

Balloon-borne laser spectrometer for UTLS water research (ALBATROSS)

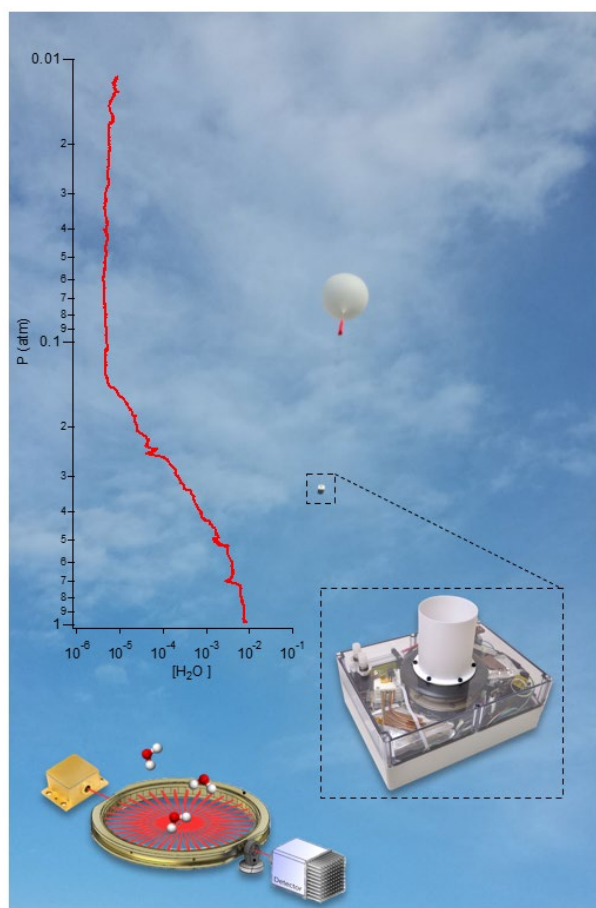
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1 Summary

Accurate and reliable measurements of water vapor (H_2O) in the upper troposphere and lower stratosphere (UTLS) are of utmost importance for a wide range of scientific research fields, such as forecast modeling, radiative transfer calculations, and climate trend studies. At present, however, in-situ measurements of UTLS H_2O are still very challenging and exhibit significant uncertainties. Recent field campaigns and laboratory experiments have provided clear evidence of large deviations between different hygrometers, making them questionable for useful determination of relative humidity at very low water vapor content (few ppm), such as in the UTLS.

The goal of this project is to establish the mid-IR laser absorption spectroscopic technique as a viable alternative method to the cryogenic frostpoint hygrometer (CFH), which is currently considered the reference method. Thereby we relied on a laser spectrometer prototype, hereafter named "ALBATROSS", previously developed at Empa (Graf et al., 2021). The instrument is based on direct absorption laser spectroscopy in the mid-IR range (around $6 \mu\text{m}$), and incorporates a monolithic, segmented circular multipass cell (SC-MPC), that was specifically developed to meet the stringent requirements posed by the harsh environmental conditions of the UTLS (e.g. Graf et al., 2018).

Three work packages (WPs) were proposed, corresponding to the three main objectives required to achieve the overall goal: enhancement and optimization of the existing prototype laser spectrometer (WP1), laboratory-based calibration and validation experiments (WP2), and in-flight validation field campaigns (WP3). WP1 represents the main activity carried out at Empa, and addressed three main tasks: minimizing interfering water absorption signal from the instrument's casing (Task A) that was observed at altitudes above the tropopause; further reduction of the payload weight (Task B), and development of improved line shape model fitting (Task C). For the laboratory-based (WP2) and in-flight (WP3) validation and calibration activities, we leveraged on the expertise and infrastructure available at two Swiss national institutions, Empa and METAS, and the Deutscher Wetterdienst/GRUAN Lead Centre in Lindenberg, Germany.

The laboratory-based validation and calibration activities were performed within the frame of two measurement campaigns: a dedicated SI-traceable validation, in collaboration with the Swiss Federal Institute of Metrology (METAS), conducted in spring 2021, and the AquaVIT4 International Intercomparison of Atmospheric Hygrometers (<https://www.hemera-h2020.eu/aquavit-4/>), held at the Aerosol Interactions and Dynamics in the Atmosphere (AIDA) climate chamber at the Karlsruhe Institute of Technology (KIT), Germany, in spring 2022. The data collected during these campaigns demonstrate the outstanding performances of ALBATROSS, in terms of accuracy, precision, linearity, and response time, as well as provided the information necessary for developing the improved line shape model fitting.

Finally, the instrument was successfully validated under real atmospheric conditions in a tandem balloon flight up to 30 km altitude against the cryogenic frostpoint hygrometer (CFH) at the Lindenberg Meteorological Observatory/GRUAN Lead Centre, Germany, in September 2022. The instrument performed well during the entire flight duration, down to pressures of 13 mbar (burst point) and temperatures of $-63 \text{ }^\circ\text{C}$ (tropopause), including the passage through one liquid water cloud (altitude 2.6 km) and two ice clouds (10.2 km and 11.5 km altitude) during the ascent of the balloon. The data (still under evaluation) show excellent agreement between ALBATROSS and CFH in the upper atmosphere.

The results obtained within the project show that ALBATROSS achieves unprecedented performance, in terms accuracy, precision, and response time for a balloon-borne hygrometer, demonstrating the real potential of mid-IR laser absorption spectroscopy as an alternative reference method to cryogenic frostpoint hygrometry for in-situ measurements of H_2O in the UTLS.

