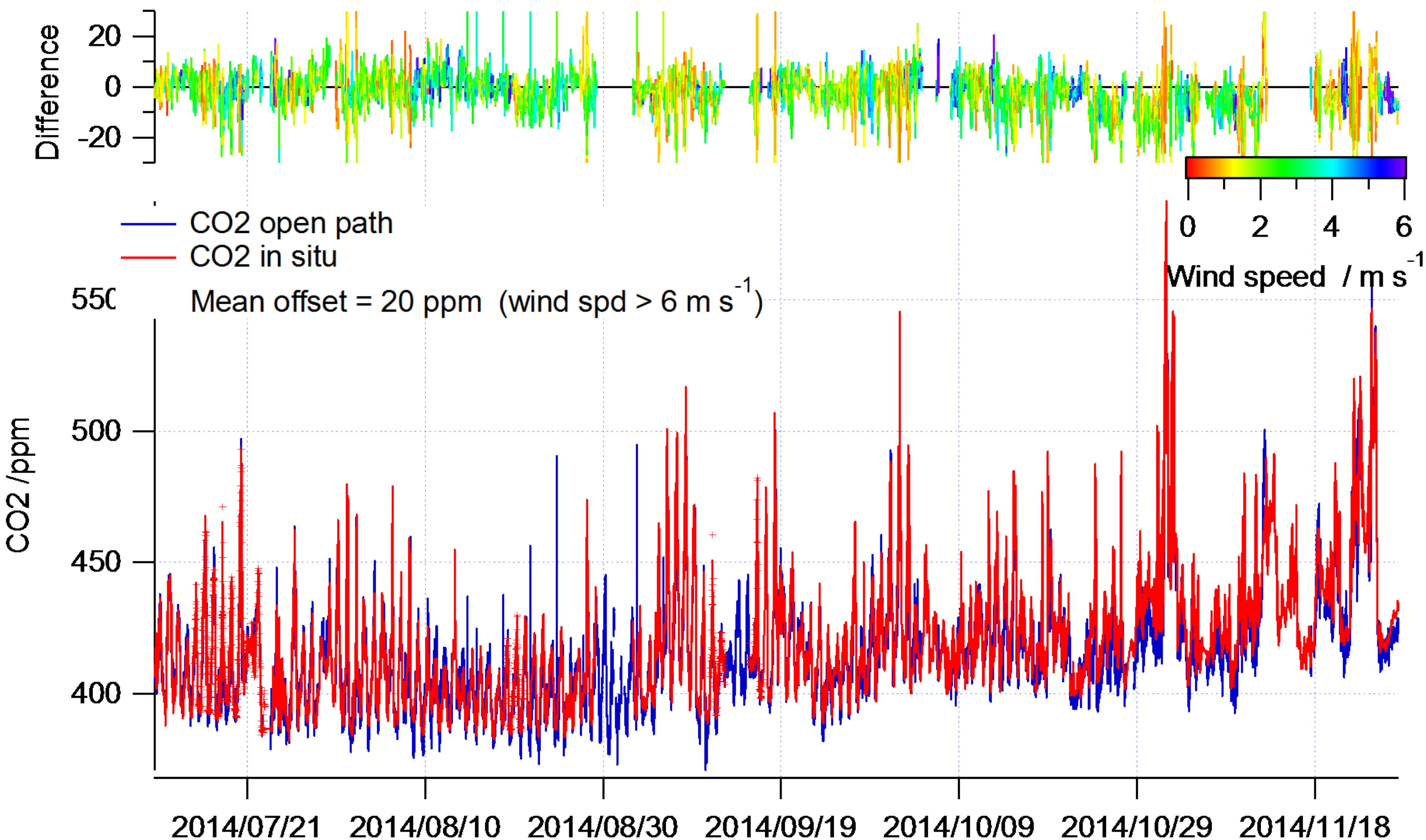
O_2 – reality check



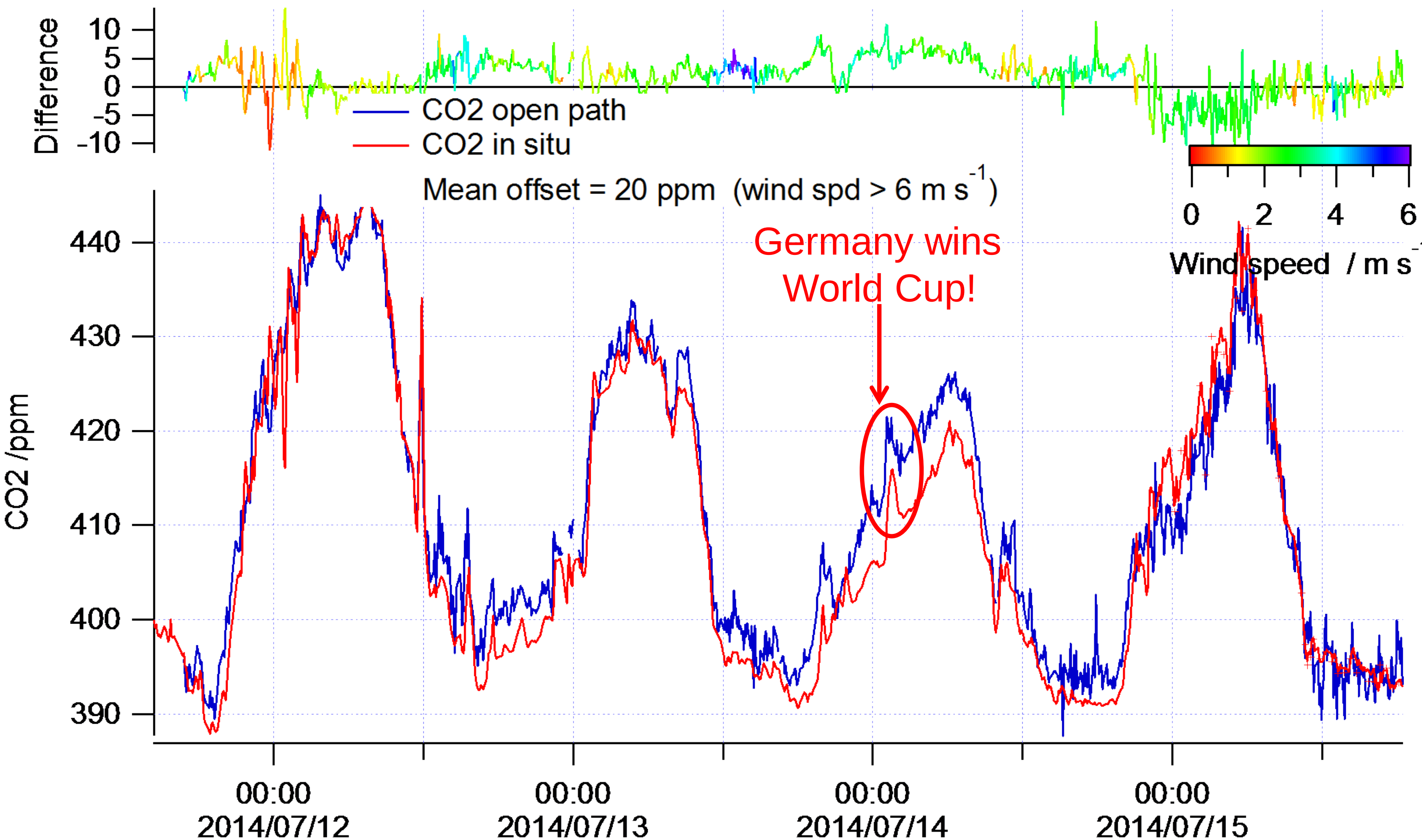
