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HAI – A new, airborne, absolute, twin dual-channel, multi-phase TDLAS-hygrometer

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Abstract

The novel **H**ygrometer for **A**tmospheric **I**nvestigations (HAI) realizes a unique concept for simultaneous gas-phase and total (gas-phase + evaporated cloud particles) water measurement. It has been developed and successfully employed for the first time on the German HALO research aircraft. This new instrument combines direct Tunable Diode Laser Absorption Spectroscopy (dTDLAS) with a first-principle evaluation method to allow absolute water vapor measurements without any initial or repetitive sensor calibration using a reference gas or a reference humidity generator. HAI contains two completely independent dual-channel (closed-path, open-path) spectrometers, one at 1.4 µm and one at 2.6 µm, which allow together to cover the entire atmospheric H_2O range from 1 to 40 000 ppmv with a single instrument. Both spectrometers comprise each a separate, wavelength-individual extractive, closed-path cell for total water (ice and gas-phase) measurements. Additionally, both spectrometers couple light into a common, open-path-cell outside of the aircraft fuselage for a direct, sampling-free and contactless determination of the gas-phase water content. This novel twin dual-channel setup allows for the first time multiple self-validation functions i.e. in particular a reliable, direct, in-flight validation of the open-path channels. During the first field campaigns, the in-flight deviations between the independent and calibration-free channels (i.e. closed-path to closed-path and open-path to closed-path) were on average in the 2% range. Further, the fully autonomous HAI hygrometer allows measurements up to 240 Hz with a minimal integration time of 1.4 ms. The best precision is achieved by the 1.4 µm closed-path cell at 3.8 Hz (0.18 ppmv) and by the 2.6 µm closed-path cell at 13 Hz (0.055 ppmv). The requirements, design, operation principle and in-flight performance of the hygrometer are described in this work.

1. Introduction

Water vapor is in many ways one of the most important measurand for atmospheric investigations (Ludlam, 1980; Möller et al., 2011; Ravishankara, 2012). Water is the most important greenhouse gas (Kiehl and Trenberth, 1997), and known as a key atmospheric coupling element of almost all microscopic (e.g. droplets/ice crystals formation), macroscopic (e.g. clouds/precipitation) and global processes (e.g. hydrological cycle). Therefore, it is strongly related to the numerous highly relevant topics of atmospheric science and closely related to "climate change" (Held and Soden, 2000; Houghton, 2009; Kiehl and Trenberth, 1997; Maycock et al., 2011). Unsurprisingly, numerous water vapor studies have been carried out targeting its atmospheric trends and variability (Lu and Takle, 2010; McCarthy et al., 2009; Ross and Elliott, 1996; Scherer et al., 2008; Trenberth et al., 2005; Xie et al., 2011), its influence on transport models (Kiemle et al., 2012; Schäfler et al., 2010), or its impact on radiation balance models (Lockwood, 1990; Ramanathan et al., 1989; Schneider, 1972). One reason for the high complexity of atmospheric water vapor is that it is one of the few atmospheric molecules that appears in all three phases. Water in the gas-phase is a very strong infrared absorber and significantly impacts atmospheric energy fluxes through latent heat transfers by the different phase transitions. Condensation to the liquid phase or freezing to solid particles leads to effective scattering of solar radiation, which directly raises links to the formation process of cirrus clouds (Krämer et al., 2009; Spichtinger et al., 2004). These relationships show the complexity from a theoretical as well as a modeling point of view. Today, however, the quality e.g. particularly accuracy and comparability of atmospheric water measurements frequently limit a better understanding of key atmospheric processes (Krämer et al., 2009; Peter et al., 2006; Scherer et al., 2008; Sherwood et al., 2014). Despite the outlined importance and the large effort invested in the developments of hygrometers in recent years, water vapor remains a target molecule that is very difficult to measure accurately.

Several major issues exacerbate water vapor measurements. Atmospheric water vapor encompasses a very large concentration range: 3 – 40 000 ppmv from troposphere up to the lower stratosphere. The spatial fluctuations of H_2O in the atmosphere are high, which leads on fast aircraft (approx. 800 km/s cruising speed) to highly dynamic H_2O variations of up to several 1000 ppmv/s in the gas-phase and up to several 10 000 ppmv/s for total water measurements. These issues require, especially for aircraft based hygrometers, very high time resolution in combination with very high precision and accuracy. Additionally, since water vapor readily changes from one phase to the other, it would be extremely helpful if hygrometer were able to differentiate between the phases or could differentiate at least between water vapor and total water in order to minimize systematic uncertainties caused by the sampling process. Last but not least, water vapor is very effectively absorbed from nearly any surface. This challenges in a highly complex manner not only the entire gas sampling system but also the calibration infrastructure which is typically required for most hygrometers. By waiving the entire calibration process, special hygrometers (Wolfrum et al., 2011) circumvent all calibration related issues efficiently which will explained in chapter 3.2.

This brief compilation illustrates the complex challenges associated with developing a water vapor instrument, especially if it should be able to measure in tropospheric and stratospheric atmospheric conditions. Numerous (mostly single-channel) hygrometers have been developed in the last decades with various advantages and drawbacks (see (Wiederhold, 1997) and e.g. (Buck, 1985; Busen and Buck, 1995; Cerni, 1994; Desjardins et al., 1989; Diskin et al., 2002; Durry et al., 2008; Ebert et al., 2000b; Gurlit et al., 2005; Hansford et al., 2006; Helten et al., 1998; Hunsmann et al., 2008; Karpechko et al., 2014; Kley and Stone, 1978; May, 1998; Meyer et al., 2015; Ohtaki and Matsui, 1982; Roths and Busen, 1996; Salasmaa and Kostamo, 1986; Schiff et al., 1994; Silver and Hovde, 1994b, 1994a; Thornberry et al., 2014; Webster et al., 2004; Zöger et al., 1999a, 1999b)). Consequently, the question should be raised from the opposite point of view: What are the important and required properties to be covered and combined for a near-universal “Hygrometer for Atmospheric Investigation” to serve as an innovative and cutting-edge tool to explore open and new scientific questions related to atmospheric water vapor?

