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Quantum-cascade laser absorption spectroscopy for balloon-borne measurements of stratospheric H₂O

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Water vapor (H₂O) is the strongest greenhouse gas in our atmosphere, and it plays a key role in multiple processes that affect weather and climate. Particularly, H₂O in the upper troposphere - lower stratosphere (UTLS) is of great importance to the Earth's radiative balance, and has a significant impact on the rate of global warming. Hence, accurate measurements of UTLS H₂O are crucial for understanding and projecting climate. Currently, the reference method used for in-situ measurements of UTLS H₂O aboard meteorological balloons is cryogenic frostpoint hygrometry (CFH) [1]. However, the cooling agent required for this technique (trifluoromethane) is phasing out as of 2020, due to its strong global warming potential. This represents a major challenge for the continuity of global, long-term stratospheric H₂O monitoring networks, such as the GCOS Reference Upper Air Network (GRUAN).

As an alternative to CFH, we developed a compact instrument based on mid-IR quantum-cascade laser absorption spectroscopy (QCLAS) [2]. The spectrometer, with a total weight of 3.9 kg, relies on a segmented circular multipass cell [3] that was specifically developed to meet the stringent requirements, in mass, size and temperature resilience, posed by the harsh environmental conditions of the UTLS. Quick response and minimal interference by H₂O outgassing from surfaces are achieved by an open-path approach. An elaborate thermal management system ensures excellent internal temperature stability, despite of outside temperature variations of up to 80 K.

In collaboration with the German Weather Service (DWD), two successful test flights were performed in December 2019 in Lindenberg, Germany. We will report on the results of these test flights, highlighting the instrument outstanding capabilities under UTLS and stratospheric conditions (up to 28 km altitude), and identifying some limitations. Further development activities triggered by the test flights, involving both hardware adaptations and spectral analysis modifications, will be also discussed. The final validation will be addressed, in cooperation with the Swiss Federal Institute of Metrology (METAS), by laboratory experiments in a custom-made climate chamber, using dynamically generated, SI-traceable reference mixtures with H₂O amount fractions below 20 ppmv and uncertainty < 1%. The ultimate goal is to demonstrate the potential of QCLAS as a highly valuable technique for quantitative balloon-borne measurements of UTLS and stratospheric H₂O.

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[2] Graf et al. (2020), Atmos. Meas. Tech. Discuss., doi.org/10.5194/amt-2020-243 (Accepted 4 January 2021).

[3] Graf, Emmenegger and Tuzson (2018), Opt. Lett., doi.org/10.1364/OL.43.002434.