Water vapour and δD



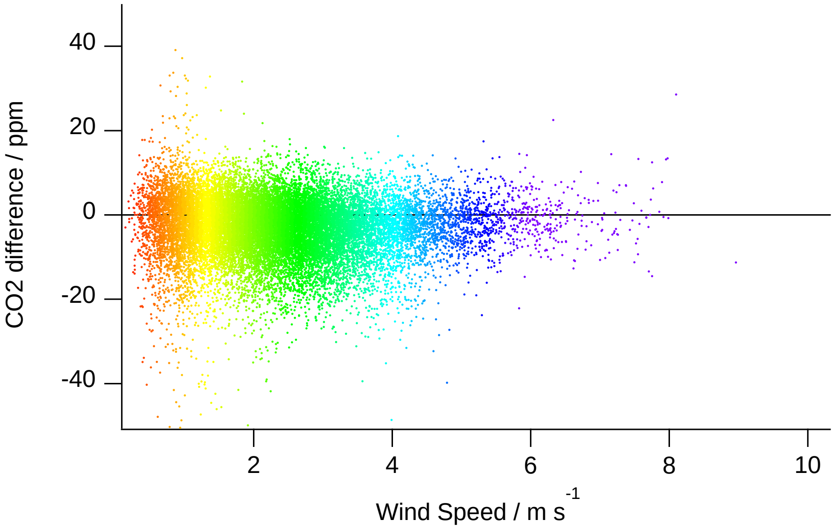
CO_2 July-October 2014



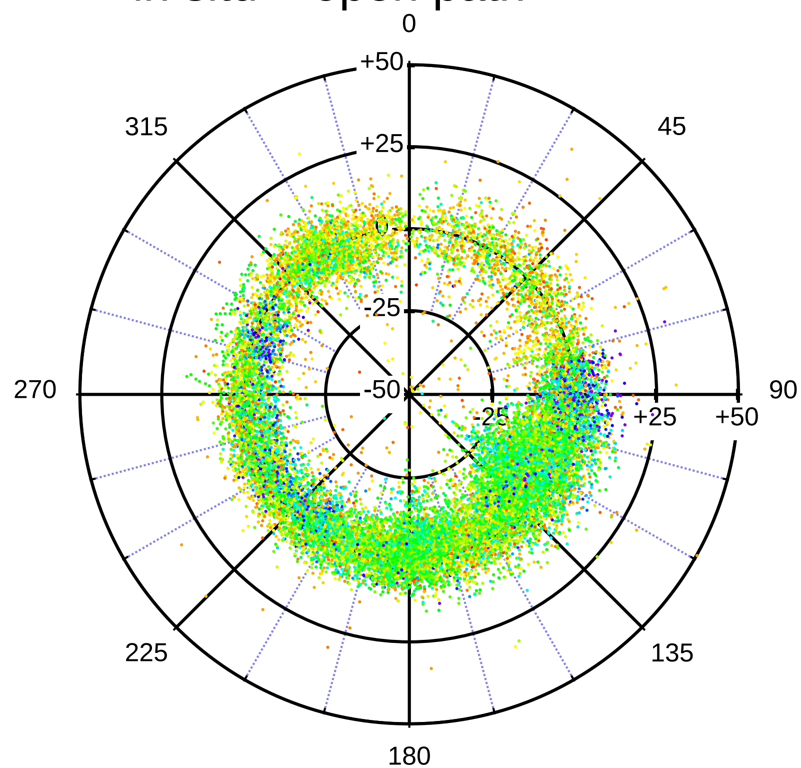