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2. Requirements for HAI, a "Hygrometer for Atmospheric Investigation"

2.1. Specific instrumental boundary conditions defined by the atmosphere

Currently, atmospheric water vapor data are generated from two instrument types: Remote sensing approaches (like satellites (Oelhaf et al., 2004) or ground-based – often FTIR based - monitoring stations (Zachariassen et al., 2003)) or in-situ hygrometers deployed directly inside of the environment to be measured. The latter are roughly separated in ground-base "weather stations", balloon-borne radiosondes (Miloshevich et al., 2006) and hygrometers on airborne carriers like aircraft (Marenco et al., 1998). While ground-based stations permanently provide local information, radiosondes deliver "only" one point like vertical profile per ascend/ descend but for heights up to 50 km above ground. Airborne vehicles like aircraft and helicopters, however, combine the possibility for arbitrary flight paths and localized measurements, e.g. for cloud investigations, with a beneficial long operation range. In particular the most modern research aircrafts such as the German HALO aircraft (Anon, 2014; Krautstrunk and Giez, 2012) or the US American HIAPER (Anon, n.d.) bridge the gap and combine broad spatial coverage (> 10 000 km), high altitudes (up to 15 km), and large payloads of up to 3000 kg payload, in a favorable pressurized, and air-conditioned cabin. The relatively high traveling speed of these aircraft (of up to 230 m/sec), however, also causes disadvantageous influences on the measurements themselves, which are difficult to take care of compared to the very slowly moving (low m/sec range), quasi-static balloon-borne radiosondes. Even relatively simple meteorological variables such as air temperature and pressure need complex retrieval algorithms (Giez, 2012) if measured on a high speed aircraft. In short, aircraft in the contemporary working equipment are indispensable for both the investigation of spatially confined effects too small to retrieve from remote sensing data and the validation of the remote sensing instruments (Oelhaf et al., 2004). It is highly desirable for an airborne system to be constructed in a way that it operates on the ground in the same manner as in flight and that every change of environment and "boundary conditions" of the instrument is logged. This allows, besides multipliable deployments e.g. in ground based stations, extensive validation as well as laboratory comparisons with other instruments and avoids systematic, barely detectable deviations only occurring in flight. This notion related to water vapor leads directly into the everlasting discussion about sampling via open-path systems operation in the free flow versus closed-path systems extracting the air to be analyzed into the instrument. Open-path hygrometers offer numerous great benefits such as: the prevention of any sampling errors or uncertainties (caused by surface absorbing effects) as well as the high response time that is limited only by the transfer functions of optical or electrical components but not by the gas exchange rate. The latter circumvents the complicated and adulterant deconvolution of smoothing effects with time-response functions caused by a sampling system. On the other hand, the boundary conditions such as gas pressure and gas temperature as well as possible spatial inhomogeneities of both parameters are difficult to accurately take into account for an open-path system. Additionally, an airborne open-path sensor has to operate in harsh boundary conditions, i.e. over a large range of temperatures (-80 to 50 °C), pressures (70 to 1000 hPa), for large ram pressures (900 km/h gas velocity) and mechanical stress through accretion of ice or liquid water.

The major problem of all present open-path systems is their highly complex calibration, or even just validation, since realistic flight conditions, in particular the dynamics, are extremely difficult to realize in the lab with sufficient accuracy. A direct metrological link to test the inflight performance, e.g. a dynamic calibration facility for open-path hygrometers, is therefore missing.

Closed-path systems, on the other side, are simply installed inside an air-conditioned cabin, in a much more protected environment. Gas is sampled with a suitable inlet and led via a tubing system to an "internal" measurement chamber, such as an optical absorption cell or a suitable "cavity", e.g. for a dew point mirror hygrometer. On one hand, it is much easier to accurately control and maintain the physical boundary conditions of the sample gas, e.g. temperature, pressure, flow, etc. in the measurement volume. On the other hand, it is difficult to ensure and maintain a representative sampling process and to quantify and correct sampling related deviations. These maybe caused by adsorption and desorption effects, which occur on all surfaces of the sampling system, and have to be carefully minimized by heated (HAI ≈80 °C) stainless steel sampling pipes, along with an instrument design ensuring high gas flows (HAI ≈100 liter/min) under all flight conditions.

Gas-phase H_2O measurements in clouds are often carried out via backward facing sampling inlets. But such inlets are readily sampling small liquid or solid water particles possibly causing systematically positive offsets. For every H_2O measurement using airborne extractive (=closed-path) instruments on aircrafts, it needs to be taken into account that the instruments response reflects contributions from the sensor element itself as well as the sampling/tubing system and their dynamic properties. One indisputable, major advantage of typical extractive instruments is the possibility for careful tests outside of the aircraft, i.e. in a hangar or laboratory. But, to take full advantage of this, it is desirable for a sophisticated instrument to integrate supervising and monitoring functions in a way that a performance comparable to the laboratory can be ensured during any inflight situation. This generates the great benefit of transferring the performance from the laboratory to the field situation, quite similar how a metrological transfer standard is typically used. This directly reinforces the question, how to assess the accuracy of airborne hygrometers.

2.2. Accuracy and state of the art instrumentation

In general, the highest measurement and preparation accuracy is realized by the validated primary standards of national metrology institutes such as PTB (Germany) or NIST (USA). The international, metrological water vapor scale is defined by traceable primary water vapor generators (Brewer et al., 2011). The mixing ratio range, required to cover the entire tropospheric and stratospheric coverage of about 3 - 40000 ppmv, is realized by a combination of generators based on different physical principles. Their typical uncertainty is in the order of 0.5% relative, but varies with the physical principle used and their realization (Brewer et al., 2011; Buchholz et al., 2014a; Mackrodt, 2012). In other words, it is not possible to validate any hygrometer with better accuracy due to the lack of a suitable accurate reference.

Comparing the available metrological accuracy to some results from field comparisons of airborne hygrometers demonstrates the large potential for improvement. For example, long-term (> 10 years) change studies of stratospheric H_2O (Oltmans and Hofmann, 1995; Rosenlof et al., 2001; Solomon et al., 2010) suffer from significant, difficult to quantify relative deviations between different instruments in the range of 50–100% (Fahey et al., 2014; Peter et al., 2006; Vömel et al., 2007) which recent studies such as (Rollins et al., 2014) confirm (±40–50% for < 3ppmv and ±20% for > 3ppmv). Radiosonde comparisons with polymer sensors and chilled dew point mirror hygrometer, covering the entire troposphere and lower stratosphere region such as (Miloshevich et al., 2006), show averaged overall agreement in the 10% range (but also, local deviations in the 30% range). These deviations are quite common for many airborne campaign results e.g. (Smit et al., 2014) and become even worse when focusing on the relative deviations in regions containing highly variable H_2O structures. Hence in 2007, an international comparison exercise, "AquaVIT" (Fahey and Gao, 2009), was organized to compare the world's best airborne hygrometers under well-controlled, quasi-static, equivalent conditions to evaluate the accuracy under well controlled laboratory conditions, without the influence of any typical airborne sampling and dynamic effects. AquaVIT comprised 22 hygrometers (tunable diode laser spectrometers, TDL, dew or frost point mirror hygrometers, D/FPH, Lyman alpha fluorescence and absorption hygrometers, LAFH and other principles) from 17 international research groups. The instruments were categorized in well validated "core" instruments (APicT, FISH, FLASH, HWV, JLH, CFH, see (Fahey and Gao, 2009) for details) and "younger, less mature" non-core instruments. Even the core-hygrometers deviated in the important 1 to 150 ppmv WVMR range by up to ± 10 % from the mean. In other words, core instruments differed by up to 20% from each other, even under quasi-static conditions. Other, less representative and extensive comparisons such as (Hoff, 2009; Mangold and Wodca Team, 2003) yielded similar results.

