Quantum-cascade laser absorption spectrometer (QCLAS) for water vapor measurements in the upper atmosphere

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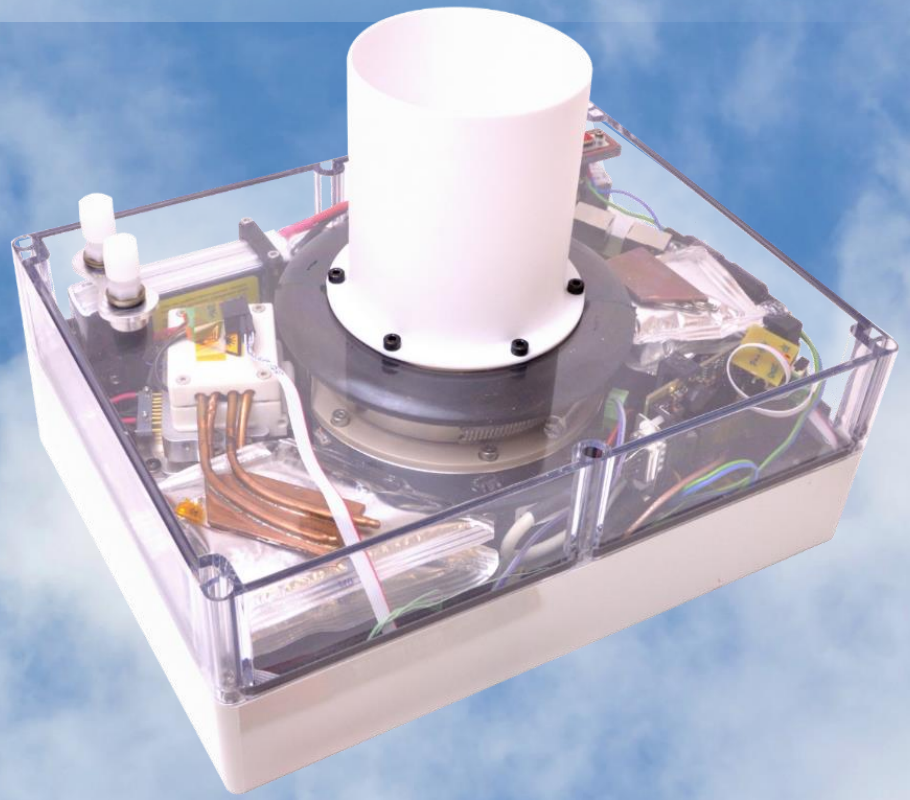
GCOS Project "ALBATROSS"



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Outline

1. Motivation
2. QCLAS instrument description
3. Test flights (2019)
4. Laboratory validation campaign (2021)
   - Precision
   - Accuracy
5. Conclusions and outlook

Balloon-borne measurements of stratospheric H_2O

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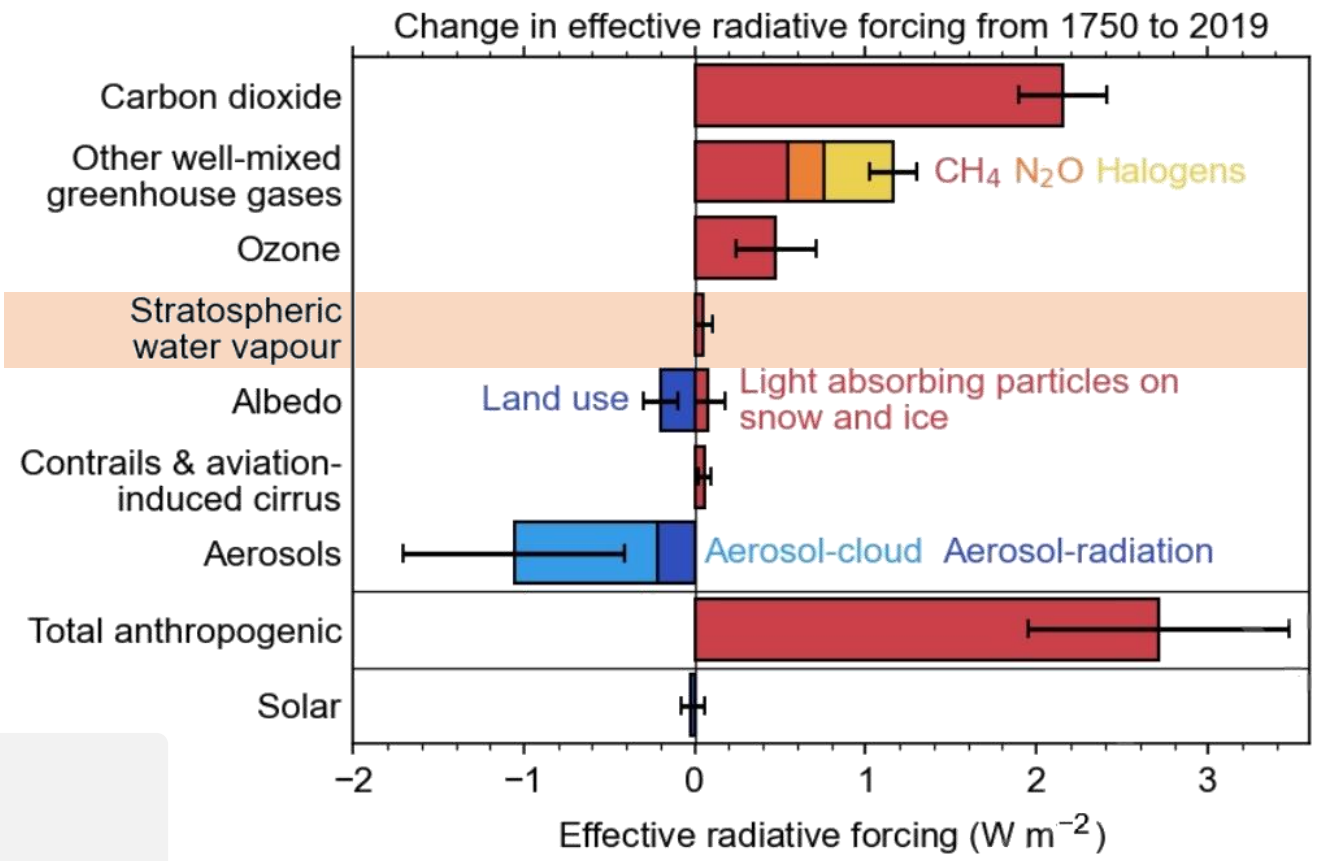
Climate relevance

- Water vapour (H_2O) is the strongest greenhouse gas in the Earth's atmosphere
- Microphysical processes at the **UTLS** determine the H_2O content of the stratosphere

In-situ techniques

- Capacitive sensors (radiosondes)
- Frostpoint hygrometry (CFH, FPH, PCFH, …)
- Lyman-α fluorescence (FLASH-B)
- **Laser spectroscopy** (Pico-Light H_2O, QCLAS)



IPCC AR6, **2021**

Challenges

- Phasing-out of R23 (CFH/FPH)
- Significant discrepancies found between different techniques (e.g. AquaVIT-1) → Accuracy?