CO_2 – 3 days in July

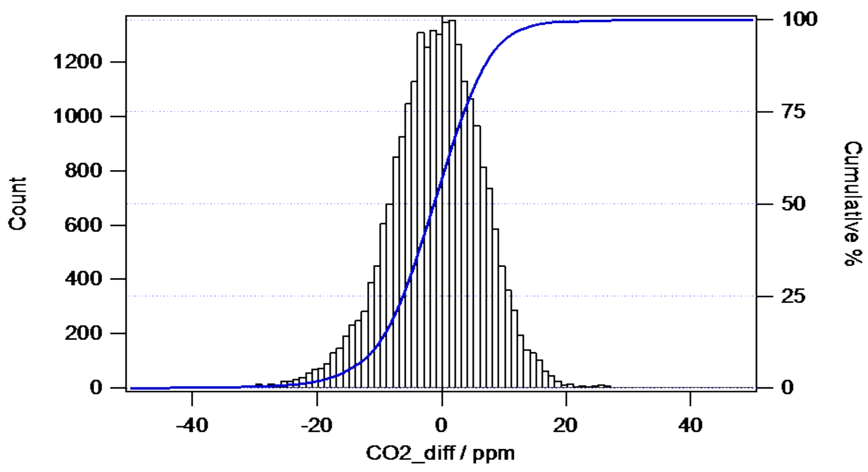


CO_2
open path – point in situ differences