Of course, the assessment of the required accuracy depends strongly on the purpose of the data. In terms of climatologies and strongly averaged or coarse validation studies such deviations can be acceptable, but as far as e.g. retrieval models for satellite data improve, such uncertainties and deviations can become critical. In many other cases such as the currently often discussed atmospheric super saturations (Peter et al., 2006), the instrument uncertainties prevent deeper investigations and therefore a better understanding. Reconsidering the entire situation and perceiving that after so much development effort over the past decades these deviations remain quite high, leads to the inevitable question of the concealed, common impact factors. Contemplating the typical metrological efforts needed at national metrology institutes (NMI) to generate an accurately humidified gas stream (with a sub-percent uncertainty) suggests that the uncertainties generated by typical calibration processes under field conditions could be a major contribution to these hygrometers deviations found in AquaVIT and other studies. In particular, comparing the performance and strategies of lab-based, metrological and portable field calibration facilities (Friehe et al., 1986; Helten et al., 1998; Podolske et al., 2003; Smit et al., 2000; Smorgon et al., 2014; Zöger et al., 1999b) show three significant discrepancies: required time for calibration, frequency of calibration, and traceability of the humidity reference itself. Calibrations in the low concentration ranges at NMIs take several hours up to days per individual humidity value. During airborne campaigns, however, calibrations often *have* to be realized (for practical reasons) in a short time, certainly less than a few hours (max) for a large number of concentration steps often including several pressure levels, thereby taking the obvious risk that the instrument/reference is not fully stabilized or equilibrated. Ideally, the time between two calibrations should be shorter than the expected time required for a drift/change exceeding the boundaries of the instrument uncertainties. Some airborne instruments require for the same reason calibrations before and after each flight in order to interpolate between both calibrations (Zöger et al., 1999b). Some even work with in-flight calibrations (Kaufmann et al., 2016) sacrificing measurement time and shifting the accuracy issue to the necessary airborne H_2O-source. Undoubtedly, many of these instruments have benefits e.g. in terms of precision, space, weight, prime cost etc., which justifies the calibration effort. However, vice versa, it is condoned that the calibration process is hampered and turns out to be the major influence on the accuracy of such a sensor.

Lastly, it seems necessary to implement a traceable link to the metrological humidity scales to improve the overall accuracy of airborne hygrometry (Joint Committee for Guides in Metrology (JCGM), 2009). By realizing an unbroken chain of calibrations, it is possible to link the instrument performance and the metrological water scale respectively the SI system of units. This also guarantees an accurate measurement/generation value with defined uncertainties.

To summarize: Fulfilling all these demands in the field similar like in a NMI laboratory is a tough task. However, as discussed later, many of the covered issues can be circumvented using first principle techniques like dTDLAS (Ebert and Wolfrum, 1994; Schulz et al., 2007) to realize optical, absolute hygrometers which avoid over certain defined operating ranges any water vapor sensor calibration.

From a user's point of view, precision and response time of an airborne hygrometer appears equivalent to accuracy if he is interested in fine structure resolving data. Precision and response time are under certain circumstances reciprocally correlated to each other (Allan, 1966). Typical figures for response time of airborne hygrometer in the literature are 0.5 – 1 Hz (Petersen et al., 2010; Szakáll et al., 2004; Zöger et al., 1999b); some instruments deliver faster data 4 Hz (Weinstock et al., 1994) or special instruments up to 25 Hz (Zondlo et al., 2010). Typical precisions at 1 Hz are in 0.1-0.2 ppmv range (Sargent et al., 2013; Zöger et al., 1999b; Zondlo et al., 2010). While in the stratosphere (<10 ppm), the precision certainly can become a limiting factor. This is much less the case inside clouds or within the troposphere, where frequent, very strong spatial variations (up to 1000 ppmv per 100 m flightpath in the gas-phase, or up to 20000 ppmv per 100 m during total H_2O phase) pose a larger problem. An instrument with a time response of just a few Hz causes significant under-sampling which can lead to strong aliasing effects at the high velocities (approx. 700-900 km/h) of many research aircrafts. Important under such conditions is the instrument's linearity to accurately cover the entire H_2O concentration range up to five magnitudes for total water vapor measurements.

Numerous additional requirements have to be fulfilled by an airborne instrument to ensure a successful operation. Due to the high operation costs for aircrafts and the high scientific demand, H_2O data have to be measured continuously without any interruptions. The instrument thus has to be highly reliable, robust, and require low maintenance. The restrictions in weight and space as associated with operation on an aircraft result in the necessity for the device to be of a compact and lightweight construction. The utterly complex and mandatory certification process (at least in Germany) quickly enforces an instrument design freeze before a campaign; this results in very stiff constraints for improvements/repairs during a campaign.

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3. Design decisions and approach

HAI reflects on these by providing four independent but coupled spectrometers in one single housing and by simultaneously combining open- with closed-path measurements in one single instrument. For all channels the evaluation is done with one common spectroscopic method: calibration-free direct Tunable Diode Laser Absorption Spectroscopy (cal-free dTDLAS).

3.1. Direct Tunable diode laser absorption spectroscopy (dTDLAS)

The requirements for fast measurements and high chemical selectivity in combination with a robust and small system calls to choose a contact-less, spectroscopic (hence optical) measurement technique rather than contact -mediated sensing methods such as dew point mirror hygrometer (DPH) or capacitive polymer sensors e.g. "Humicap" (Salasmaa and Kostamo, 1986; Smit et al., 2000). These are quite often used in air-borne hygrometry and especially in meteorological environments (Anon, 2010; Busen and Buck, 1995; Hansford et al., 2006; Wiederhold, 1997). Optical hygrometers can be set up to become quite immune to hydrophilic/hydrophobicsubstances (unavoidable in the vicinity of aircraft) as well as particles (dust, soot, ice, etc.) contained by the gas to be analyzed. These capabilities were e.g. extensively demonstrated via measurements inside of combustion processes in industrial power plants (Ebert et al., 2000a; Schlosser et al., 2002; Sun et al., 2013; Teichert et al., 2003). Even response times in the several 10 kHz range could be demonstrated recently by (Witzel et al., 2013) for measurements in combustion engines.

Tunable diode laser absorption spectroscopy (TDLAS) is a powerful as well as versatile diagnostic tech-nique which is frequently employed in the near infrared spectral range and led to numerous applications in atmospheric hygrometry (Diskin et al., 2002; Fahey and Gao, 2009; Gurlit et al., 2005; May, 1998; Schiff et al., 1994; Thornberry et al., 2014) . Advantageous diode lasers properties are the very high spectral resolution and power density, the continuous tuneability in combination with interesting technical features such as low cost, very low size/weight/power consumption, long life time, excellent beam quality, and optical fiber coupling to name just a few.