2 Scientific report

2.1 Introduction

Water vapor (H_2O) is the strongest greenhouse gas in our atmosphere, and its abundance in the upper troposphere-lower stratosphere (UTLS) is of great importance to the Earth's radiative balance. Hence, accurate and reliable measurements of H_2O in the UTLS are of utmost importance for a wide range of scientific research fields, such as forecast modeling, radiative transfer calculations, and climate trend studies. At present, however, in-situ measurements of UTLS H_2O are still very challenging and exhibit significant uncertainties. Recent field campaigns and laboratory experiments have provided clear evidence of large deviations between different hygrometers (e.g., Fahey et al., 2014), making them questionable for useful determination of relative humidity at very low water vapor content (few parts per million - ppm), such as in the UTLS. Currently, the reference method for balloon-borne measurements of UTLS H_2O is cryogenic frost-point hygrometry (CFH) (Vömel et al. 2016). However, these devices are currently undergoing a fundamental re-conception due their use of fluoroform (HFC-23) as cooling agent, which must be phased out due of its high global warming potential (UNEP, 2016). The goal of this project is to establish mid-IR laser absorption spectroscopy as a viable alternative technique for balloon-borne measurements of UTLS H_2O .

This project aims at the enhancement, optimization and calibration/validation of an existing laser spectrometer prototype, named "ALBATROSS", previously developed at Empa. The instrument called ALBATROSS was described in detail by Graf et al. (2021). Briefly, it incorporates a monolithic segmented circular multipass cell (SC-MPC), consisting of a rotationally symmetric arrangement of individual mirror segments carved into its inner surface (Graf et al., 2018). This geometry was found to be highly tolerant to thermally induced distortion, robust to mechanical stress, and thus, well suited for open-path applications (Tuzson et al., 2020). The SC-MPC contains 57 circularly arranged mirror segments with a diagonal distance of 108.8 mm, resulting in an optical path length (OPL) of 609.4 cm. A distributed feedback quantum cascade laser (DFB-QCL) is used as a light source. The laser is tuned across $\sim 1 \text{ cm}^{-1}$ spectral window, centered around an isolated H_2O absorption line at 1662.809 cm^{-1} ($\lambda \approx 6.01 \mu\text{m}$). Rapid spectral sweeping of the QCL is achieved by periodic modulation of the laser driving current, following the intermittent continuous wave (ICW) modulation approach (Fischer et al., 2014, Liu et al., 2018). The spectra are acquired at a frequency of 3 kHz and co-averaged in real-time to 1 s.

2.2 Methods and activities

The various activities within the project were allocated to three different work packages (WPs). Here we review the main tasks of each individual WP.

WP1. This dealt with the enhancement and optimization of the existing prototype laser spectrometer (ALBATROSS), and consisted of three main tasks: minimizing interfering water absorption signal from inside the instrument's enclosure (Task A), payload reduction by optimizing weight intensive elements (Task B), and developing improved line shape model fitting (Task C).

Task A refers to the observed interference by outgassing H_2O from the surfaces inside the instrument's enclosure when flying above the tropopause. In particular, this led to a substantial moist bias in the stratosphere (Graf et al., 2021). We identified its source as being the additional absorption of the residual H_2O within the spectrometer's housing along the short free-space optical path ($<1 \text{ cm}$) between the laser housing/detector and the multipass cell. Although this internal "gap" is only 0.14 % of the total OPL, the corresponding absorption can still generate a significant offset at high altitudes, where outgassing is enhanced by the low atmospheric pressure (Graf et al., 2021) and the ambient water vapor reaches very low (ppm-level) amount fractions.

We addressed this limiting issue by designing and implementing two custom-made connectors that hermetically connect the optoelectronic components (detector and laser housing) to the multipass cell (Figure 1, left). The connectors provide both the required *mechanical flexibility* for optical alignment and *thermal decoupling* between the components, while *shielding* the internal free optical path from water vapor that outgasses from the surfaces within the enclosure of the instrument. In addition, the connectors feature a gas fitting that is used to actively purge their internal volume with dry air, thereby further reducing the effect of any spurious absorption contribution from internal water vapor. We found

that purging with dry N₂ at a flow rate of 200 sccm, the proposed solution efficiently solves the interfering internal signal issue (Figure 1, right). This was a prerequisite for conducting all the subsequent validation activities (WPs 2-3).

For the flight operation of ALBATROSS, we built an *in-flight purging* system to keep the connectors dry for the duration of a balloon sounding. This consists of a miniaturized cartridge containing 25 g CO₂, combined with two critical orifices (diameter ~10 μm) that restrict the flow rate to roughly 100 sccm, resulting in a slow release of the CO₂ lasting around 3 hours. This solution was successfully tested in a climate chamber down to temperatures of -50 °C prior to the in-flight validation. While, of high importance, the purging system unavoidably involved additional weight (~200 g) to the overall payload, which made the task of the next WP, addressing the reduction of payload, even more challenging.

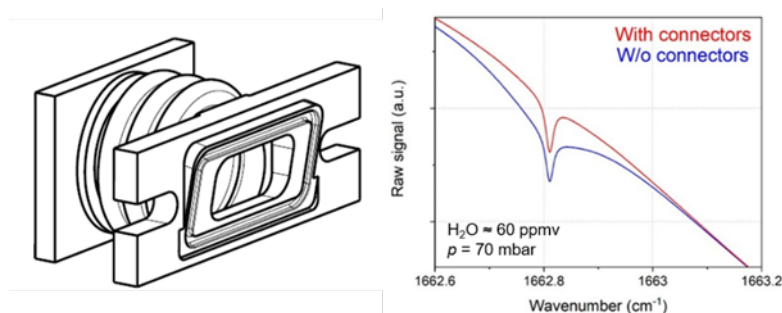


Figure 1. 3D-drawing of the connectors developed to eliminate the interfering internal absorption (left). Effect of purging the connectors on the absorption spectrum measured at 70 mbar pressure and 60 ppm H₂O amount fraction (right). The broad absorption feature, which is due to ambient H₂O absorption outside the multipass cell is eliminated.