Quantum-cascade laser absorption spectrometer (QCLAS)

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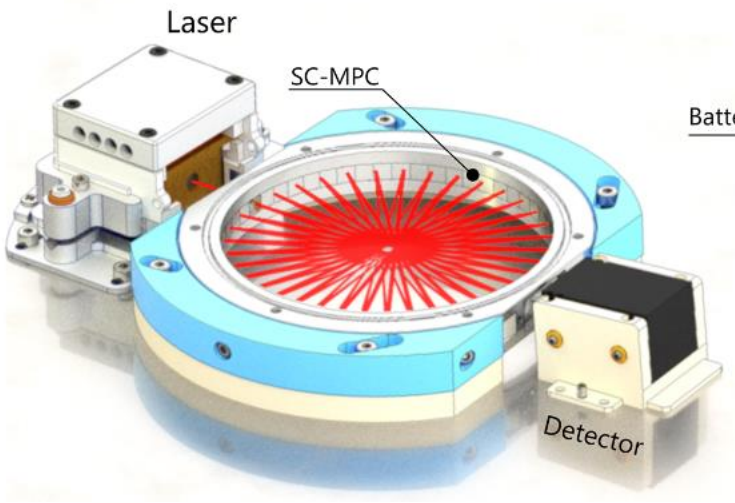
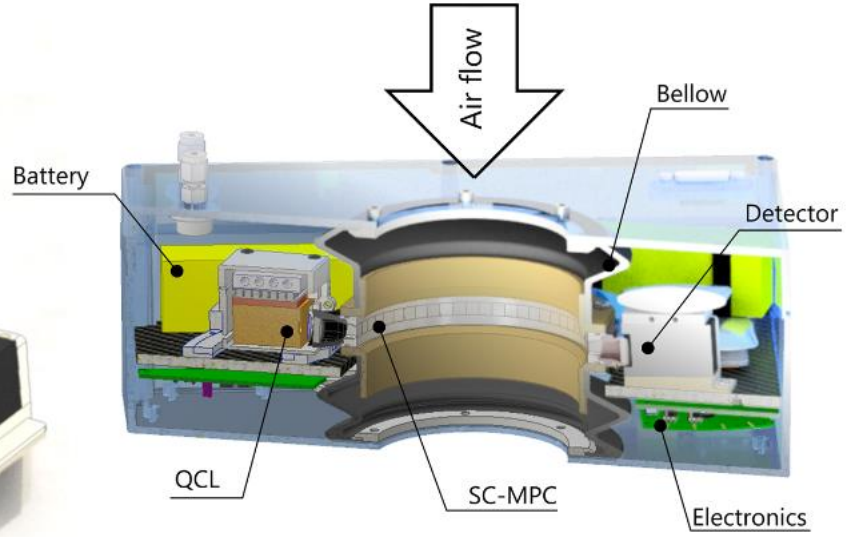
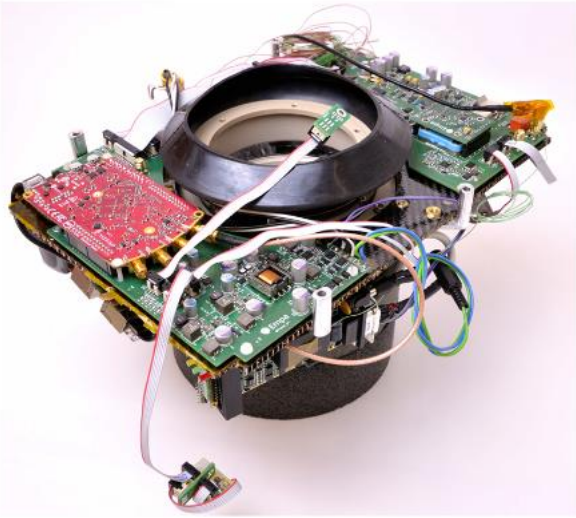
Main components

- Segmented circular multipass cell (SC-MPC)
- Quantum-cascade laser (QCL)
- Mid-IR detector (MCT)
- Thermal management system (TEC & PCM)





- Compact and robust optical design
- Lightweight (< 4 kg)
- High-precision and accuracy
- Fast response (1 Hz)
- Calibration-free retrieval



Graf *et al.*, AMT, **2021**

Quantum-cascade laser absorption spectrometer (QCLAS)

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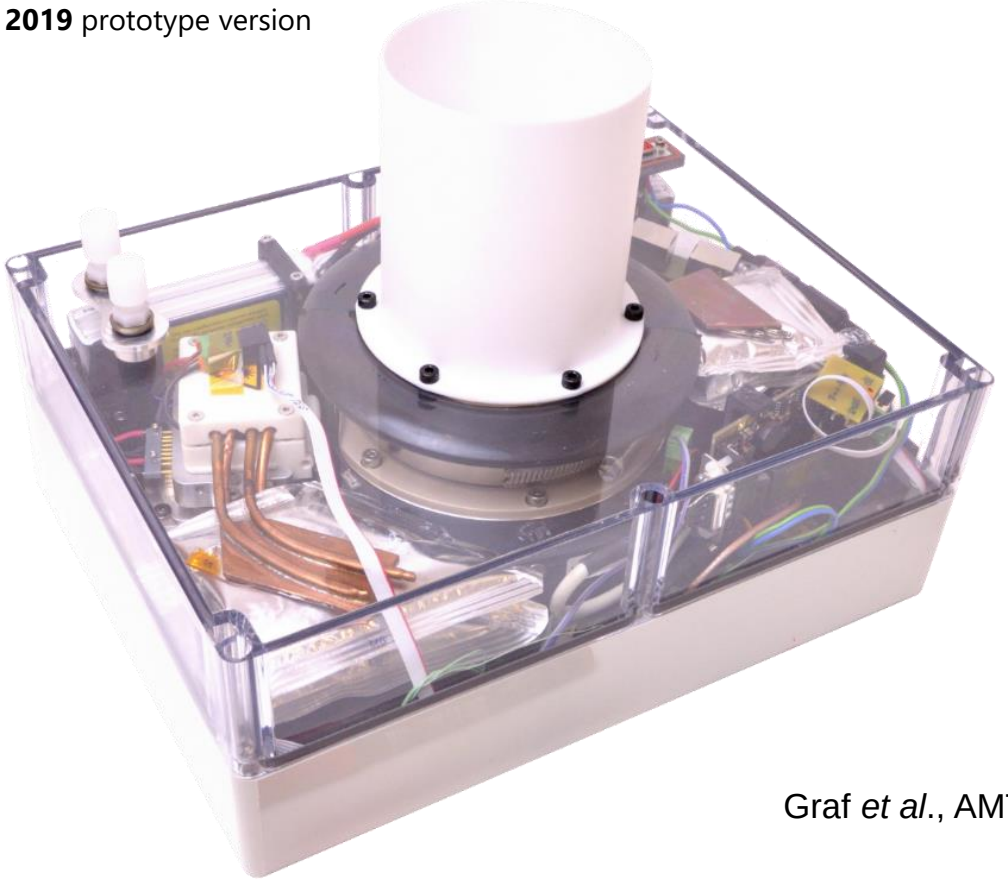
Technical properties

Method	QCL direct absorption spectroscopy
Sampling technique	Open-path
SC-MPC diameter	10.8 cm
Optical path length	6 m
H_2O transition used	1662.809 cm^{-1} ($\lambda \approx 6$ µm)
QCL tuning range	~ 1 cm^{-1} (icw driving)
Acquisition rate	1 Hz
Acquisition method	3000 on-board co-averaged spectra
Dimensions	30 × 23 × 12 cm^3 (with insulation box)
Total weight	3.8 kg

2019 prototype version



Graf *et al.*, AMT, **2021**

Laboratory validation performance

- Precision **< 0.1 %** at 1 s resolution
- Accuracy **< 1.5 %** at UTLS-relevant conditions