The typical setup and working principle of a TDLAS instrument has been frequently described in detail (Lackner, 2011; Schiff et al., 1994; Schulz et al., 2007; Werle, 1998). Therefore, only the HAI design relevant topics are discussed here. Important for an understanding of the novel HAI instrument is the classification of TDLAS instruments by their optical detection schemes in classical single- (Ebert et al., 2000b) or multi-path (Gurlit et al., 2005; Hunsmann et al., 2008; Lübken et al., 1999; May, 1998; McManus et al., 1995) beam setups. Further major categorizing distinguishes between the wavelength-modulation schemes like single modulation frequency = direct TDLAS or dTDLAS (Ebert and Wolfrum, 1994) or double modulation schemes like wavelength modulation spectroscopy WMS (Podolske and Loewenstein, 1993; Silver, 1992; Silver and Hovde, 1994a; Silver and Zondlo, 2006; Vance et al., 2011; Webster et al., 2004). WMS which is often used for very compact airborne sensors, provides on the first glance higher sensitivities by using lock-in technologies to efficiently filter noise. This, however, sacrifices the possibility of direct physics-based quality and reliability checks, since the actual measured WMS raw signal contains less spectral information than a dTDLAS raw signal. This aggravates or sometimes even prevents detailed signal analysis based on fundamental physical explanations. Using dTDLAS instead with a special, but less common, first principles evaluation procedure (Ebert and Wolfrum, 2000; Farooq et al., 2008; Mihalcea et al., 1997; Schulz et al., 2007) yields a sophisticated evaluation, characterization, and quality management and a holistic view on the physical principles behind the data. This circumstance can even be used to avoid typical calibration procedures with reference gas standards.

Explanation of the term "calibration-free"

The term "calibration-free" is often used in different communities with dissimilar meanings. To distinguish one should consider how calibration is defined by metrology (JCGM 2008, 2008): "calibration (…) in a first step, establishes a relation between the measured values of a quantity with measurement uncertainties provided by measurement standards (…), in a second step, uses this information to establish a relation for obtaining a measurement result from an indication (of the device to be calibrated)". In other words, an instrument with a deterministic relation between indication and quantity can be calibrated, without knowing the physics behind it. This particularly allows for compensating of non-linearity, offsets, drifts or response changes over time as long as they are stable, predictable, or can be extrapolated. We use the term "calibration-free" to emphasize that HAI does not rely on such a correction process. The hygrometer described in this paper has never been calibrated or adjusted to a water vapor primary standard. Of course, parameters like gas pressure and temperature that are used for the calculation of the water vapor content via a first principle model are measured with calibrated sensors. This is done from a practical point because a) primary standards for temperature and pressure are by themselves large facilities and b) the influence in the final uncertainty budget doesn't justify that approach. Calibration-free doesn't mean that the whole setup only uses primary principles. The whole idea behind traceability (JCGM 2008, 2008) is to use other sizes, higher in the hierarchy of the SI units of course, to generate/analyze the target value. Metrologists name that an unbroken chain of measurements. To visualize, this means that the first water vapor value delivered by the HAI instrument is the final value. Everything has to be characterized in advance on such a level that the first measurement value is determined within its uncertainty limits. Therefore, since there was no calibration, it's termed *calibration- free*.

Obviously, the calibration-free approach can always be enhanced with a calibration (such as (Muecke et al., 1994)) at any time (Buchholz et al., 2013b), even after a campaign if this seems advantageous, since the requirements for a calibrated instrument are lower than for a calibration-free instrument.

3.2. Non-calibrated direct TDLAS (dTDLAS)

The principle of non-calibrated absolute dTDLAS is very briefly presented in the following section. More detailed information regarding TDLAS is referred to in the above-mentioned literature. The sketch in Figure 1 shows the schematics of a dTDLAS spectrometer with two independent channels. For low light intensities $I_0(\lambda)$ in the mW-range, the transmitted light $I(\lambda)$ can be described by the extended Lambert-Beer equa-

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tion (Equation 1) including possible disturbances or the absorption measurement by background radiation E(t) or broadband transmission losses Tr(t).

Equation 1: $I(\lambda) = E(t) + I_0(\lambda) \cdot Tr(t) \cdot exp[-S(T) \cdot g(\lambda - \lambda_0) \cdot N \cdot L]$

By applying the ideal gas law Equation 1 can be used to retrieve the H_2O volume mixing ratio c.

Equation 2: $$c = -\frac{k_B \cdot T}{S(T) \cdot L \cdot p} \int ln\left(\frac{I(\nu) - E(t)}{I_0(\nu) \cdot Tr(t)}\right) \frac{d\nu}{dt} dt$$

The *amount fraction* c is in metrological units officially specified as [mol/mol = mol absorber per mol gas] which is in the environmental community better known as "volume fraction" e.g. in units of ppmv or Vol.-%. The term $\frac{d\nu}{dt}$ is called the *dynamic tuning coefficient* of the used laser. It can be determined experimentally and is directly linked to the SI units (length) by using the Airy signal of the laser light passing through a planar, air-spaced etalon (Ebert and Wolfrum, 2000; Schlosser et al., 2002). Yet, unpublished, on-going, and long-term $\frac{d\nu}{dt}$ measurements for the used 1.37 µm DFB laser type over several years indicate a long-term stability of its tuning characteristics better than 1%, which is within the current uncertainties of the tuning characterisation. k_B is the *Boltzmann constant* and L is the *optical path length*. S(T) is the *line strength* of the selected molecular transition (see chapter below) and therefore a physical property of the molecule to be measured. The gas *pressure* (p) and gas *temperature* (T) can be accurately acquired in a closed-path cell; the quality of the respective measurements in the open-path cell are discussed in the following chapter describing the construction of the open-path sensor. Equation 2 also "explains" the term calibration-free quantitatively as there are no other "hidden" parameters used to derive the water vapour concentration which require a calibration.