Nevertheless, the payload reduction (Task B) was accomplished by implementing a custom-made enclosure, made out of lightweight carbon-aramid honeycomb plates, which reduces the weight of the existing (polycarbonate) casing by more than a factor of 3 (i.e., from 1 kg to 0.3 kg). Furthermore, an improved intake tube design has been implemented, with the purpose of reducing both weight and surface adsorption effects. This comprises two lightweight aluminum funnels and two custom-made stainless-steel extension tubes (thickness 0.5 mm). This results in an overall weight reduction of 550 g, from the previous 3.95 kg.

Task C (improved line shape model fitting) was addressed using of the data collected during the SI-traceable validation campaign conducted at METAS in 2021 (part of WP2). Using a multi-spectral fitting approach, we empirically determined the molecular parameters required for the application of a quadratic speed-dependent Voigt profile (qSDVP) line shape model to our investigated H₂O line (not available in the literature). As discussed below, the upgrade to the qSDVP parameterization leads to a 2–6 % improvement in accuracy compared to the standard Voigt profile.

WP2. The laboratory-based validation and calibration activities were performed in two intensive measurement campaigns, conducted respectively at the Swiss Federal Institute of Metrology (METAS) in April-May 2021, and at the AIDA climate chamber of the Karlsruhe Institute of Technology (KIT), Germany, in March-April 2022. The latter was part of the AquaVIT4 International Intercomparison of Atmospheric Hygrometers, the 4th edition of a well-known series of hygrometer intercomparisons conducted at the AIDA chamber since 2007 (Fahey et al., 2014).

Prior to the measurement campaigns, a major activity consisted in the development of an experimental setup that addresses the numerous issues posed by measuring H₂O with high precision in a laboratory setting in the low ppm concentration range. This preparatory step was necessary for the assessment and validation measurements planned at both METAS and AquaVIT. Thus, a custom-made and fully automatized gas handling unit was developed to control the flow rate and pressure in the multipass cell (MPC), configured to close cell operation, and to allow for a wide range of measurement conditions, i.e. calibration, dilution, purging, etc. A detailed schematics of the sampling system used for the validation campaign at METAS is shown in Figure 2.

SI-traceable validation. The laboratory campaign dedicated to the SI-traceable validation of the instrument was conducted in collaboration with METAS. Using their established dynamic-gravimetric permeation method combined with dynamic dilution (Guillevic et al., 2018), we generated SI-traceable reference gas mixtures of H₂O in a synthetic air matrix, with H₂O amount fractions between 2.5–35 ppm

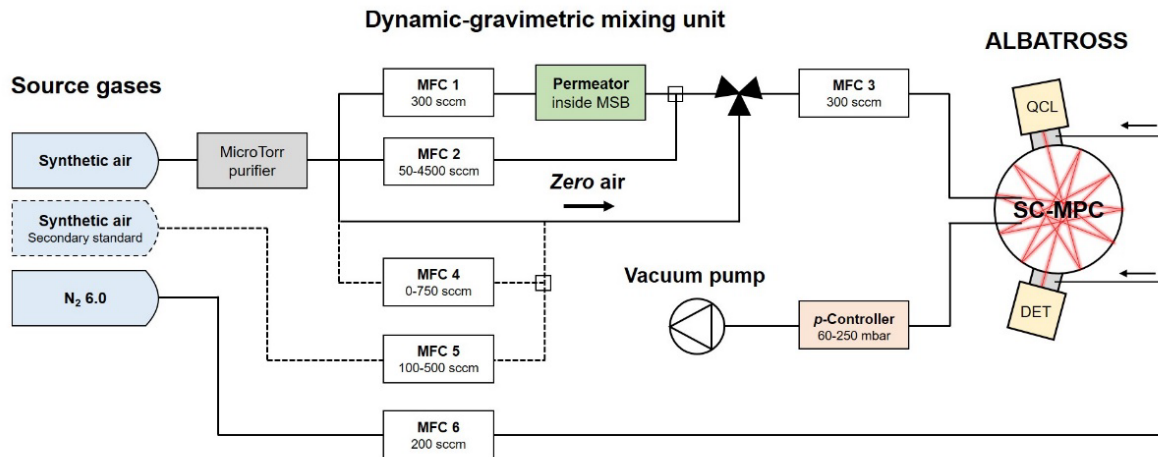


Figure 2. Schematics of the sampling system used for the SI-traceable validation campaign at METAS.

and uncertainty $<1.5\%$, which were measured by the spectrometer at pressures between 30–250 mbar. A total of 300 h of measurements were performed. This dataset provides the basis for the absolute validation, in terms of accuracy, precision and linearity of the ALBATROSS spectrometer at UTLS-relevant conditions. Furthermore, it allows for investigating the effects originating from the assumption of a simplified shape model (Voigt profile), and also for assessing the benefits of using more elaborated profiles (e.g. the quadratic speed-dependent Voigt profile, qSDVP) for the absorption lines in the fitting algorithm.

Figure 3 shows an overview of all H_2O setpoints and pressure levels investigated during the METAS validation campaign (panel a), overlaid with two atmospheric H_2O amount fraction profiles measured by CFH during recent field campaigns, and a representative time series of the measurements performed at 150 mbar (b). We selected five H_2O setpoints for our SI-traceable validation, covering the full range allowed by the dynamic-gravimetric permeation source (2.5–35 ppm), and eight pressure levels between 30–250 mbar, for a total of 40 possible combinations. This corresponds to the expected variability range of UTLS H_2O (Figure 3a).

Each H_2O setpoint generated by the permeator was measured for two hours to allow for the equilibration of the H_2O amount fraction in the SC-MPC (Figure 3b). The last 30 min of each interval are then selected for the precision assessment. Before and after each experiment, the SC-MPC is purged with zero air for at least 3 h to obtain the "empty-cell" spectrum, i.e. the transmission signal in the absence of the reference gas. This is used to normalize the consecutive raw absorption spectra.