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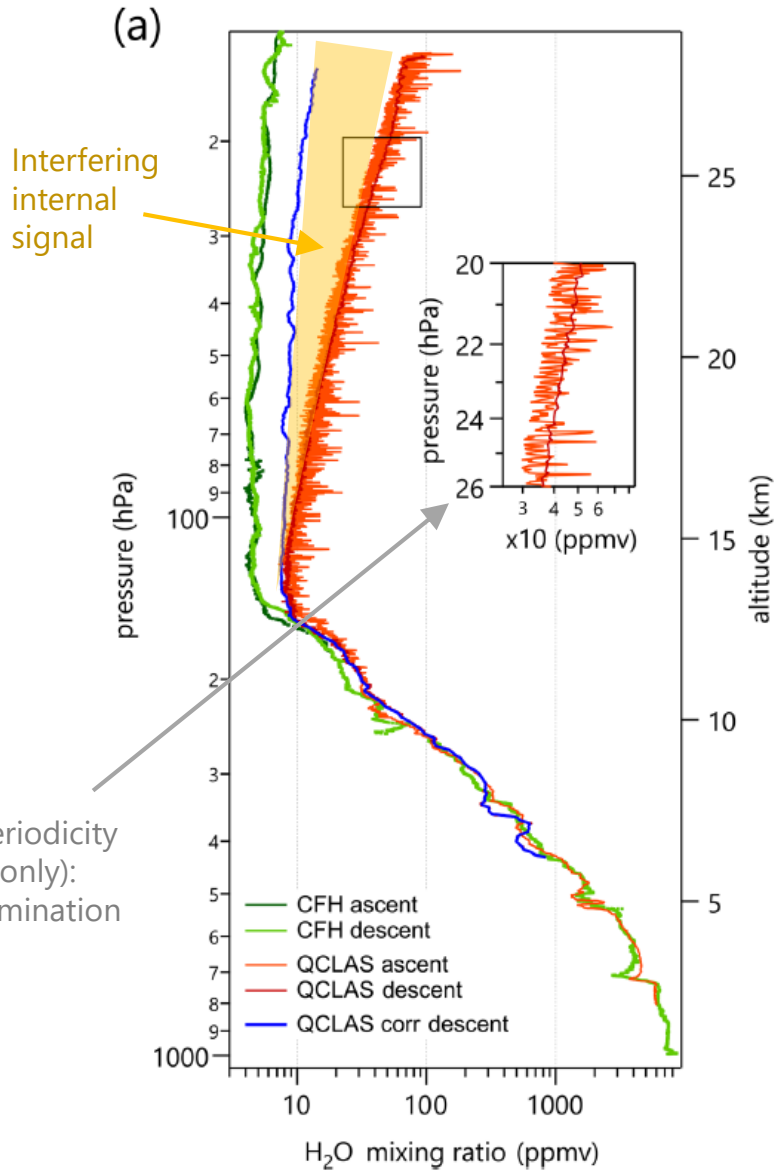
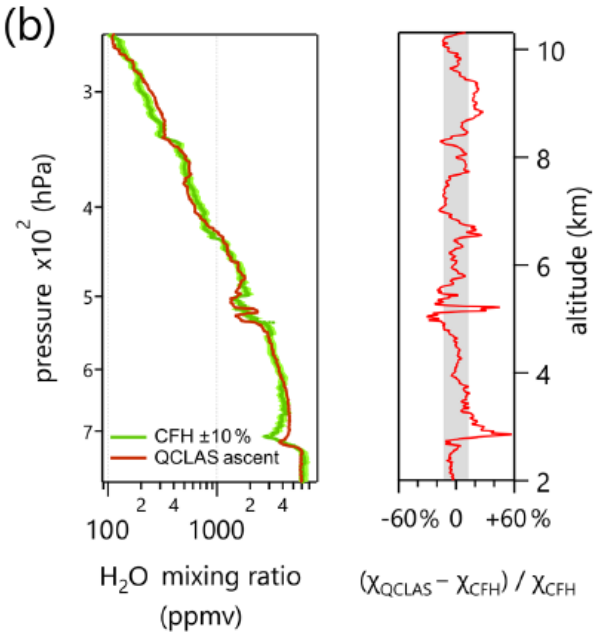
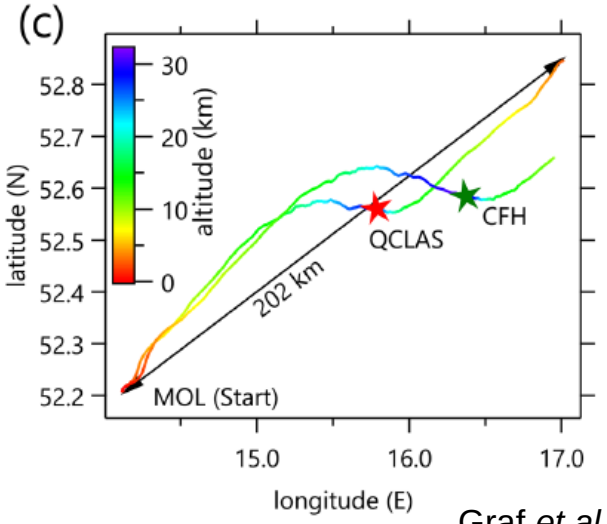
Test flights in Lindenberg (2019)

- Good operation during entire flight (up to 28 km altitude)
- **Troposphere:** excellent agreement with CFH (± 3 %)
- **Stratosphere:** moist bias due to *interfering absorption signal* (outgassing H_2O from inside the instrument's casing) → Solved by mechanical engineering
- New in-flight validation campaign in collaboration with DWD Lindenberg in preparation







Graf *et al.*, AMT, **2021**

Laboratory validation campaign (2021)

- Collaboration with Swiss Federal Institute of Metrology (METAS)
- **Objective:** absolute validation of QCLAS in terms of precision, accuracy, and linearity using SI-traceable reference mixtures at UTLS-relevant conditions