3.3. Absorption line selection

Suitable absorption lines have to be selected for a specific application by several criteria (Wunderle et al., 2006) (Wagner et al., 2012). Besides a line strength maximization to ensure high sensitivity, other important parameters have to be taken into account. For atmospheric measurements, the cross sensitivity to other gases such as CO_2 needs to be minimized. This ensures a better control of the fitting process due to the fewer degrees of freedom. Similarly, the line should be isolated from other lines to simplify the retrieval of the baseline function. For the open-path measurements, it is highly important to minimize temperature dependence of the line strength in order to minimize the influence of gas temperature uncertainties. Lastly, sometimes the primary constraint is the availability of suitable laser diodes and additional accessories such as fibers and optic components. As the certification for airborne instruments nearly prevents improvements/repairs during a campaign, all components need to be very reliable. For HAI, we selected two specific water lines at 2596 nm and 1370 nm in two wavelength ranges, which also had been used before (Buchholz et al., 2012, 2013b; Ebert, 2006; Ebert et al., 2004; Fahey and Gao, 2009; Hovde et al., 2001; Hunsmann et al., 2008; May, 1998; Seidel et al., 2012; Witzel et al., 2012; Wunderle et al., 2008) and for which improved spectral parameters were generated (Hunsmann et al., 2006). The 2.6 µm laser is not fiber-coupled, doesn't have an optical isolator, is less stable in terms of temperature fluctuations and has a lower

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beam quality, but accesses a factor of 20 stronger line and thus ultimately promises 20 x higher sensitivity. Both lines are shown (Figure 2) as a simulation based on the HITRAN (Rothman et al., 2009) database. Assuming an optical resolution of 5.10^{-4} OD, the hygrometer is expected to provide for a 1.4 µm closed-path cell (assuming a 1.5 m path length) under atmospheric conditions a water vapor concentration range from 10 to 40 000 ppmv and for the 2.6 µm closed-path channel from 0.5 to 2000 ppmv. The lower limit will finally be defined by the capability of minimizing and compensating the effects of parasitic H_2O offsets (Buchholz and Ebert, 2014b) and their related uncertainties. For the 2.6 µm channel, the upper limit depends strongly on the retrieval quality of the baseline. This means e.g. under lower pressure conditions (<500 hPa) evaluations up to 40 000 ppmv can be performed but for high pressures the absorption line at these high concentrations is saturated and so broadened that the baseline cannot be retrieved with a low uncertainty. The upper limit in general, i.e. also for the 1.4 µm channels, is defined by the dew point related to the instrument temperature; 40 000 ppmv is equal to 100 %RH at 28.5 °C and 1013 hPa.

4. HAI configuration

HAI can be installed on an aircraft in several different ways and configurations. This paper focuses on the installation during the first HALO (Krautstrunk and Giez, 2012) campaigns (TACTS (Engel et al., 2013) and ESMVal (Schlager, 2014)) which targeted multiphase H_2O measurements. Figure 3 (right) shows the fuselage of HALO with the two mountings belonging to HAI. In the very front is a trace gas inlet (TGI), where HAI uses the second opening from the top to sample air in flight. The sampling height is approx. 25 cm above the aircraft skin. Next to the TGI is the HAI open-path cell (OPc), which is a fiber-coupled, White-type three-mirror configuration mounted on two pylons. The mirrors fold the laser light beam shown in the middle (yellow). Both the TGI and the OPc are installed on HALO specific aperture plates. Figure 3 (left) depicts the cabin installations with the golden instrument in the middle being the rack of the HAI main unit. The black insulated ½" pipes on the right side of the rack are the inlet lines providing the air sampled by the TGI to the main unit. These pipes are heated (70 °C) to evaporate ice particles and droplets in the gas stream at a flow of approximately 100 volume liter/min. The open-path sensor is not visible in this photo since it is on the left side of the rack.

4.1. HAI main unit

The HAI main unit is comprised of individual modules except for some electronics of the OPc which are directly mounted cabin sidewise on the OPc: preamplifier for detectors, five temperature measurement converters, mirror heating controller, and two pressure sensors. Many of the modules contain innovative developments, which are or will be published individually to prevent an overload of technical details in this paper. The important functions/modules to understand the HAI concept are as follows:

a. Automatic line identification and spectral locking to compensate temperature and electrical influences as well as drifts
b. Internal mechanism (Buchholz and Ebert, 2014b) to minimize and compensate effects from parasitic absorption which usually leads to variable offset problems in spectrometers like HAI

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c. Gas handling system which tempers the air to be analyzed to the temperature of the instrument and therefore minimizes the risk of temperature inhomogeneity even at high gas flows as well as to provide detailed information about temperature, pressure, and gas flow in and through the system
d. Several individual sensors and electronics to collect in total more than 120 system parameters about the HAI system's health
e. Sequence control system for steering, controlling, and exception treatment of HAI with all its submodules

4.2. HAI's closed-path cells (CPc)

Core parts of the HAI main unit are the extractive optical closed-path cells (Figure 4) installed in the individual 1.4 µm and 2.6 µm optics modules within the HAI main unit. Both optics modules are completely independent, and both cells use a new, highly compact miniature White type (White, 1976) design that folds the laser beam (total approx. 1.5m) with three mirrors (approx. 7.5 cm distance). The 1.4 µm cell is an improved fiber-coupled version of the White cell in (Kühnreich et al., 2013). Combined with a new fiber fed through (Buchholz and Ebert, 2014a), it reaches a very low leakage rate of $1.9 \cdot 10^{-6}$ hPaL/s and prevents parasitic offsets due to ambient air. Three mirrors, one fiber, and one detector are the only optical parts in the cell. This reduced amount of optics minimizes fringing effects efficiently down to a long-term 10^{-4} level (no readjustment needed for approx. 400 flight hours within 4 years). The temperature is measured via a 3 mm² accurate (0.3 K) PT100 sensor for slow temperature changes ($t_{0.5}$ approx. 2.5sec) and also with a thermocouple (type T, 0.5 mm diameter) for faster temperature changes ($t_{0.5}$ approx. 0.5sec). Due to the above mentioned gas handling system and the thermal mass of the whole instrument (approx. 31 kg) the temperature fluctuations are below 0.1 K during aircraft operation. Each cell with a volume of approx. 300 ccm is typically flushed during flight with a gas flow of 20 – 50 vol.liter/min per cell (depending of the installing scenario) leading to a 0.7 sec flushing time using bulk flow estimation. The 2.6 µm closed-path cell is by its design quite similar to the 1.4 µm. The most significant difference is the free-space beam guidance into the cell, since fibers at 2.6 µm are quite critical to handle and suitable beam splitter etc. weren't available. Therefore, the 2.6 µm laser, the reference cell for spectral stabilization, the beam splitters, the mirrors, and the fiber coupling section for the open-path cell are enclosed in a specially designed gas tight 2.6µm optics module box. This module is internally dried by a purging cycle containing a H_2O purifier based on the same idea as for removing parasitic offsets in fiber coupled hygrometer (Buchholz and Ebert, 2014b). The 2.6 µm closed-path cell is directly attached to this laser module. Before reaching the cell, the laser beam is therefore "free-space" guided in a water vapor free environment (typ. <0.5 ppmv) which efficiently avoids parasitic effects in the beam path.