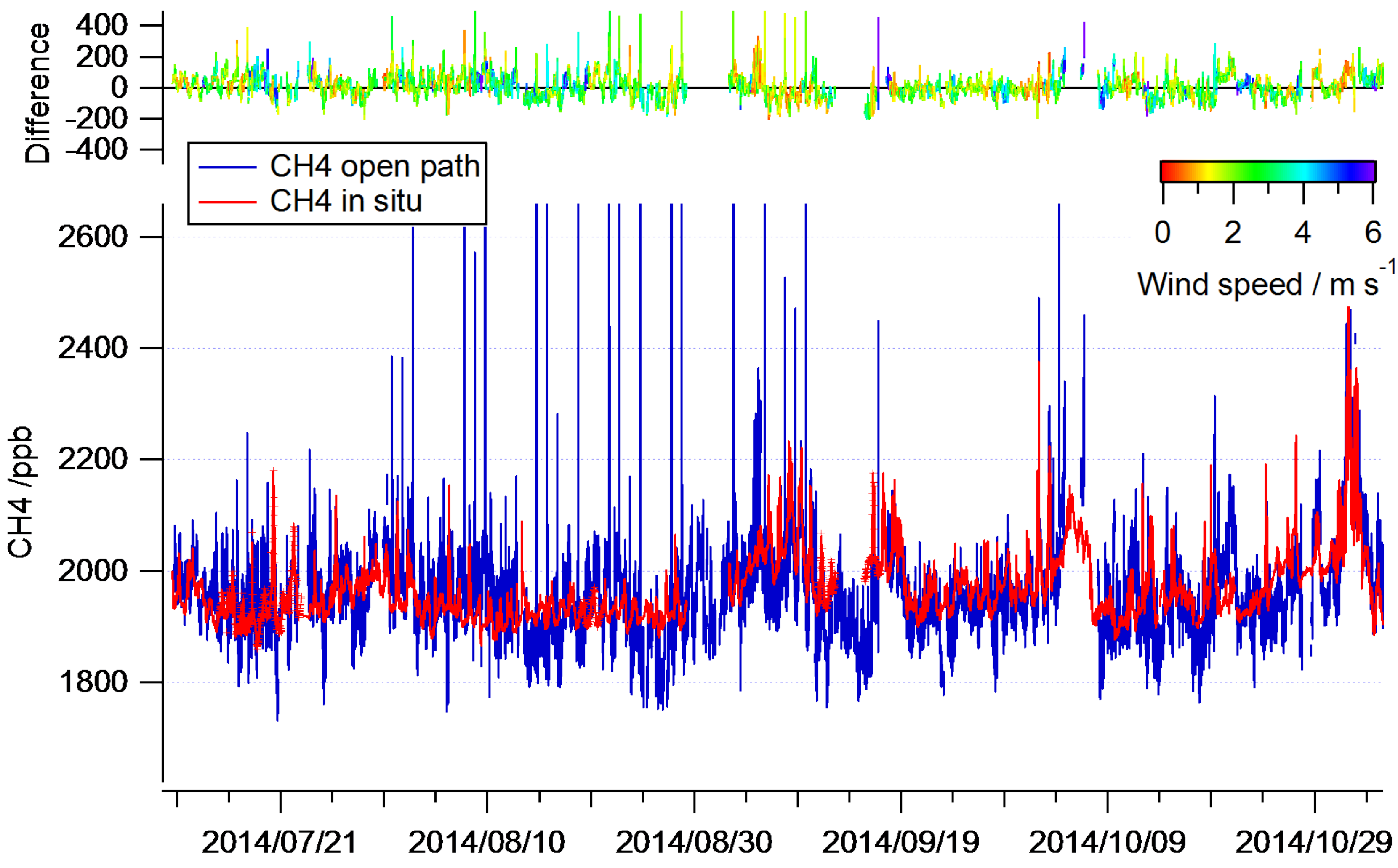


- "1σ" = 7ppm (n=21,800)
- Slight skew from SE (city)
  - in situ > open path