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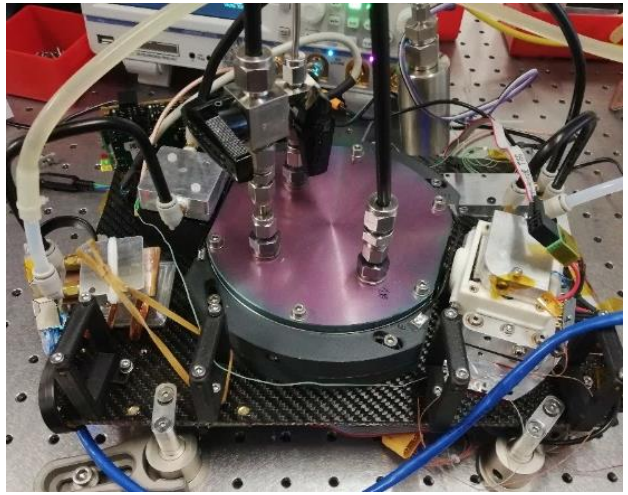
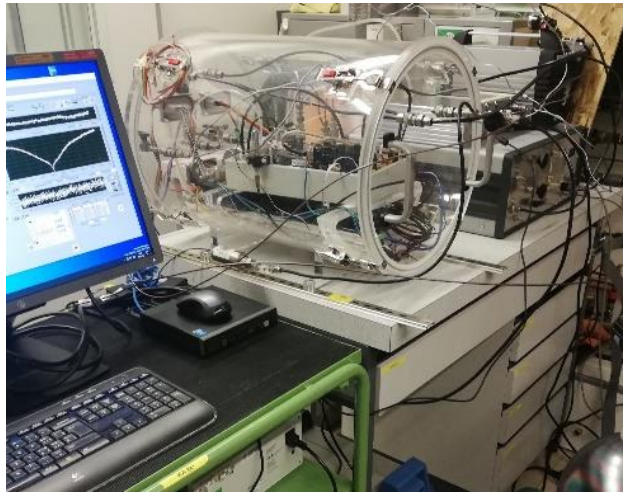
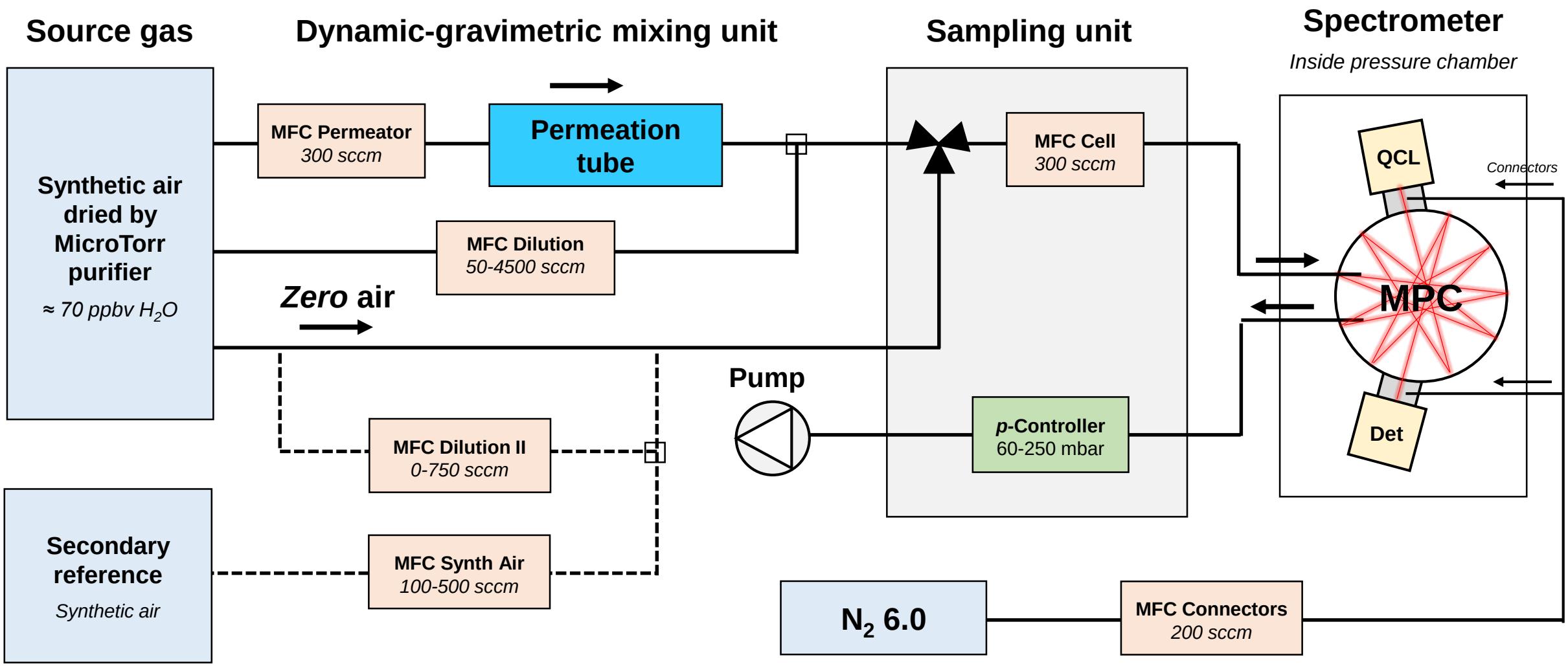
Technical setup

- **Dynamic-gravimetric permeation source**
  - H_2O amount fraction = 2.5 – 35 ppmv
  - Uncertainty < 1.5 %
  - SI-traceable
- **QCLAS in "lab configuration"**
  - MPC sealed by stainless-steel lids
  - Inert coating of all surfaces → minimize desorption effects
  - Pressure range 30 – 250 mbar