4.3. HAI's open-path cell (OPc)

HAI's open-path cell (Buchholz et al., 2013a) is – just like the closed-path cells – based on the White (White, 1976) principle and installed in the fuselage of HALO. Contrary to the closed-path cells, both wavelengths (2.6 µm and 1.4 µm) are simultaneously coupled into the same 3 mirror cell via two independent glass fibers. The cell (made of 2" diameter mirrors) has a mirror base distance of approximately 15 cm and is set to an optical path length of 4.2 m. The optical measurement volume is located approximately 23 cm above the aircraft skin. The air temperature within the open-path cell is measured via two surface-mounted platinum PT100 sensors. These temperature sensors provide together with the HALO's temperature measurement (Giez, 2012) and our CFD simulation (not shown here) an estimated (±5 K) temperature in the actual measurement volume of the open-path cell. Improved temperature field simulations will be realized in the future as soon as a full CFD model of the aircraft and HAI is accessible. To prevent a dew or frost buildup on the mirror surface, all three mirrors are individually heated (using approx. 20W electrical power each). This raises the mirror temperature, defined by core temperature measurements of the copper mirrors, typically 15 – 20 K above the ambient temperature. The gas pressure measurements are done by two commercial piezo pressure transmitters. Their individual ports are located on either side of the open-path cell. An in-flight pressure sensor validation has been realized recently via the first optical airborne pressure measurements. This calibration-free approach exploits the pressure dependence of the collisional broadening of the water absorption line (Buchholz et al., 2014b).

4.4. TDLAS based multi-phase H_2O measurements

By simultaneously combining closed-path and open-path water measurements, HAI is the first TDLAS hygrometer to allow calibration-free multi-phase water detection. The schematic of the HAI configuration is shown in Figure 4. Both independent closed-path cells are connected via heated inlet lines (visible in Figure 3 left) to the forward facing TGI. The TGI thus samples in clouds both the *gas-phase water vapor* in conjunction with the *ice particles* or H_2O *droplets*. Droplets and ice particles are evaporated in the heated inlet lines, so that both closed-path cells analyze the so called *total water* content, i.e. the sum of the interstitial gas phase water plus the evaporated condensed phase water. This evaporation step is necessary since dTDLAS is typically evaluated in a way to suppress the broad band spectra of condensed phase water and only targets the narrow spectral structure of water vapor. The combination of the narrow absorptions line width with a narrow tuning range of diode lasers allows this discrimination of gas-path water vapor from the broad spectra of ice or liquid water. The open-path cell, which is directly located on the fuselage of HALO, however, measures the pure interstitial gas-phase water vapor of the air which flows through the cell. By combining the total water and the gas phase water measurement, i.e. closed-path and open path signals, it is possible to derive from the difference of both HAI signals the pure condensed water phase, i.e. the ice/droplet phase. To our knowledge, HAI is the first airborne instrument that can measure all water phases with the same detection principle, the same evaluation strategy, and with two channels, each spectroscopically and temporally linked by one single laser. When independent devices have been used previously to measure these water products, even with similar TDLAS instruments like in (Vance et al., 2015), this always led to the discussion on how much of the deviations are caused by atmospheric effects and how many are linked to discrepancies between the participating instruments caused by different characteristics, response times or nonlinearities of the individual instruments. Indeed, the calibration or even validation of all other open-path hygrometers cited above is based on laboratory measurement or comparisons with other hy-

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grometers with different properties which makes it nearly impossible to justify more than statistical (=averaged) statements. HAI avoids these discussions by various self-validations.

4.5. Self-validation

The new self-validating capabilities are a consequence of the unique 2x2 multi-channel configuration, which enables six individual validation pathways, as illustrated in Figure 5. Under clear-sky (= cloud-free) conditions, the independent open-path and closed-path measurements should each deliver the same value. Important to note is that both the 1.4 µm and the 2.6 µm spectrometer channels are evaluated without common parameters in calibration-free first principles mode. This means in detail: two different lasers, two different absorption paths, two different absorption lines, different line parameters, different electronics etc. Nevertheless, both spectrometer channels have to agree within their uncertainty if the first principle evaluation is applicable. Due to the large overlapping H_2O concentration range of approximately 50 to 5000 ppmv, all disturbances can be clearly identified as long as they affect both spectrometers differently. Outside of clouds (clear-sky) – i.e. in ice particle/droplet-free air – all four channels spectrometer channels have to agree. This in particular allows to validate (or even if necessary to calibrate) the closed-path cells in a laboratory environment and to use them as an airborne transfer standard to validate or even calibrate in turn the open-path sensor in flight. This is a unique property of HAI, since each open- and closed-path spectrometer are coupled by one single laser and all four spectrometers are employing the same evaluation procedure. Hence, HAI is called a 2x2 spectrometer.

5. <u>Results</u>

5.1. Signal assessment

Figure 6 shows - at the same instant of time - all seven raw optical signals from HAI's measurement channels at approx. 15000 ppmv and 900 hPa. For visualization, the signals of each laser are scaled to the same peak maximum, since transmission changes are irrelevant according to the equation for concentration retrieval (Equation 2). The time axis (240 Hz laser repetition rate) is already converted to the wavelength axis using the dynamic tuning rate. The triangular diode laser current modulation yields to a triangular modulation of the beam intensity and a similar shape in the wavelength. The tuning range of the 2.6 µm laser is set to be broader in order to allow the evaluation of high absorbance spectra when the peak absorption is in saturation in the absorption line centre. In this case, the Lorenz wings carry the concentration information. The absorbance of the 1.4 µm open-path signal (purple) is higher than the one from the closed-path cell, due to the threefold longer optical path length in the open-path cell. The 1.4 µm reference cell signal (blue) is used for spectral stabilization and acquired from a low pressure (approx. 20 hPa) fibre coupled reference cell. The signal for retrieving and compensating parasitic absorption is shown in green and the associated evaluation is described in (Buchholz and Ebert, 2014b). Accordingly, the three 2.6µm signals are shown. The parasitic absorption compensation of the 2.6 µm system is due to the "free-space" signal in the sealed laser module (see description above) at least as important as the one for the 1.4 μm spectrometer. This compensation treatment is done using an ultra-low pressure (approx. 0.5 hPa) reference cell which provides information for both, the spectral stabilization and the parasitic absorption correction. The latter approach is described as "two pressure separation method" in (Buchholz and Ebert, 2014b).

Figure 7 and Figure 8 show four typical absorption signals in flight with a pre-average of 50 raw measurements done at 240 Hz. This leads to 4.8 measurements per second with a time resolution of 70 ms for each reading while only approx. 1/6 of the whole spectral scan is necessary for the evaluation. Figure 7 allows the simultaneous comparison of both closed-path cells. Gas pressure and temperature are slightly different due to the different piping and installation placement inside of HAI. Both H_2O concentrations agree (Δ = 1.6%) within the combined uncertainties (7.9%) of both channels. Each graph in Figure 7 shows in the top the measured data (green dots) with the fitted Voigt profile (black line). The measurement data are shown as absorbance ($OD_e = -\ln(I/I_0)$) values, i.e. the detector (I) signal divided by the so called "base line" (I_0). The baseline resembles the absorption spectrum in the absence of the molecular target absorption. It is retrieved from the raw spectrum (I) by applying a synchronous polynomial fit of the baseline and a Voigt profile for the absorption line. The residuals between measured data and the model function are depicted below. The remaining structures visible in the residual are optical interference fringes, caused by light scattered from imperfect mirrors, the fiber surface, or the detector arrangement. The repetition frequency of a fringe can be linked to the optical distances between the interfering surfaces. So far, these baseline structures as well as the cell alignment remained stable over the past years. The mirrors never required cleaning during the last four years. Of course, the entire fringe structure can be phase shifted by temperature. However, compared to WMS systems, where fringe levels have to be reduced down to the 10^{-6} OD level to achieve high sensitivities, our calibration-free dTDLAS evaluation is designed to require a stable baseline on the 10^{-4} OD level, which makes it inherently robust. In other words, dTDLAS systems like HAI are usually not as sensitive to misalignments, dirt, and optical structures as WMS systems. On the other hand, they require higher absorbance for the calibration-free approach.