Figure 4 (left column) shows such normalized spectra measured at 35 ppm, color-coded with pressure (top), and the corresponding residuals (i.e., observed – fitted spectrum) obtained using a Voigt profile (centre) and qSDVP (bottom) line shape model. The VP retrieval is performed using the line parameters from the HITRAN2020 database (Gordon et al., 2022), while for qSDVP we use our own molecular coefficients, empirically determined by a multi-spectral fitting approach. This consists in simultaneously minimizing the residuals of all 8 spectra measured at 35 ppm and different pressures by a least-squares fitting algorithm. The results show that residuals as large as 4×10^{-3} (i.e., $\sim 0.4\%$ of the transmission signal) occur near the absorption line center using VP, which is a known artifact of this parameterization (e.g., Tennyson et al., 2014), whereas with the qSDVP model fitting, the shape of the absorption line is correctly represented, and the residuals stay below 10^{-3} ($\sim 0.1\%$ of the transmission signal) at all wavelengths. This shows the improvement in terms of spectral accuracy resulting by the implementation of the qSDVP line shape model.

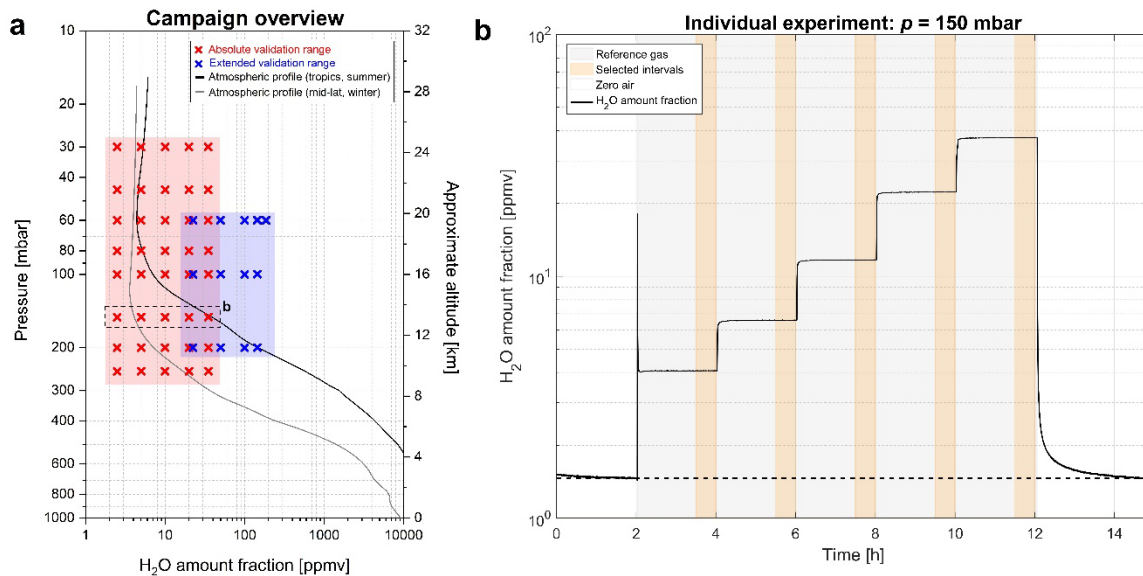


Figure 3. (Left) Overview of all H₂O setpoints and pressure levels investigated during the METAS validation experiments, overlaid with two atmospheric H₂O amount fraction profiles measured by CFH during recent field campaigns. The profiles correspond to moist, tropical summer conditions (black: Brunamonti et al., 2018) and dry, mid-latitude winter conditions (grey: Graf et al., 2021). (Right) Time-series of H₂O amount fraction measured by ALBATROSS during the experiment performed at 150 mbar.

Figure 4 (right) shows the accuracy assessment, i.e. the difference in H₂O amount fraction (in %) as function of pressure between the ALBATROSS measurements and the SI-traceable reference values, for all the investigated H₂O amount fractions. Results obtained with the VP line shape model are shown as open markers and dashed lines, while qSDVP results as solid markers and solid lines. This indicates that the VP model overestimates the H₂O amount fractions by 2–6 % compared to the SI-traceable reference. Conversely, all data points retrieved using the qSDVP model fall within the ± 1.5 % uncertainty range of the SI-traceable reference (grey shading).

The stability and precision of the instrument was systematically evaluated for each setpoint and pressure level using the Allan variance approach (not shown here). This indicates that ALBATROSS achieves a precision better than 30 ppb (i.e. 0.1 % at 35 ppm) at 1 s resolution at all conditions, which can be further improved to about 5 ppb (i.e. 0.02 % at 35 ppm) using integration time up to 50 s.

The METAS campaign also featured a number of additional experiments using synthetic air with higher H₂O concentrations than the permeator's upper range of 35 ppm ("extended-range validation", see Figure 3a). Their analysis shows an excellent linearity (within ± 1.5 %) of the spectrometer's retrieval up to amount fractions of 180 ppm, i.e. upper tropospheric conditions.

AquaVIT4. The AquaVIT4 (<https://www.hemera-h2020.eu/aquavit-4/>) intercomparison was conducted at the AIDA chamber (KIT, Germany), a large climate chamber (84 m³) able to simulate the atmospheric conditions of the UTLS in terms of temperature, pressure and H₂O amount fraction. Six instruments participated to the campaign: four prototype hygrometers under development for airborne platforms (balloon/aircraft), including ALBATROSS and two laboratory-based reference instruments. The airborne instruments included a balloon-borne laser spectrometer (PicoLight H₂O; CNRS, France), an airborne laser spectrometer (DLH; NASA, USA), and a balloon-borne frostpoint hygrometer (SAWfPHY; LMD, France). Reference instruments were the MBW373LX dewpoint mirror hygrometer (MBW Calibration, Switzerland), operated by KIT and the ApicT spectrometer integrated in the AIDA chamber.