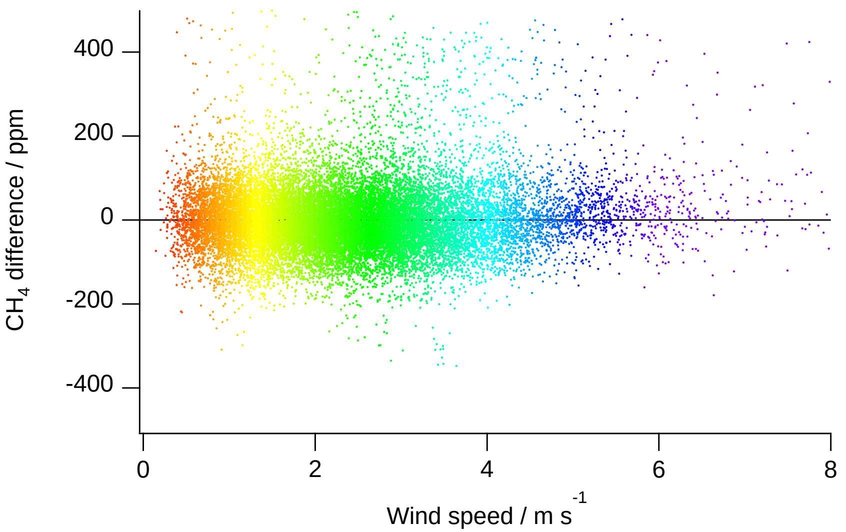


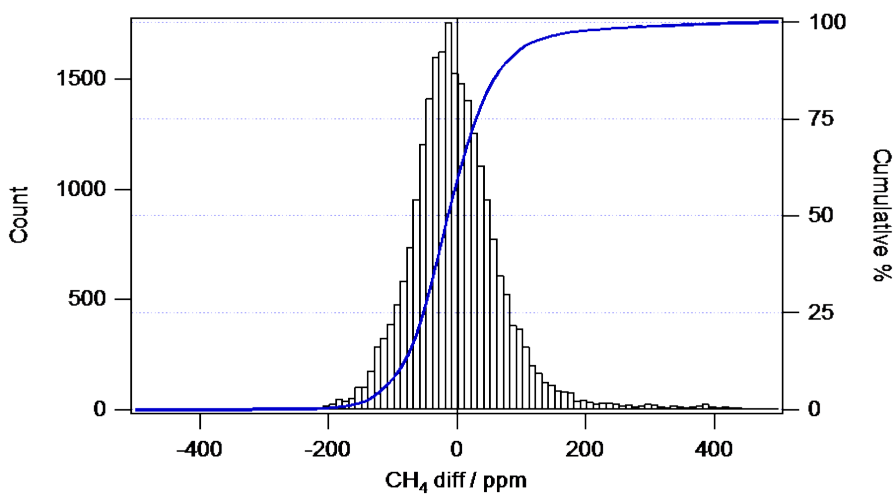


CH_4
July-October 2014

