Long open path Fourier transform spectroscopy measurements of greenhouse gases in the Near-IR

Open path FTIR meets DOAS

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History: open path FTIR – mid IR
Measuring greenhouse gases from agriculture

- Tracer release (N$_2$O)
- FTIR beam
- Retroreflector
- FTIR & telescope
- Wind
$F_{CH_4} = F_{Tracer} \frac{C_{CH_4}}{C_{Tracer}}$
TCCON and OCO-2: Near IR remote sensing of CO$_2$, CH$_4$, N$_2$O, CO, H$_2$O

- Infrared absorption spectroscopy in the near IR
- Retrieve total column amounts of trace gases
- Derive column average dry air mole fractions (e.g. $X_{CO_2}$)
- TCCON precision/accuracy 0.1 – 0.2% (0.4 – 0.8 ppm)
Open path FTIR meets DOAS in the NIR

- Accurate greenhouse gas measurements are mostly point-based
  - Using calibrated in situ analysers or flask samples
- Open path spectroscopy provides spatial averaging
  - Better match to regional-scale model resolution
  - How spatially representative are point measurements?
  - How accurate are open path measurements?
- We know that TCCON provides precise measurements of GHGs:
  - Solar absorption spectroscopy, 8-20 km-atm atmospheric path
  - TCCON precision/accuracy 0.1 – 0.2% (0.4 – 0.8 ppm)
- 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

- Source
- FT Interferometer (0.5 cm\(^{-1}\) resoln, Bruker IRcube)
- FT Detector
- 1.5 km path
- Retroreflectors

丈量参数:
- φ (直径): 30 cm
- FL (焦距): 150 cm

光路参数:
- 17 x 70 mm
- Illuminating bundle
- Receiving fibre
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…
Spectrum analysis: MALT least squares fit CO$_2$
Temperatures
point vs path averaged

Meteo temperature
Spectrum temperature

Air temperature / °C

00:00 00:00 00:00 00:00
2014/07/17 2014/07/18 2014/07/19
$O_2$ – reality check

Irradiance

$O_2$ mol fraction

2014/08/10 2014/09/19 2014/10/29

0.20 0.22 0.24 0.26 0.28 0.30

Stray sunlight

0.21
CO₂ – 3 days in July

Germany wins World Cup!

Mean offset = 20 ppm (wind spd > 6 m s⁻¹)
CO$_2$  July-October 2014

Mean offset = 20 ppm (wind spd > 6 m s$^{-1}$)
CO$_2$
open path - in situ differences

![Graph showing CO$_2$ open path - in situ differences](image-url)
CO₂
open path - in situ differences

Wind speed / m s⁻¹

|→ mean diff=0 wind spd > 6 m s⁻¹

Low bias (~ 2ppm, OP < in situ)

p-p noise 10 ppm

City centre

Wind direction / °
Summary: CO$_2$ precision and accuracy

**Precision/repeatability**
- ~1-2 ppm ($1\sigma$) for 5 min averages
  - limited mainly by spectrum SNR
  - 300mm telescope, 17 retros, 50 W source
- ~ <40 ppm p-p differences to in situ measurements
  - Real differences, larger at low wind speeds

**Accuracy/bias**
- ~ 20 ppm if uncalibrated (5%)
  - Limited by
    - HITRAN, simulation ~ 2-4%
    - Temperature ~ 3°C (1%)
    - Pressure <1 hPa, (0.1%)
    - Pathlength <3 m (0.1%)
    - Fibre residual ~1%
An interesting artefact …

18:15 every sunny day!
Solar beam travels through ~17 km path
- Apparent increase in CO₂, CH₄, O₂
Current setup

- **Source**: 30 cm
- **FL**: 150 cm
- **φ**: 30 cm
- **FT Interferometer**: 0.5 cm⁻¹ resolution
- **Bruker IRcube**: 1-3 km
- **Illuminating bundle**
- **Receiving fibre**
- **17 x 70mm retroreflectors**

Diagram:
- Source
- FT Interferometer (0.5 cm⁻¹ resolution, Bruker IRcube)
- Illuminating bundle
- Receiving fibre
- 1-3 km
- Retroreflectors

Spectrum
Stray and scattered radiation (e.g., sunlight) is not modulated, appears only as DC and does not appear in the FT spectrum.
Potential improvements & future work

- Pre-modulate IR source before transmission
  - Removes stray (sun)light
- More photons
  - Precision is detector noise limited
- Remove or co-fit fibre spectral structures
- Higher resolution?
  - Better discrimination against fibre residuals
  - Lower SNR => lower precision
  - Less portable
- Temperature measurement
  - Improve retrieval
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 CO₂, CH₄, H₂O, HDO, and ¹³CO₂, across a 2 km path. The spectra have sub-1-kHz frequency accuracy, no instrument lineshape, and a 0.0033 cm⁻¹ point spacing. They are fit with different absorption models to yield dry-air mole-fraction...
Thank you!