EGU General Assembly 2023

EGU23-13161, updated on 29 Mar 2024
https://doi.org/10.5194/egusphere-egu23-13161
EGU General Assembly 2023

SI-traceable balloon-borne measurements of water vapor in the upper atmosphere

Béla Tuzson[1], Simone Brunamonti[1], Manuel Graf[1], Tobias Bühlmann[2], and Lukas Emmenegger[1]
[1]EMPA, Air Pollution/Environmental Technologies, Duebendorf, Switzerland (bela.tuzson@empa.ch)
[2]METAS, Swiss Federal Institute of Metrology, Laboratory Gas Analysis, Bern, Switzerland

Water vapor (H_2O) in the upper troposphere-lower stratosphere (UTLS) is of great importance to the Earth's radiative balance. Yet, accurate measurements of H_2O in this region are notoriously difficult, and significant discrepancies were found in the past between different techniques (both in-situ and remote sensing). Currently, cryogenic frostpoint hygrometry is considered as the reference method for balloon-borne measurements of UTLS H_2O [1]. However, these devices must be fundamentally reconceived due their use of fluoroform (HFC-23) as cooling agent, which has to be phased out due to its high global warming potential. There is thus an urgent need for alternative, reliable technologies to monitor UTLS H_2O, e.g. in long-term global observing networks, such as the GCOS Reference Upper Air Network (GRUAN).

Here we present a new mid-IR quantum-cascade laser absorption spectrometer for balloon-borne measurements of UTLS H_2O (ALBATROSS). The spectrometer incorporates a specifically designed segmented circular multipass cell (SC-MPC) that allows for an optical path length of 6 m [2], and it fulfills stringent requirements in terms of mass (< 3.5 kg), size, and temperature resilience. Two successful test flights demonstrated the instrument's outstanding capabilities under real atmospheric conditions up to 28 km altitude [3].

During flights, the instrument experiences a harsh environment with up to 80 K temperature variations and two orders of magnitude change in pressure and H_2O amount fraction. To achieve reliable and SI-traceable, we determined the spectral performance and the accuracy of the retrieved amount fractions of ALBATROSS at UTLS-relevant conditions using a dynamic-gravimetric permeation method [4]. SI-traceable reference mixtures were generated with H_2O amount fractions as low as 2.5 μmol/mol (or parts per million, ppm) in synthetic air. The results show that ALBATROSS achieves an accuracy better than ±1.5 % at all investigated pressures (30–250 mbar) and H_2O amount fractions (2.5–35 ppm). The 1 s relative precision is better than 0.3 %, while 5 nmol/mol (i.e., parts per billion, ppb) is reached by averaging for 100 s. Furthermore, ALBATROSS reaches a linear response within ±2 ppm up to 180 ppm H_2O.

To achieve this level of performance, the Voigt profile was found to be inadequate. Therefore, we empirically determined all parameters needed to implement the quadratic speed-dependent Voigt profile (qSDVP), which accounts for speed-dependent collision broadening. The qSDVP more accurately captures the H_2O line shape at all pressure conditions, and thus significantly improves the accuracy of the retrieved water vapor amount fraction.

Overall, ALBATROSS achieves an unprecedented level of accuracy and precision for a balloon-borne hygrometer, and it demonstrates the exceptional potential of mid-IR laser absorption spectroscopy for in-situ measurements of UTLS H_2O. Further in-flight validation campaigns from Lindenberg (Germany) and Payerne (Switzerland) are currently in preparation.

[1] Brunamonti et al., *J. Geophys. Res. Atmos.*, **2019**, 124, 13, 7053-7068.

[2] Graf et al., *Atmos. Meas. Tech.*, **2021**, 14, 1365-1378.

[3] Graf, Emmenegger and Tuzson, *Opt. Lett.*, **2018**, 43, 2434-2437.