Figure 8 shows a simultaneous comparison between the open-path and the closed-path channel. The outside gas temperature in the open-path cell is approx. 52 K lower than in the closed-path cell. The gas pressure in the closed-path cell, on the other hand, is much higher than in the open-path cell, due to the ramp pressure guiding the air through the TGI and the piping. The measured water vapour concentrations are slightly shifted in time due to the gas transport and the sampling delay in the pipes. The 1σ residual is a factor of six higher in the open-path than in the closed-path sensor. This mainly results from the high wind speed through the open-path cell (approx. 800 km/h). It is important to mention that for diode laser instruments a change of gas temperature, pressure, and concentration over a large range can commonly cause large systematic offsets, which frequently would require at a least three dimensional calibration surface (depending on pressure, temperature, concentration) or to the integration of complex assumptions (Duffin et al., 2007; Goldenstein et al., 2014; Rieker et al., 2009) to correct the data via a simulation model. WMS closed-path systems usually avoid this problem by always keeping temperature and pressure as close and constant as possible to the calibrated level which is obviously not a viable approach for an open-path system.

5.2. Precision

One common figure of merit to quantify the short term optical precision of a spectrometer is the 1σ noise level in the residuum. The idea is based on the assumption that the residual is governed by random noise which limits the precision of the system. The Signal-to-Noise Ratio (SNR) is then defined by the *signal* as the ODe_{peak} value and the *noise* by the statistical standard deviation of the residual and thus equals to OD_{noise}. This definition yields for the 1.4 µm/2.6µm closed-path cells at 4.8 Hz time resolution a SNR of 54 respectively to an SNR of 204. The corresponding precision is then 100/54 ppmv = 1.9 ppmv (1.4 µm) respectively 100/204 ppmv = 0.49 ppmv (2.6 µm). This determination is quite consistent with Figure 8 where the precision of the 1.4 µm closed-path can be estimated to 1.3 ppmv in the same way. However, as the fringe structure of the closed-path cell is quite stable it is better to determine the instrument precision via the Allan variance (Allan, 1966). Figure 9 shows the Allan plot for both closed-path cells. The measurement was done at 255 ppmv by measuring gas from a big vessel. Under these conditions, the precision at 4.8 Hz is 0.22 ppmv (for the 1.4 µm closed-path cell) respectively 0.065 ppmv (2.6 µm closed-path cell). Both values are approximately a factor of 8 lower than the one derived from the single scan analysis, which confirms the stability thesis of fringes. Vice versa we can derive an actual (= unstable, random) 1σ residual OD noise level of 2.3E-5 for the 1.4 µm closed-path and 1.7E-4 ppmv for the 2.6 µm closed-path cell. The best precision is achieved at 3.9 Hz (0.18 ppmv) for the 1.4 µm cell and at 13 Hz (0.055 ppmv) for the 2.6 µm cell, normalizing this with respect to time resolution and optical path length yields a 1σ precision of 187 ppbv·m·$Hz^{-½}$ (1.4 µm) and 31 ppbv·m·$Hz^{-½}$ (2.6 µm).

Comparing that precision ratio of 6 between the 1.4 µm and the 2.6 µm to the ratio in line strength of 20 (see line selection chapter 3.3) confirms the statement mentioned above that drawbacks of the longer wavelength technology significantly constrain the practically achievable maximum gain.

For the open-path sensor, we have not yet been able to calculate an Allan variance so far, since we haven't acquired a data set in flight where atmospheric water was constant enough not to dominate the Allan variance via natural H2O fluctuations . However, using the short term single spectrum evaluation method in Figure 8, one can calculate a SNR of 145, which yields a precision of 2.1 ppmv (= 4.6 ppmv·m·$Hz^{-½}$).

5.3. Uncertainty consideration

One of the major benefits of the dTDLAS evaluation done with HAI is the high level of control over the fitting/evaluation process. In particular, when it is combined with the complete storage of all the raw data, this allows for a dedicated and highly flexible post-flight analysis or post-flight processing. Raw data storage for HAI not only covers all raw spectra but also approximately 120 measured parameters depicting the complete status of the HAI instrument. This "housekeeping data" for example, encompasses 15 temperature measurements that allow for diagnostic statements e.g. about temperature inhomogeneity, electronic drift, or sensor malfunctions. Housekeeping data like these facilitate assignments of a measurement uncertainty closer to the one in a metrological sense (Joint Committee for Guides in Metrology (JCGM), 2008). If measurements are done in harsh environments or under rapidly changing conditions, typical concepts of metrological uncertainty assessments fall short since most of their concepts are developed for laboratory applications under quasi-static conditions. But even in this case the housekeeping parameters provide information for a trustful uncertainty consideration. The uncertainty calculations below are based on a physical model (Equation 2) and are performed with an approach independent from the actual measurement. Depending on the individual science community uncertainties, errors, reproducibility, or misreading are often determined by comparing the readings of an instrument to reference instrument. This is contrary to a metrological uncertainty and furthermore a non-independent approach; we call that therefore simply *deviation*, *standard deviation* etc.

According to the Equation 2 for retrieving the final concentration the individual contributions to the total uncertainty budget can be assessed as followed for both closed-path cells. The optical path length was primarily determined with a Zemax based ray tracing simulation. In addition, it was calculated by the mechanical mirror distance and compared to a test measurement with a known amount of CH_4 gas in the cell. These measurements lead to an uncertainty of the optical-path length of 15 mm (1.1%). The uncertainty for the used H_2O absorption line strength is 3.5% [44]. The temperature sensors (PT100 and thermocouple type E as described above) are calibrated against metrological transfer standards (Rosemount platinum resistance thermometer type 162CE (PRT-25), uncertainty 1.5 mK). Due to the well-known fact that temperature measurements in moving gases suffer from many issues, we use a conservative estimation of 1K (0.3%). The manufacture of the pressure sensors (Omega PAA33X) advertises a resolution of 0.02 hPa and a long-term accuracy of 0.5 hPa. We use 1 hPa as a conservative uncertainty estimation. A general statement for the laser tuning uncertainty is difficult in a metrological sense, since it depends on local effects in the spectra, pressure range, concentration level and number of absorption lines fitted. From our experience, we assume that deviations related to the fitting process in total (tuning + fit process) are in the range of below 1.5%. As a conservative approximation we use 2% for the fit and 1% for the tuning. This yields to a total uncertainty of 4.3%. for the 1.4 µm closed-path cell.