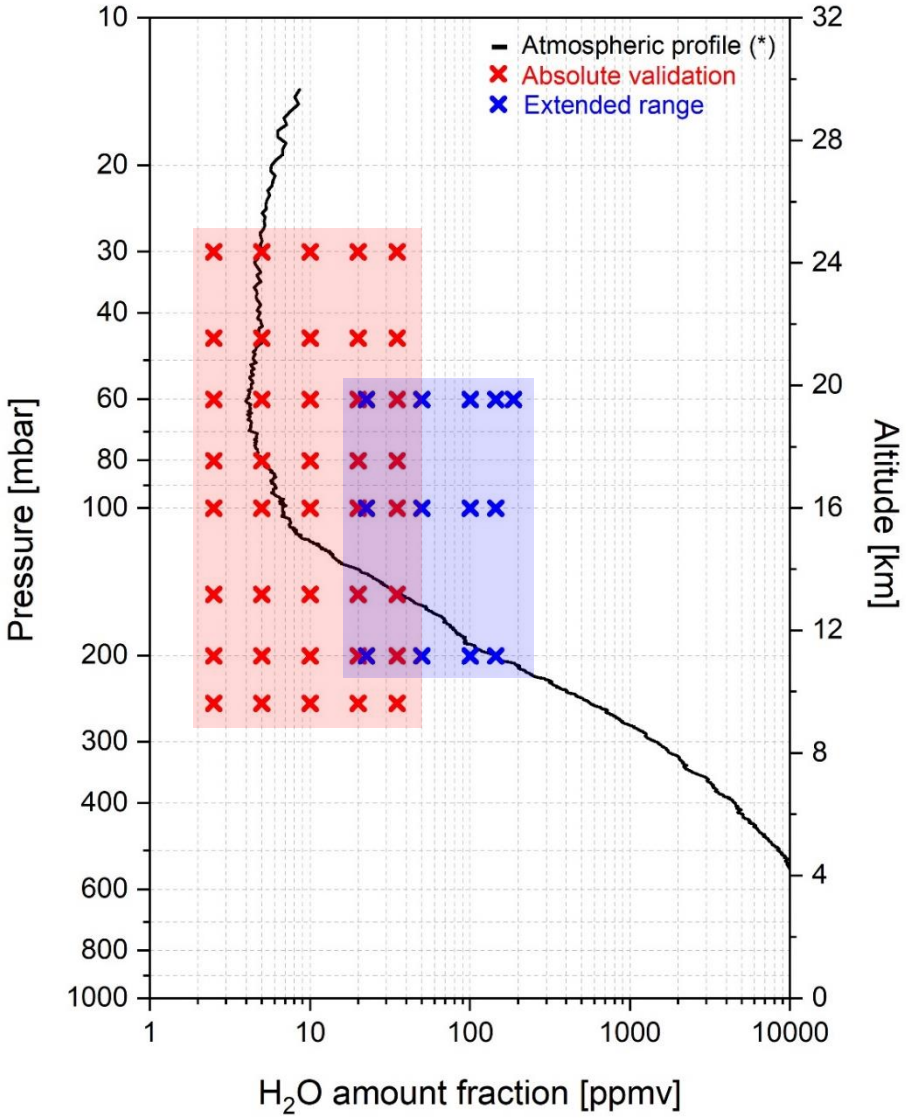




Experimental setup



Measurement strategy



Absolute validation range

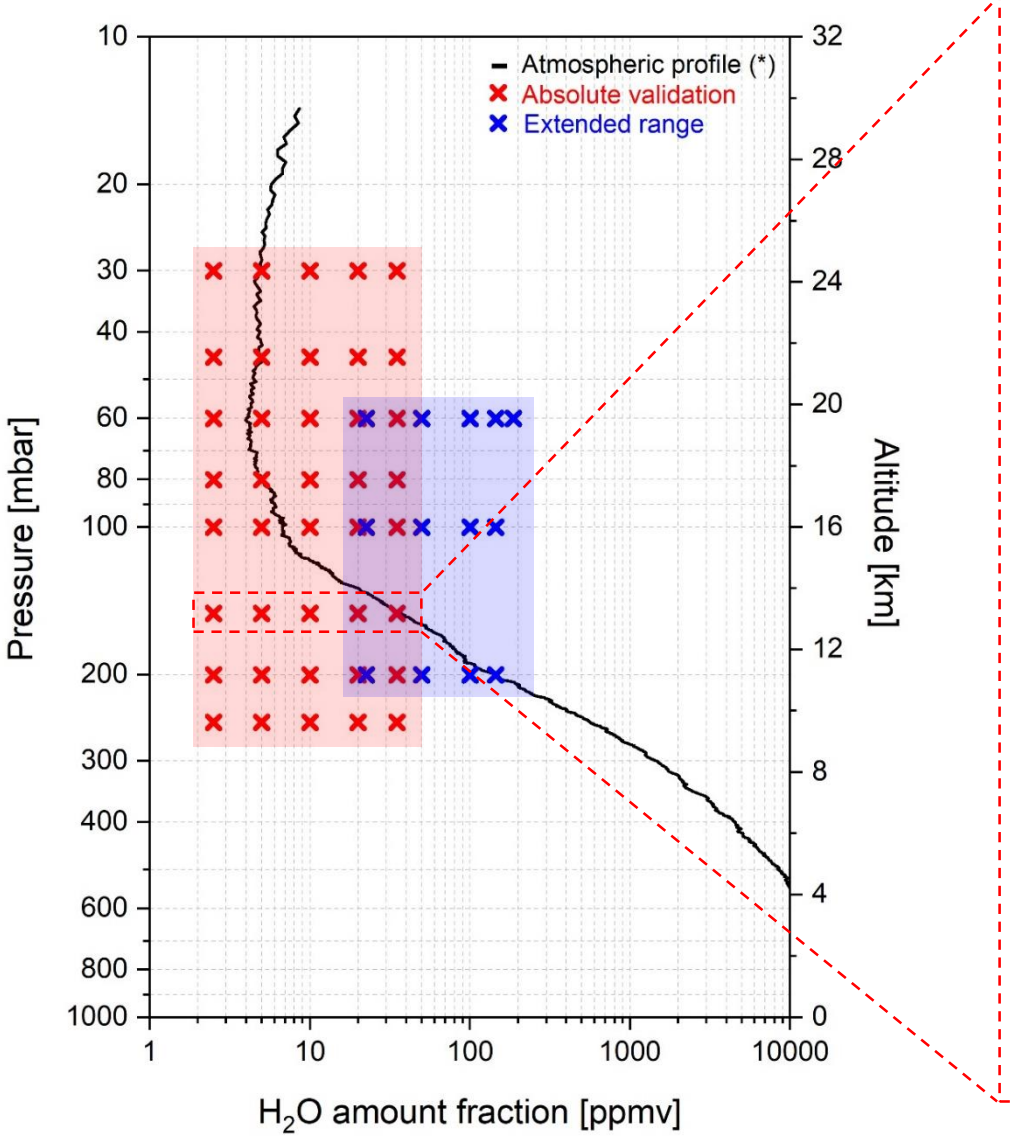
Source gas	Gravimetric permeation (SI-traceable)
H_2O amount fraction	2.5 – 35 ppmv (5× levels)
Pressure	30 – 250 mbar (8× levels)

Extended range

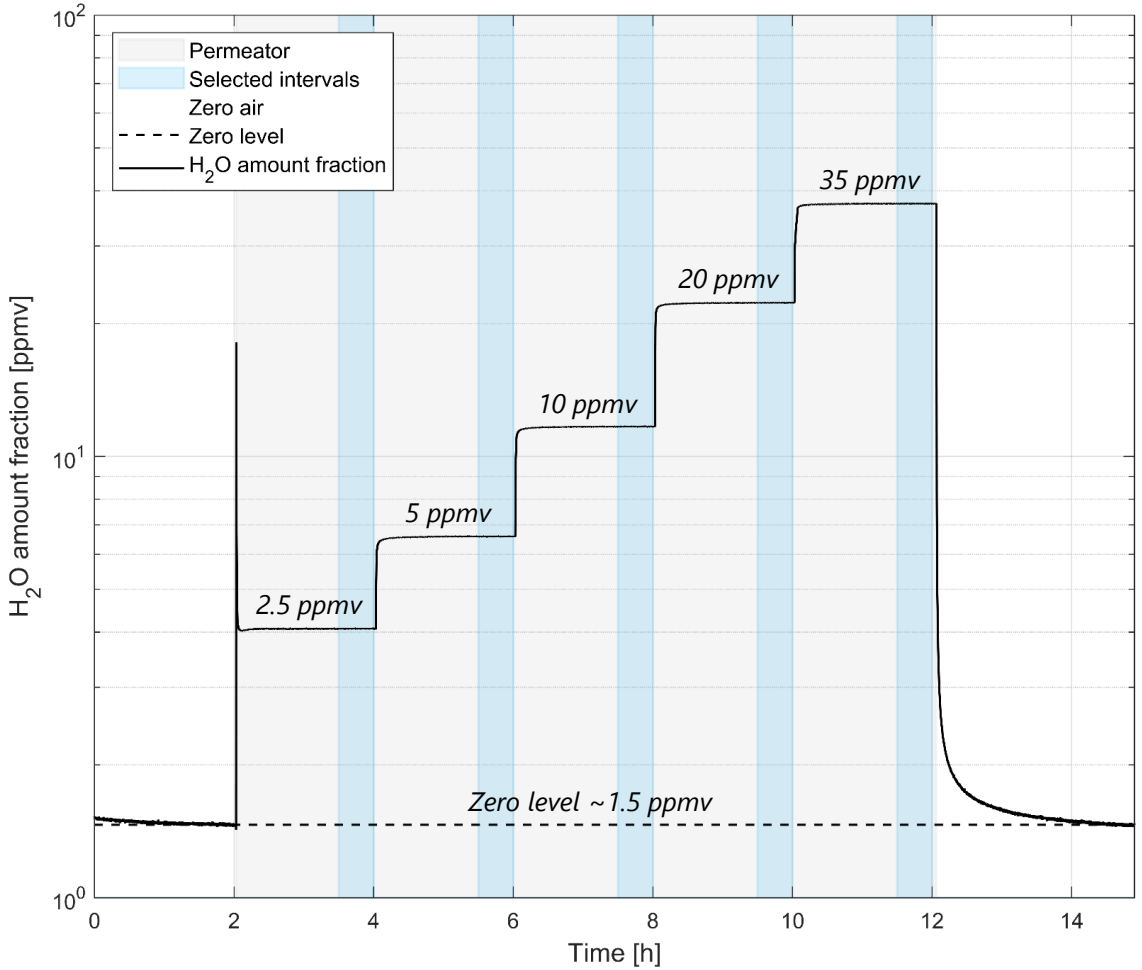
Source gas	Synthetic air (secondary reference)
H_2O amount fraction	22 – 150 ppmv (4× levels)
Pressure	60 – 200 mbar (3× levels)