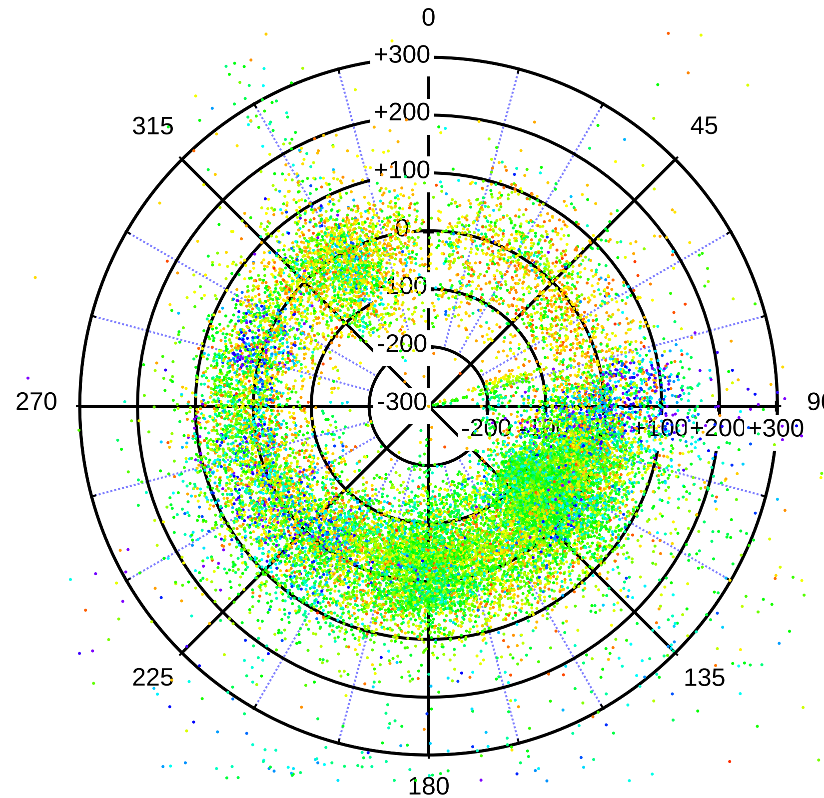


CH_4
open path – point in situ differences





- "1σ" = 93 ppb (n=21,800)