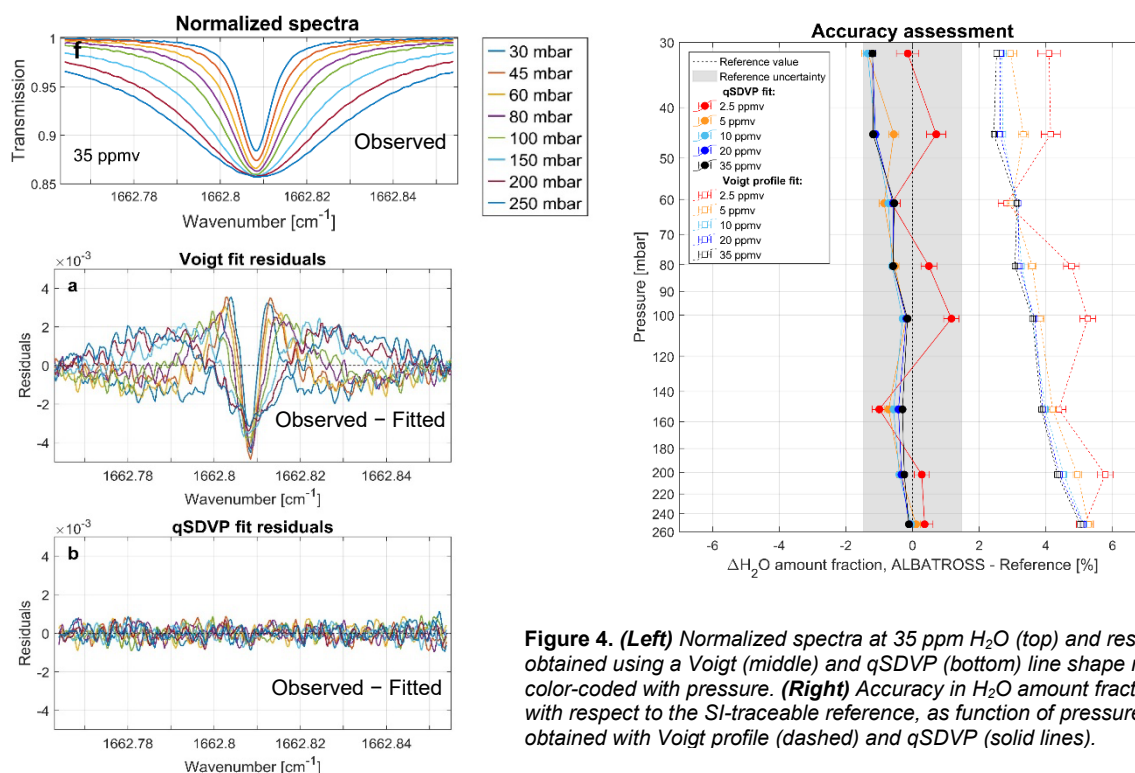


Figure 4. (Left) Normalized spectra at 35 ppm H₂O (top) and residuals obtained using a Voigt (middle) and qSDVP (bottom) line shape models, color-coded with pressure. (Right) Accuracy in H₂O amount fraction (%) with respect to the SI-traceable reference, as function of pressure, obtained with Voigt profile (dashed) and qSDVP (solid lines).

During the campaign a total of 10 days (~80 h) of measurement at UTLS-relevant conditions were performed, including 6 days of open testing and 4 days of *blind* intercomparison. The data of the *blind* intercomparison will be evaluated separately by each group and compared by an independent board of international referees (in progress). The simulated environmental conditions during AquaVIT4 ranged between pressures of 15–500 mbar, temperatures of 185–245 K, and H₂O amount fractions between 1–1000 ppm.

Figure 6 shows a timeseries of the AquaVIT-4 measurements performed on 2022-03-31, in terms of measured H₂O amount fraction from ALBATROSS (blue) and MBW373LX (red) (top panel) and their relative H₂O amount fraction difference (in %) (bottom panel). This demonstrates the excellent agreement (within ±5 %) between ALBATROSS and the reference instrument at H₂O amount fraction conditions relevant for the stratosphere (<10 ppm). The small-scale H₂O fluctuations, visible in both retrievals (due to the heating system of their common sampling line), further illustrates the capabilities of ALBATROSS to deliver high precision data even under fast varying humidity conditions. This is particularly remarkable considering the differences in weight (3.4 kg ALBATROSS vs. 40 kg MBW373LX) and power consumption (15 W vs. 300 W) between the two instruments.

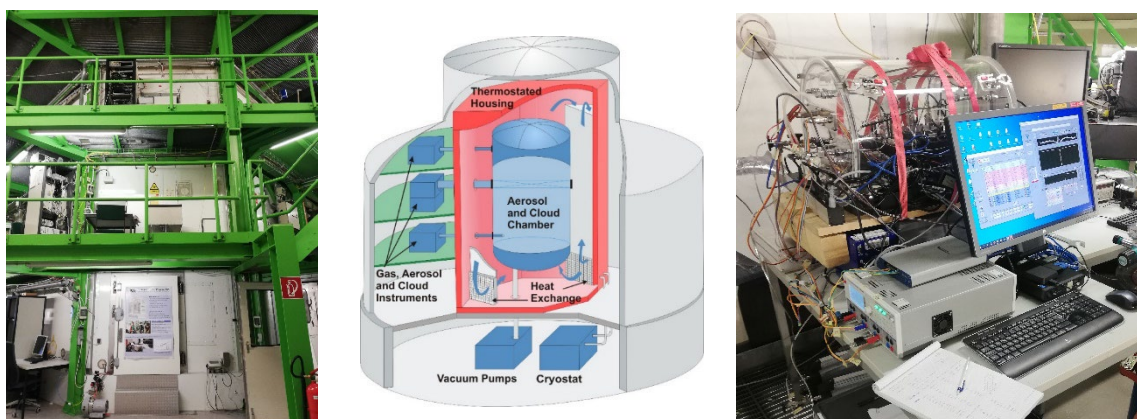


Figure 5. (Left) AIDA chamber hall. (Center) Schematics of the AIDA chamber, KIT, Germany (source: https://www.imk-aaf.kit.edu/AIDA_facilities.php). (Right) ALBATROSS experimental setup.