(*) Brunamonti *et al.*, ACP, **2018**

Measurement strategy



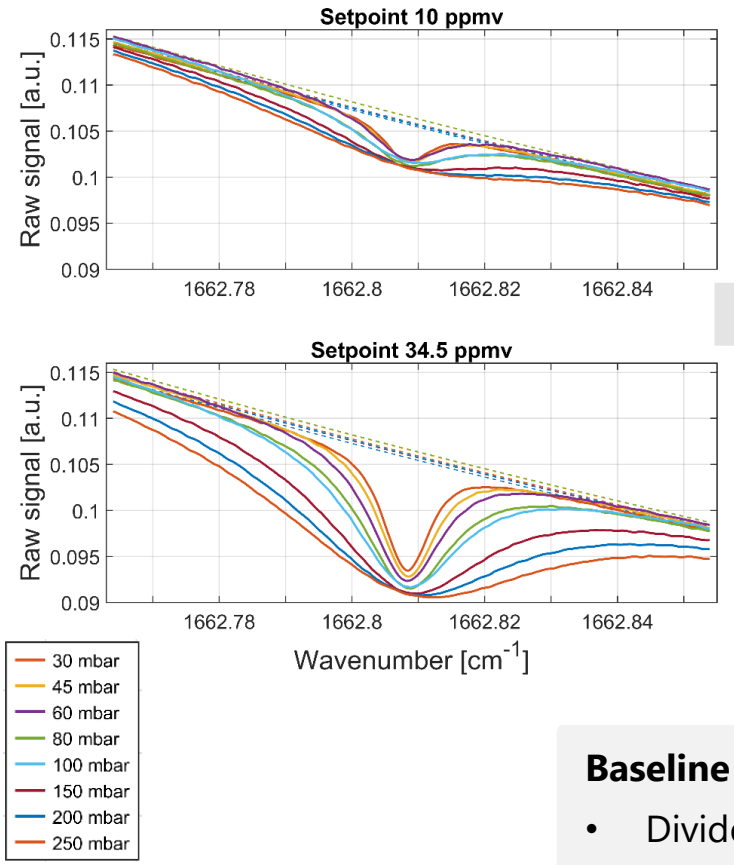
Single experiment: p = 150 mbar



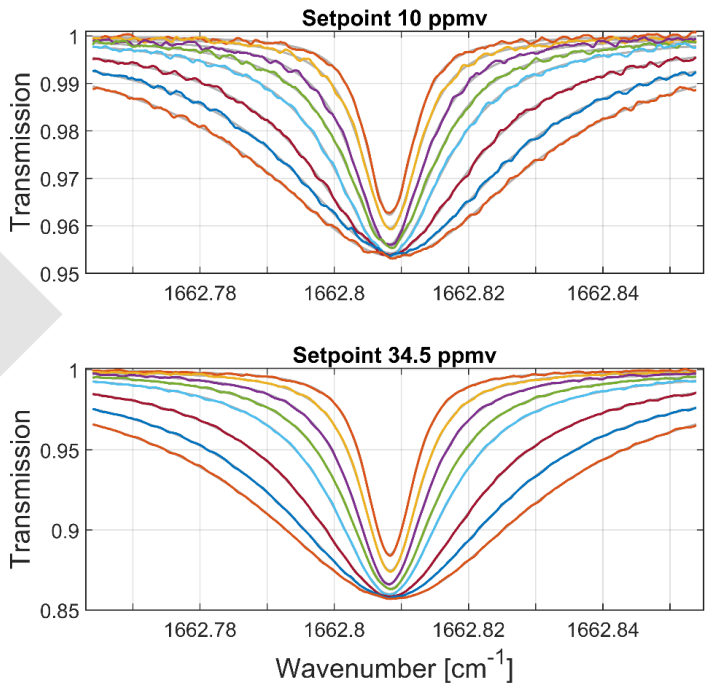
(*) Brunamonti *et al.*, ACP, **2018**

Spectroscopic retrieval

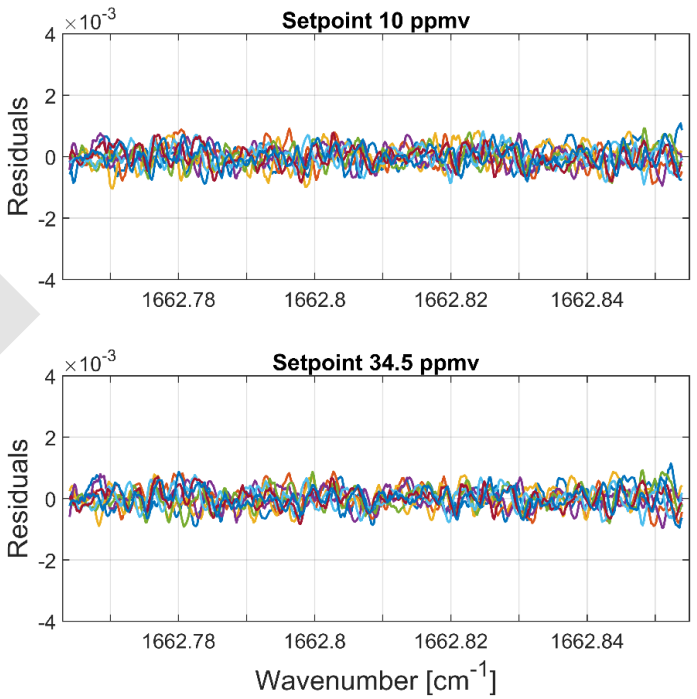
Raw spectra



Normalized spectra



Residuals (i.e. observed – fitted)



Baseline normalization

- Divide each spectrum by corresponding "zero level" spectrum (*lab*)
- Polynomial fitting (*flight*)

Fitting

➢ **Calculate H_2O amount fraction**

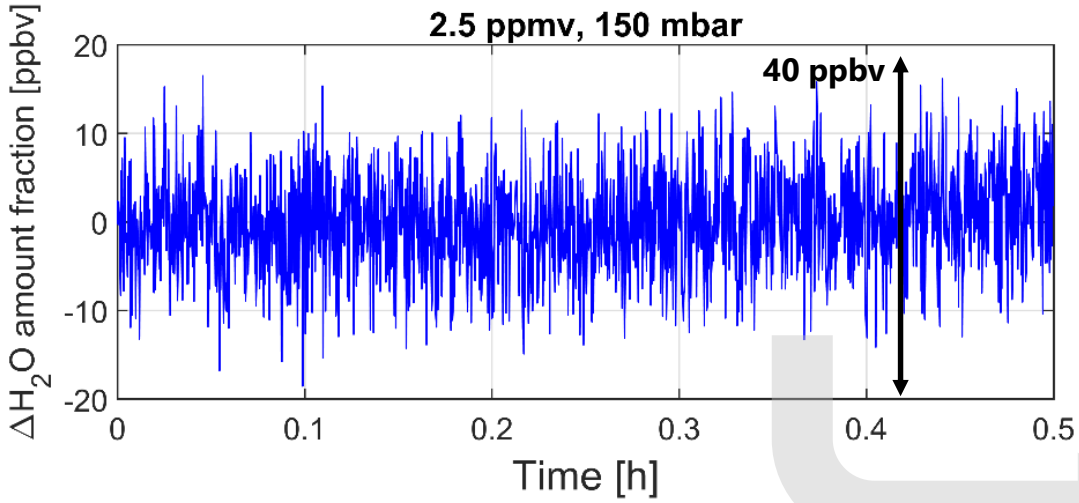
Requires:

- Environmental parameters (*p*, *T*)
- Absorption line shape model (qSDVP)
- Molecular parameters

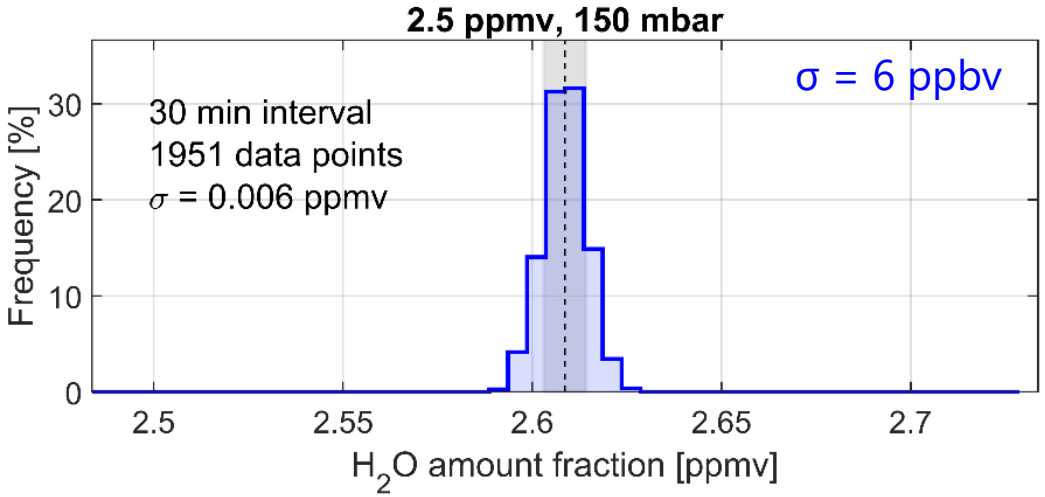
Brunamonti *et al.*, **In prep.**

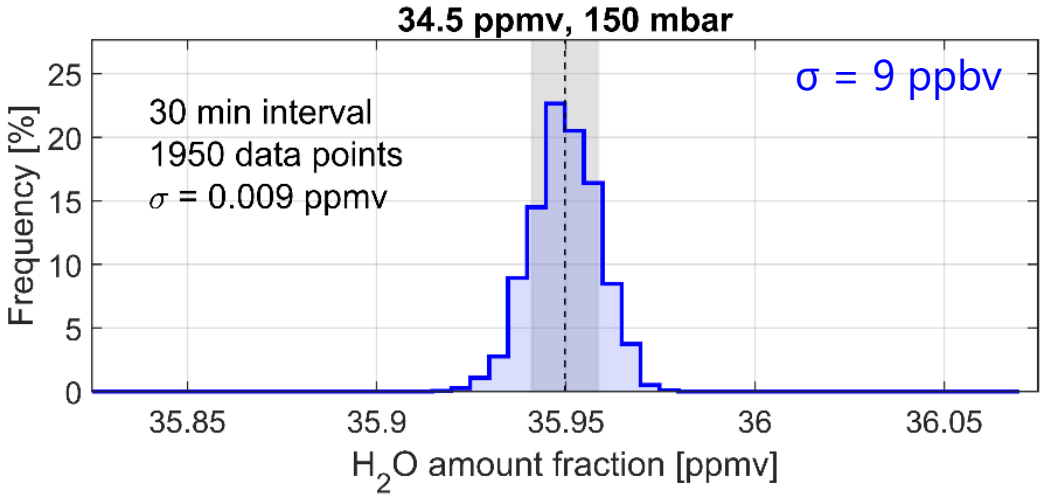
Results: Precision, long-term stability

Timeseries (30 min, 1 s resolution)



Frequency of occurrence distributions





- **Standard deviation < 30 ppbv (i.e. < 0.1 %)** at 1 s resolution at all conditions

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Results: Accuracy (SI-traceable)

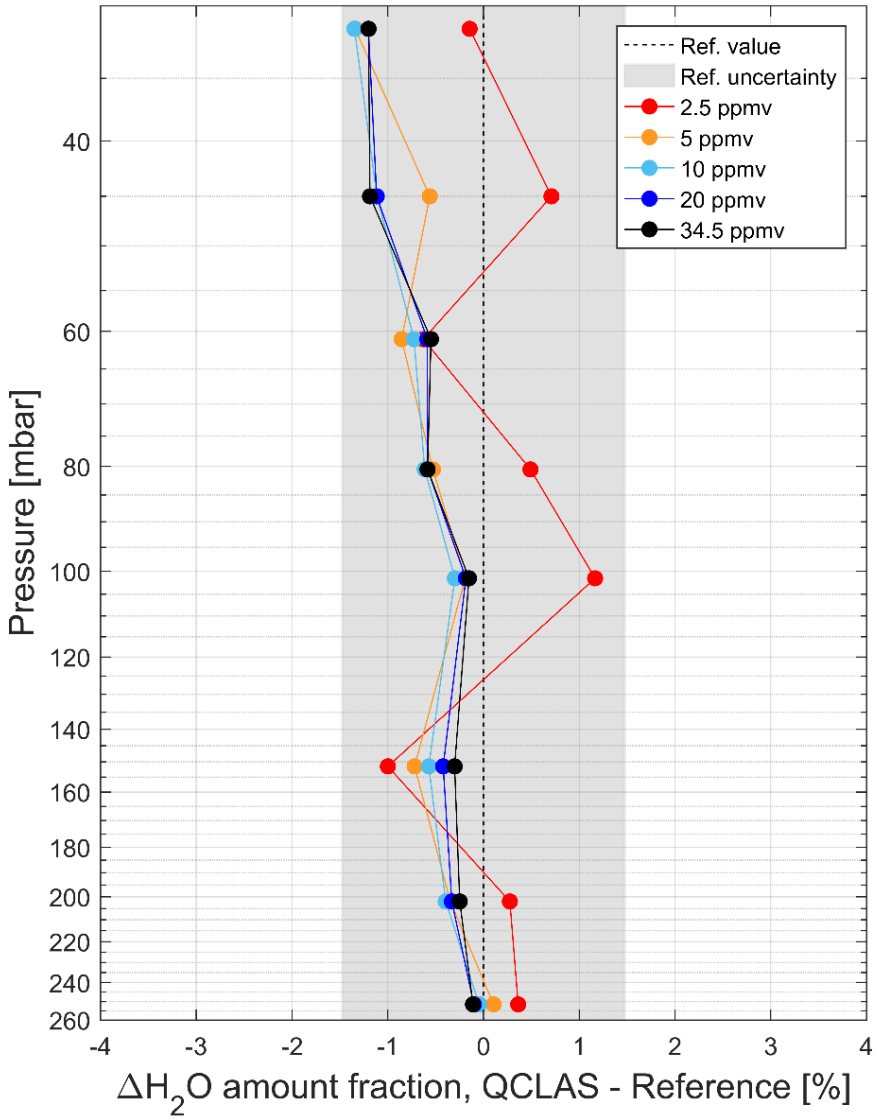
Fitting algorithm

- Least-squared minimum multi-spectral fitting
- 8 spectra fitted simultaneously (different *p*)
- Integration time: 50 s
- Line shape model: quadratic Speed-Dependent Voigt Profile (qSDVP)
- Molecular parameters (Γ_0, Γ_2) determined *a-priori* by the fitting algorithm

➢ **All measurements** within ± 1.5 % uncertainty of the reference

➢ Maximum absolute deviation from reference = 210 ppbv

Deviation from reference [%]