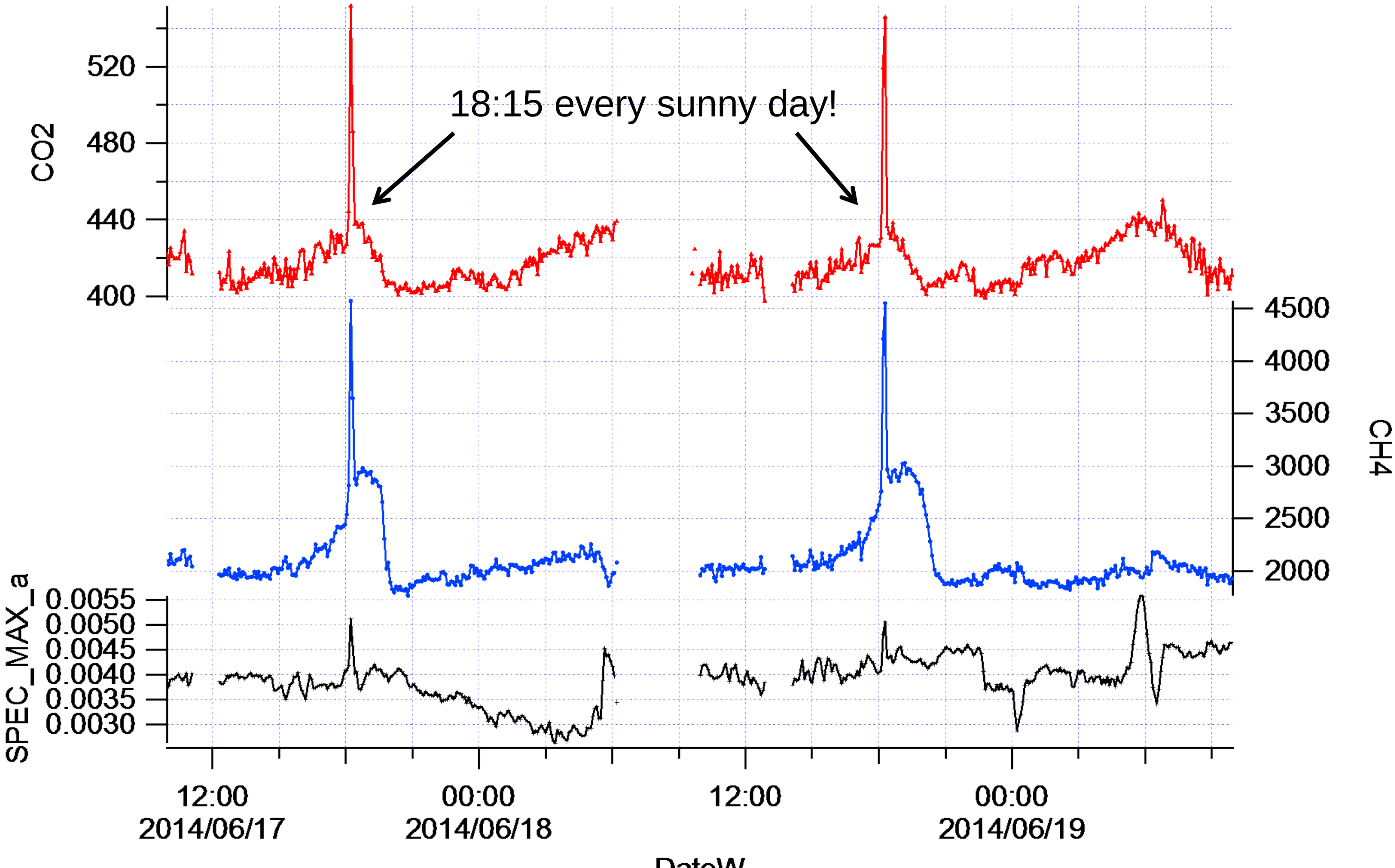
Precision and accuracy - summary

	CO_2	CH_4
OP-point difference (“1σ”) - all data	7 ppm	93 ppb
Short term repeatability (1σ) - estimated	3 ppm	37 ppb
Bias OP – point in situ	20 ppm	14 ppb

- Repeatability limited by
  - signal noise
  - optical throughput

- Accuracy/bias limited by
  - HITRAN/MALT ~ 2-4%
  - Temperature ~ 3°C (1%)
  - Pressure <1 hPa, (0.1%)
  - Pathlength <3 m (0.1%)
  - Fibre residual ~1%