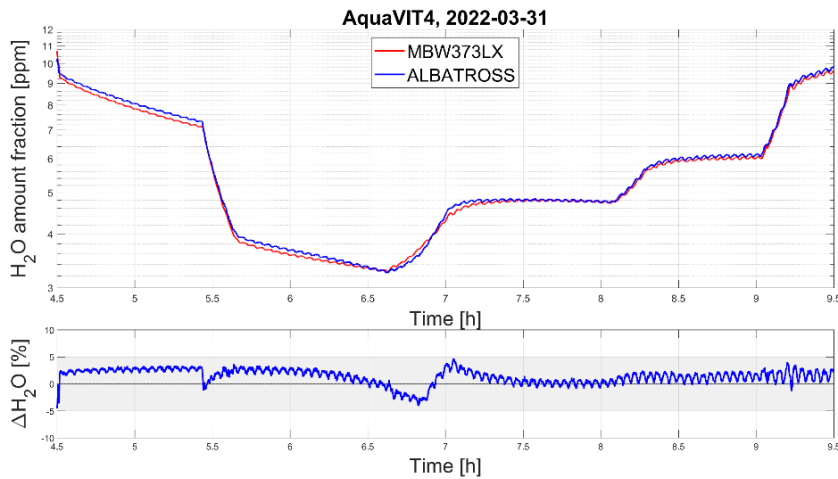


Figure 6. Time series of AquaVIT-4 measurements on 2022-03-31. (Top) H₂O amount fraction from ALBATROSS (blue) and MBW373LX (red). (Bottom) H₂O amount fraction difference (in %) between ALBATROSS and MBW373LX. The grey shading indicates the area of relative differences smaller than $\pm 5\%$.

WP3. A balloon test flight of the upgraded ALBATROSS spectrometer was performed at the Lindenberg Meteorological Observatory (Deutscher Wetterdienst, Germany) in September 2022. This was a tandem flight with a cryogenic frostpoint hygrometer (CFH, EnSci, USA) flown in parallel on a separate weather balloons. The ALBATROSS payload comprised, besides the spectrometer, an RS41 radiosonde (Vaisala, Finland) for pressure, temperature and RH measurements, and an additional GPS tracker for the recovery of the instrument after landing. The CFH payload also comprised an RS41 radiosonde and an aerosol backscatter sonde for cloud and aerosol measurements.

Both launches were conducted successfully and the two instruments reached the target burst altitude (ALBATROSS 29.5 km, CFH 33.4 km). The instrument performed well in terms of temperature stability and laser intensity during the entire flight duration, down to pressures of 13 mbar (burst point) and temperatures of $-63\text{ }^{\circ}\text{C}$ (tropopause). This also includes the passage within one liquid water cloud (at altitude 2.6 km) and two ice clouds (altitudes 10.2 km and 11.5 km) during the ascent of the balloon.

Figure 8 shows a preliminary evaluation of the vertical profiles of H₂O amount fraction measured by ALBATROSS (blue) and CFH (red) during the ascent of the test flight. In the upper troposphere (i.e. altitudes between 5 km and the tropopause), the ALBATROSS measurements show excellent agreement with CFH, including fine features in the vertical distribution of H₂O (e.g. the dry layers at 5–6 and 7–8 km), as well as in-cloud measurements. In the stratosphere, the ALBATROSS measurements show a high-frequency variability between approximately 4–10 ppm, hence above the stratospheric "background" H₂O content of ~ 4 ppm. It can be deduced from the analysis of the raw spectra (not shown here) that this signal is not due to an instrumental artifact, but rather to contamination by an external water vapor source (e.g., the balloon, the string connecting the payload to the balloon, or ice deposited inside the intake tube of the instrument). Nevertheless, we observe that the "minimum" H₂O amount fraction measured by ALBATROSS is in good agreement with the CFH measurements (at around 4 ppm) up to approximately 25 km altitude. The analysis of the stratospheric data and evaluation of the observed variability/suspect contamination are still ongoing.

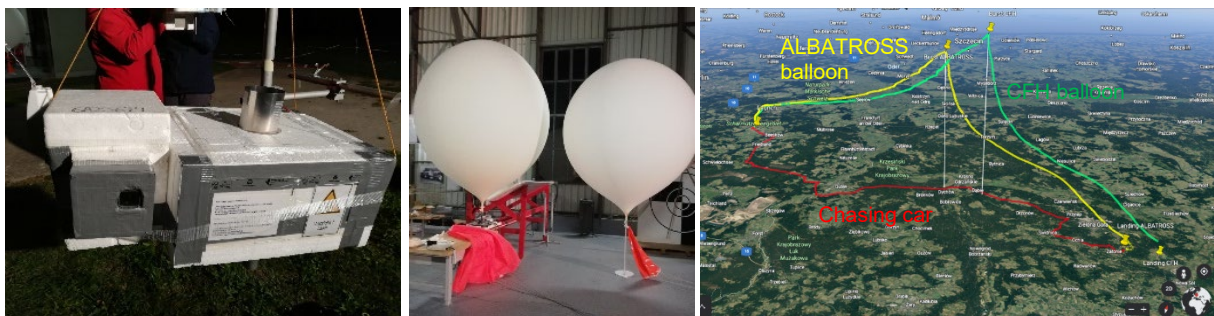


Figure 7. ALBATROSS spectrometer payload (left) and tandem balloons (center) ready for flight. (Right) 3D trajectory of the two balloons and the chasing car used for recovery (graphics: Google Earth, <https://earth.google.com/web/>).