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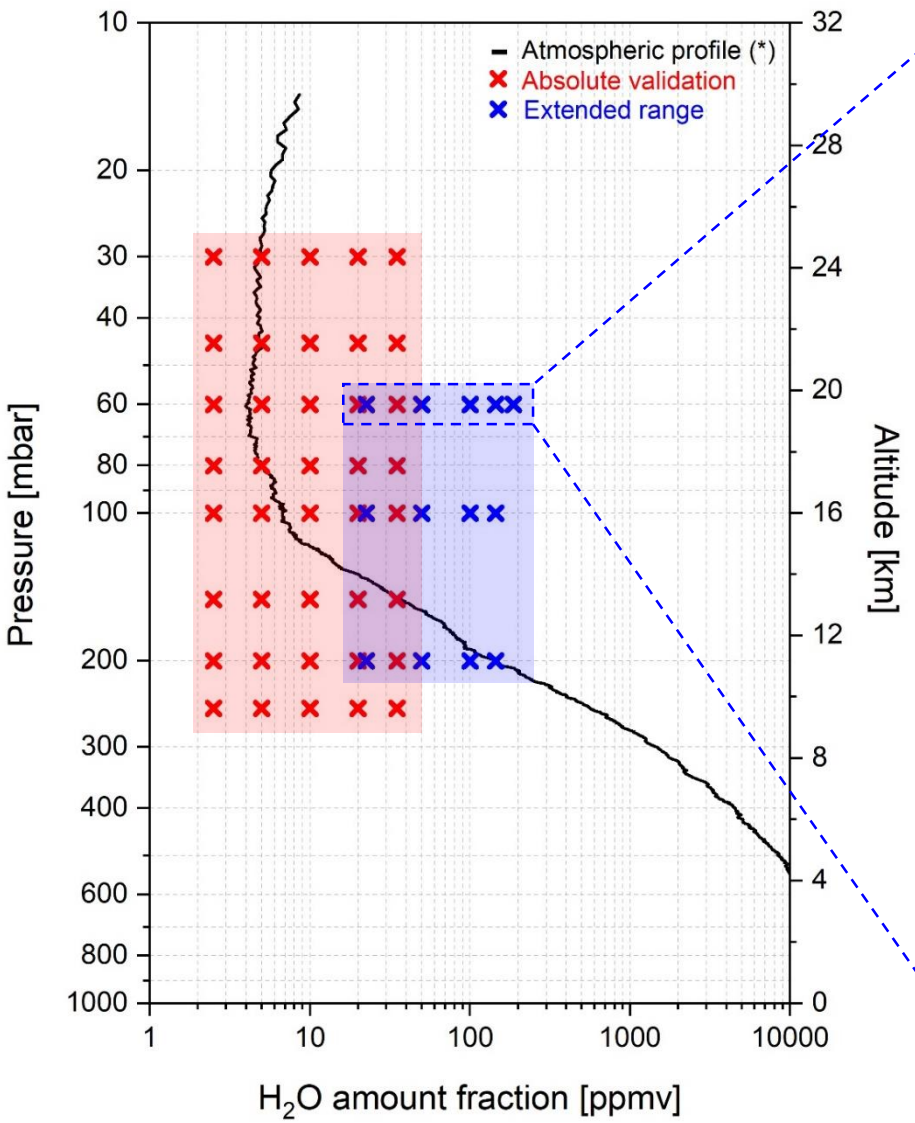
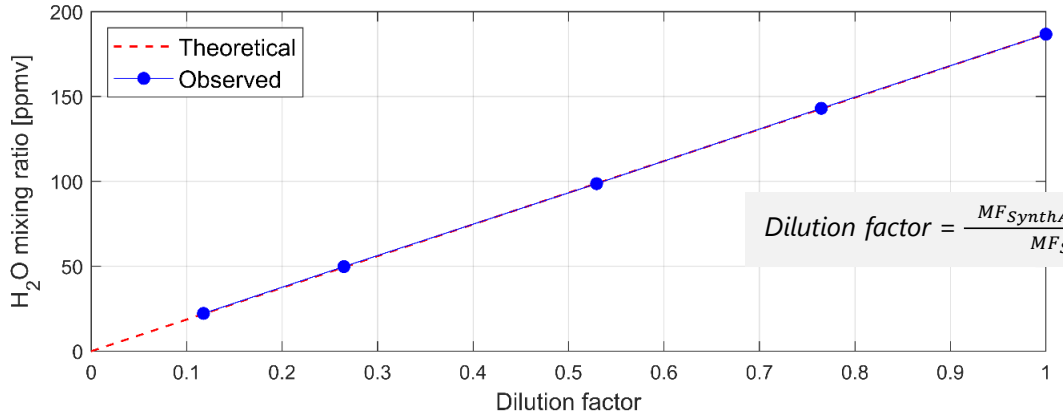
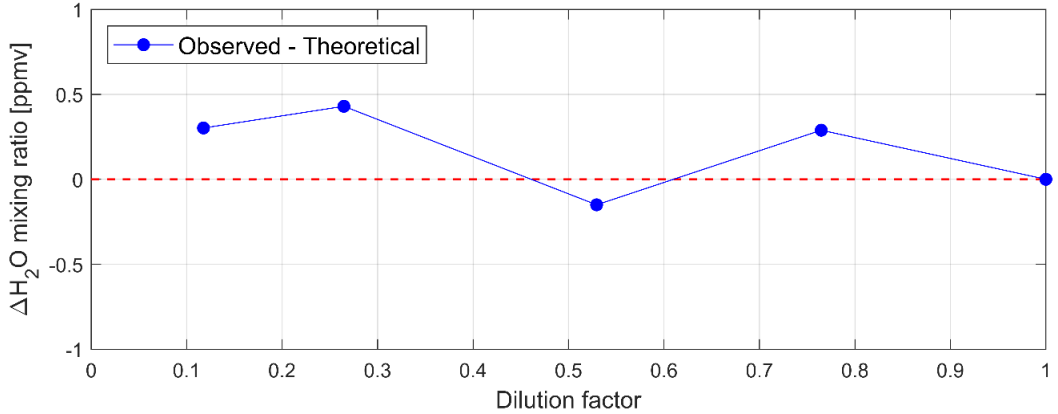
Results: Linearity up to 150 ppmv H_2O



Secondary reference (p = 60 mbar)



$$\text{Dilution factor} = \frac{MF_{SynthAir} - MF_{ZeroAir}}{MF_{SynthAir}}$$



➢ **Linearity** within ± 1.5 % from theoretical value up to 150 ppmv

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(*) Brunamonti *et al.*, ACP, **2018**

Conclusions and outlook

- We developed a compact quantum-cascade laser absorption spectrometer (**QCLAS**) for balloon-borne measurements of H_2O in the UTLS
- Test flights performed in 2019 show excellent agreement with CFH in the troposphere. The moist bias in the stratosphere due to interfering internal signal was solved by mechanical engineering
- Laboratory validation using SI-traceable reference mixtures of H_2O at UTLS-relevant conditions show outstanding performance for a balloon-borne hygrometer:

Precision **< 0.1 %** (i.e. < 30 ppbv) for 2.5–35 ppmv H_2O at 1 s resolution

Accuracy **< 1.5 %** (i.e. < 210 ppbv) for 2.5–35 ppmv H_2O

Linearity **< 1.5 %** (i.e. < 1.5 ppmv) up to 150 ppmv H_2O

- New test flights with optimized/improved instrument are in preparation in collaboration with DWD Lindenberg (Germany).

Acknowledgements

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Thanks for your attention!