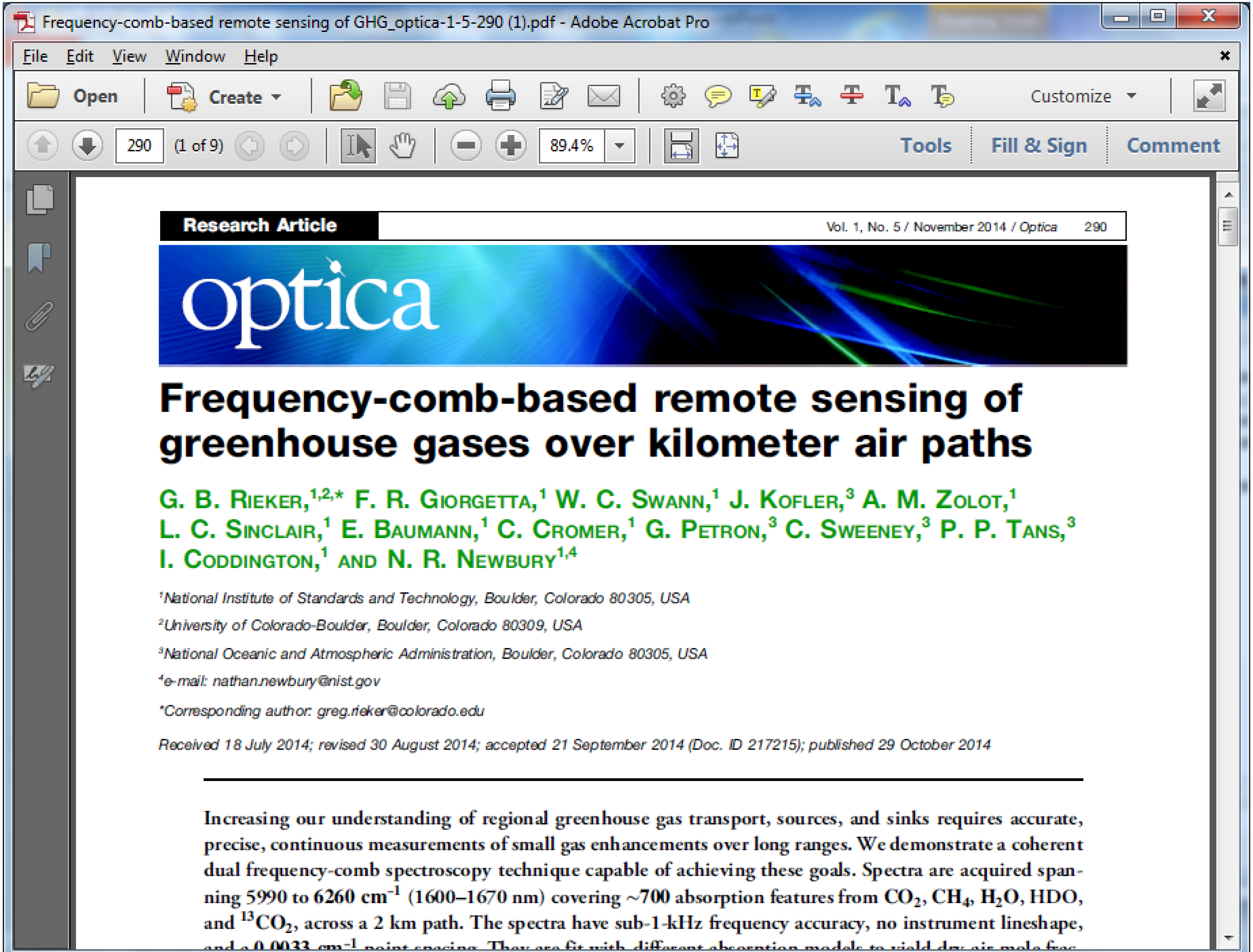
An interesting artefact …



Solar beam travels through ~17 km path
- Apparent increase in CO_2, CH_4, O_2

Potential improvements & future work

- More light!
  - Precision is detector noise limited
    - Brighter source
    - larger telescope/retro reflector area
- Pre-modulate IR source before transmission
  - Removes stray (sun)light artefacts
- Remove or co-fit fibre spectral structures
- Higher resolution?
  - Better discrimination against interferences, but…
  - Lower SNR => lower precision
  - Less portable



Research Article
Vol. 1, No. 5 / November 2014 / *Optica* 290

optica

Frequency-comb-based remote sensing of greenhouse gases over kilometer air paths

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Received 18 July 2014; revised 30 August 2014; accepted 21 September 2014 (Doc. ID 217215); published 29 October 2014

Increasing our understanding of regional greenhouse gas transport, sources, and sinks requires accurate, precise, continuous measurements of small gas enhancements over long ranges. We demonstrate a coherent dual frequency-comb spectroscopy technique capable of achieving these goals. Spectra are acquired spanning 5990 to 6260 cm^{-1} (1600–1670 nm) covering ~700 absorption features from CO_2, CH_4, H_2O, HDO, and $^{13}CO_2$, across a 2 km path. The spectra have sub-1-kHz frequency accuracy, no instrument lineshape,


Thank you !