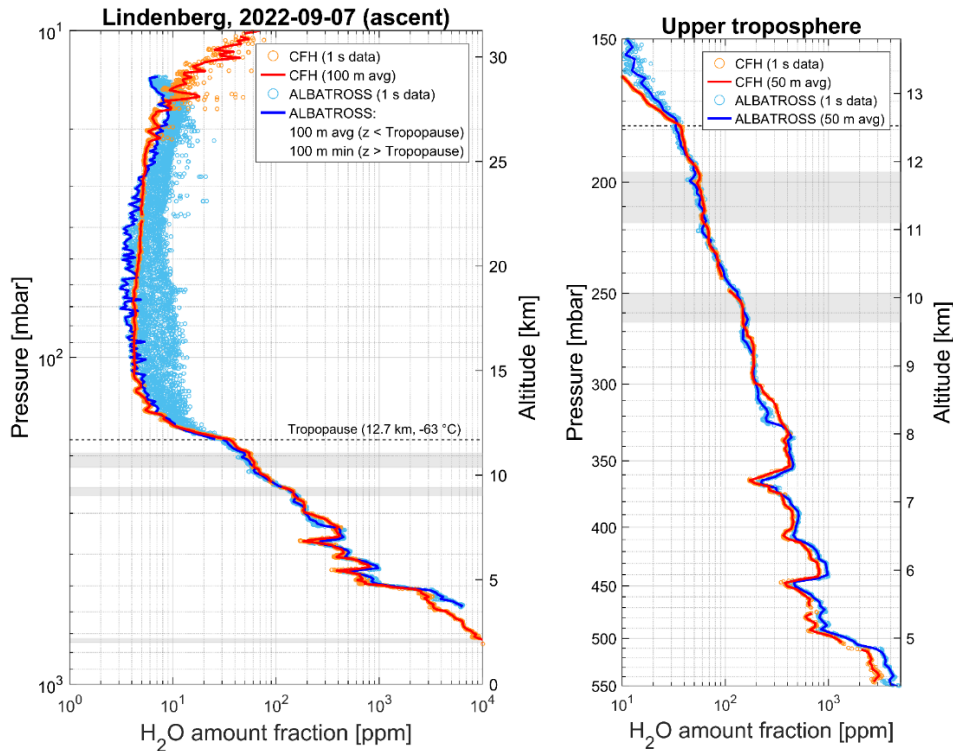


Figure 8. Preliminary evaluation of the H₂O amount fraction profiles measured by ALBATROSS (blue) and CFH (red) during the test flight performed in Lindenberg, Germany, on 2022-09-07 (left panel: entire flight, right panel: zoom into the upper troposphere region, ca. 4–14 km altitude). The observed tropopause level (at 12.7 km, temperature $-63\text{ }^{\circ}\text{C}$) is shown as a black dashed line. Raw data at 1 s resolution are shown as open circles, while the averages of 100 m and 50 m are shown as solid lines. For ALBATROSS, we show the average profile for altitudes below the tropopause, while for the altitudes above the tropopause we consider the profile across the lowest (i.e. "non-contaminated") values.

2.3 Results

Overall, our laboratory-based validation activities show that ALBATROSS achieves unprecedented accuracy and precision levels for a balloon-borne hygrometer. Particularly, the results of our validation campaign at METAS show an accuracy better than $\pm 1.5\%$ with respect to an SI-traceable reference source at a large variety of UTLS-relevant conditions, in terms of pressure (30–250 mbar) and H₂O amount fractions (2.5–35 ppm). The measurement precision was better than 30 ppb (0.1 % at 35 ppm H₂O) at 1 s resolution, and as low as 5 ppb (0.02 % at 35 ppm H₂O) upon integrating the data over 50 s (i.e., the Allan minimum). These results are particularly remarkable also considering the technical challenges associated with maintaining a stable H₂O amount fraction level in the low-ppm range in a laboratory setting, due to the strong surface adsorption/ desorption properties of H₂O.

A major improvement on the accuracy of ALBATROSS was achieved by implementing a quadratic Speed-Dependent Voigt Profile (qSDVP) line shape model for the spectroscopic retrieval algorithm, replacing the standard Voigt profile (VP). The molecular parameters required by this parameterization for our investigated H₂O line (not available in the literature) were determined experimentally using a multi-spectrum fitting approach. We demonstrated that the implementation of the qSDVP line shape mode improves the accuracy by 2–6 % compared to the VP model.

The results obtained at the AquaVIT4 intercomparison campaign further demonstrate that ALBATROSS achieves the same performance (in terms of accuracy, precision and response time) of a high-end laboratory instrument, such the reference MBW373LX dewpoint mirror hygrometer. This is particularly remarkable considering the differences in weight (3.4 kg ALBATROSS vs. 40 kg MBW373LX) and power consumption (15 W vs. 300 W) between the two instruments.

Finally, the results of the balloon test flight against CFH performed in Lindenberg in September 2022 show the capabilities of ALBATROSS under real atmospheric conditions. The instrument performed well during the entire flight duration, down to pressures of 13 mbar (burst point) and temperatures of $-63\text{ }^{\circ}\text{C}$ (tropopause), including the passage through one liquid water and two ice clouds, and shows excellent

agreement with the reference instrument until the tropopause (~13 km altitude). The stratospheric data also show consistent measurements with CFH until approximately 25 km altitude, although affected by a contamination due to an external water vapor source.

2.4 Conclusions and limitations

The results obtained within this project demonstrate the potential of mid-IR laser absorption spectroscopy, and specifically of our prototype, ALBATROSS, as an alternative reference method to cryogenic frostpoint hygrometry (CFH) for in-situ measurements of H₂O in the UTLS. This is particularly relevant considering the ongoing phasing out of the cooling agent required by CFH (fluoroform, HFC-23), which urges the need of an alternative solution to maintain the monitoring of UTLS H₂O in global, long-term monitoring networks, such as the GCOS Reference Upper Air Network (GRUAN).

The results of the test flight revealed a potential contamination by an external water source (which could be associated either to the balloon, the string connecting the payload to the balloon, or to other sources such as ice deposited in the intake tube of the instrument) in the stratosphere. The data are still being investigated to identify the source of the contamination. However, we are confident that such artifacts can be addressed in future flights, in order to complete the instrument's validation until mid-stratospheric altitudes of ~30 km (and potentially above).

2.5 Outreach work, publication of data and results

Conference contributions:

1. S. Brunamonti, M. Graf, L. Emmenegger and B. Tuzson: *Quantum-cascade laser absorption spectroscopy (QCLAS) for balloon-borne measurements of stratospheric H₂O*, European Geophysical Union (EGU) General Assembly 2021, 30 April 2021, (oral presentation, virtual meeting).
2. B. Tuzson, M. Graf, P. Scheidegger, H. Looser, A. Kupferschmid, and L. Emmenegger: *Up in the air! Trace-gas sensing aboard flying platforms*, Conference on Lasers and Electro-Optics/Europe (CLEO Europe), 21 June 2021, (oral presentation, virtual meeting).
3. S. Brunamonti, M. Graf, T. Bühlmann, C. Pascale, H. Looser, L. Emmenegger and B. Tuzson: *Absolute validation of a balloon-borne spectrometer for water vapor measurements in the upper atmosphere*, Swiss Chemical Society (SCS) Fall Meeting 2021, 10 September 2021, (oral presentation, virtual meeting)
4. S. Brunamonti, M. Graf, L. Emmenegger and B. Tuzson: *Absolute validation of a balloon-borne spectrometer for water vapor measurements in the upper atmosphere*, Swiss National GAW/GCOS Symposium, 13-14 September 2021, (oral presentation, virtual meeting)
5. S. Brunamonti, M. Graf, L. Emmenegger and B. Tuzson: *Quantum-cascade laser absorption spectrometer (QCLAS) for water vapor measurements in the upper atmosphere*, GRUAN (GCOS Reference Upper Air Network) Implementation and Coordination meeting (ICM-13), 16 November 2021, (oral presentation, virtual meeting)
6. S. Brunamonti, M. Graf, , L. Emmenegger and B. Tuzson: *Quantum-cascade laser absorption spectroscopy for water vapor measurements in the upper atmosphere*, GAW-CH Landesausschuss Spring Meeting, 4 May 2022, (oral presentation, ETH Zürich, Switzerland)
7. S. Brunamonti, M. Graf, L. Emmenegger and B. Tuzson: *Quantum-cascade laser absorption spectroscopy for balloon-borne measurements of UTLS water vapor*, European Geosciences Union (EGU) General Assembly 2022, 23 May 2022, (oral presentation, Vienna, Austria)
8. S. Brunamonti, M. Graf, L. Emmenegger and B. Tuzson: *Laboratory validation of a laser absorption spectrometer for balloon-borne measurements of UTLS water vapor*, NOAA Global Monitoring Annual Conference (eGMAC 2022), 26 May 2022, (oral presentation, virtual meeting)
9. B. Tuzson, S. Brunamonti, M. Graf, T. Bühlmann and L. Emmenegger: *QCL spectrometer for balloon-borne measurements of water vapour in the UTLS*, FLAIR (Field Laser Applications in Industry and Research) 2022, 12-16 September 2022, (poster presentation, Aix-les-Bains, France)
10. B. Tuzson S. Brunamonti, M. Graf, T. Bühlmann, L. Emmenegger and: *Laboratory validation of a*

laser absorption spectrometer for balloon-borne measurements of upper air water vapor, Swiss Chemical Society (SCS) Fall Meeting 2022, 8 September 2022, (oral presentation, Zürich, Switzerland)

11. S. Brunamonti, M. Graf, T. Bühlmann, L. Emmenegger and B. Tuzson: *SI-traceable validation of a laser absorption spectrometer for water vapor measurements in the upper atmosphere*, BIPM-WMO Workshop "Metrology for Climate Action", 26-30 September 2022, (poster presentation, virtual meeting)
12. S. Brunamonti, M. Graf, T. Bühlmann, L. Emmenegger and B. Tuzson: *Balloon-borne laser absorption spectroscopy for UTLS water vapor: ALBATROSS*, SPARC (Stratospheric Processes and their Role in Climate) General Assembly 2022, 24-28 October 2022, (poster presentation, ECMWF Reading, UK)

Peer-reviewed publications:

1. M. Graf, P. Scheidegger, A. Kupferschmid, H. Looser, T. Peter, R. Dirksen, L. Emmenegger, and B. Tuzson: *Compact and lightweight mid-infrared laser spectrometer for balloon-borne water vapor measurements in the UTLS*, Atmos. Meas. Tech., 14, 1365–1378, 2021 <https://doi.org/10.5194/amt-14-1365-2021>.
2. S. Brunamonti, M. Graf, T. Bühlmann, C. Pascale, I. Ilak, L. Emmenegger and B. Tuzson: *SI-traceable validation of a balloon-borne laser absorption spectrometer for water vapor measurements in the upper atmosphere*, in preparation for Atmospheric Measurement Techniques.

2.6 Outlook

The development and testing of ALBATROSS will continue in the framework of the "Swiss H₂O Hub" project (2023–2026), recently funded by MeteoSwiss in the context of GAW-CH. This collaborative project, involving a consortium of four Swiss institutions (Empa, ETH Zürich, University of Bern and MeteoSwiss), aims at providing high-quality measurements of atmospheric water vapor from the ground to the mesosphere (i.e., up to 80 km altitude) by combining the laser spectroscopy (ALBATROSS, Empa), frostpoint hygrometry (PCFH, ETH Zürich), microwave radiometry (MIAWARA, University of Bern) and Raman lidar (RALMO, MeteoSwiss) techniques.

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