The largest relative influence to this total uncertainty budget results from the line strength (66%), followed by the fitting uncertainty (21%), as well as the optical path length (7%). Additionally, the offset uncertainty is defined in a calibration-free dTDLAS system by the capability of minimizing and determining parasitic effects. It is ±3 ppmv for the 1.4 µm closed-path cell of HAI, which uses the same parasitic prevention treatment as SEALDH-II (Buchholz and Ebert, 2014b). Thus, the total uncertainty of the 1.4 µm closed-path system is 4.3% ±3 ppmv. Similar considerations lead to an uncertainty of 5.9% ±0.4 ppmv for the 2.6 µm closed-path cell.

Related to the open-path cell, the statement is more difficult. First of all, a full CFD model of the boundary layer around HALO is currently missing, which would allow a retrieval of the gas temperature in the open-path cell. The first CFD test runs of HAI's open-path sensor and its immediate surroundings have been realized (A. Afchine (FZJ) , published in (Klostermann, 2011)), and led to a first temperature uncertainty estimate of ±7 K. The full CFD model could also provide pressure field calculations. However, in this case, we developed a method on how to retrieve the pressure directly from the dTDLAS signal (Buchholz et al., 2014b) and use that for a validation of the built-in micromechanical pressure sensor. The pressure dependent uncertainty is in the range of 3-5%. The uncertainty of the fitting process reveals a much higher complexity. It depends on all impacts on the spectral signal itself such as: background radiation on the detector, misalignment due to the distortion of the HALO fuselage, forming of an ice layer on the mirrors, high optical dense cloud transects, eddies around the aircraft etc. From all the data gathered and compared during the HALO missions TACTS and ESMVal, we couldn't find any deviation larger than ±5% in clear sky conditions between the open-path and the closed-path sensor which couldn't be clearly linked to explainable effects. Besides of the availability of an entire CFD model in the future, HAI will be validated by sampling the pure water vapor gas-phase (backward measurement) with its closed-path cells in future campaigns. These datasets will sustainably prove if the ± 5% deviation is valid under all conditions, since then all four channels of HAI (Figure 5) would permanently measure the same atmospheric value. Compared to the deviations, revealed in the AQUAVit (Fahey et al., 2014) intercomparison of state of the art, airborne hygrometer in 2007, this 5% is fourfold smaller than the span (20%) of those instruments there. Additionally, one has to keep in mind that this comparison was done under quasi-static laboratory conditions. Therefore, this in-flight 5% deviation between the open-path and the closed-path sensor shows the great performance of HAI, even if a general metrological uncertainty is difficult to be given yet at this time.

5.4. First airborne measurements with HAI on the HALO aircraft

Figure 10 shows typical HAI measurements during the first HALO science mission "TACTS/ESMVal". The chosen flight profile shows 15 min of a slow ascent from the lower to the upper troposphere. Depicted in Figure 10 are: the H_2O concentration measured with the 1.4 µm CPc (black), 1.4 µm OPc (magenta), and 2.6 µm CPc (red) channel of HAI (left axis) as well as the ambient gas pressure (right y-axis, in blue), which is directly connected to the flight level (height). Other instruments indicated that HALO flew in the early part of the flight (left) in clear sky conditions (no clouds). All Figure 10 data were plotted with 1 Hz temporal resolution. With increasing height, the H_2O concentration declines about 2.5 magnitudes from approximately 2000 ppmv to 75 ppmv, in combination with simultaneous relative pressure variation from approx. 175 to 450 hPa (Δp=275 hPa). The H_2O concentrations, measured with HAI's two independent closed-path spectrometer channels, fit nicely to each other and show only a small average relative deviation of 1.9% (bottom graph in red), which is entirely covered by the uncertainty range of each spectrometer (4.3% resp. 5.9%). This result is consistent with other measurements, where the 2.6 µm path was persistently on average approx. 2% higher than the 1.4µm channel. This could be easily explained with the 2.6µm line strength from the HITRAN database being 2% too small. The 1.9% deviation also needs to be related to the fact that both hygrometer channels are evaluated completely independently, without any calibration of the instrument and most importantly under real-world flight conditions. Despite these demanding conditions HAI's two closed-path channels generate a relative deviation which is just 1/10 of the above mentioned 20% in AquaVIT (Fahey et al., 2014), which demonstrates the excellent accuracy of the HAI spectrometers.

The purple signal also depicted in Figure 10 shows the measurement of the 1.4 µm open-path sensor (1.4 µm OPc). Compared to the 1.4 µm closed-path cell (1.4µm CPc), the 1.4 µm OPc shows a relative deviation of ±5% peak-to-peak or 2% on average. This performance of the 1.4µm OPc is to be compared with previously described airborne hygrometer which resemble the current state of the art. However, it has to be noted that a huge number of airborne hygrometers with an extreme diversity in sensing principles and

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specific performances have been published and described, so that a general overall statement about the "state of the art" is very difficult to make. In addition, there is a very large variation in sensor accuracy and the performance figures stated are sometime misleading and contradictory to the results of the few rare large scale intercomparisons like AquaVIT. However, a short incomprehensive review on the performance of previous open-path instruments should be given here:

a) A calibrated, 1 m direct absorption, open-path sensor at 1.4μm was developed in a feasibility study for the Strato 2C research aircraft and described to cover a concentration range of 2-2000 ppmv with 100 ppb precision at 1 Hz. It was claimed to have an accuracy of 5% , but this hygrometer never flew (Roths and Busen, 1996).

b) A calibrated open-path sensor with an optical path length of 375 cm, folded in a Herriot cell, and a time resolution of 25 Hz gave a precision of 80 ppbv. This instrument is designed for the American HAIPER (Anon, n.d.) aircraft, a Gulfstream 550 like the HALO aircraft. To our knowledge this is the only **open-path** hygrometer which was frequently installed and used during the last years on an aircraft. Typical deviations to other instruments were in the range between 2% and 10% (Zondlo et al., 2010).

c) Another calibrated, Herriot cell based open-path hygrometer with a total path length of 11 m, a measurement frequency up to 10 Hz and a precision of 50 ppbv at 0.5 Hz. This instrument claims to have an uncertainty of 5% to 10% (May, 1998). However, to the best of our knowledge, except of a few test flights, no further flight was nor published or performed.

In summary, HAI's 2% deviation appears to be much better than typical statements in publications.

To further evaluate the performance of HAI's open-path channels we further discuss the TACTS data. The dark purple line shows the data after smoothing them with a 60 second sliding window algorithm. Quite interesting are the two dents at 13:41 and 13:44 in the relative deviation, which means the closed-path measurement was higher than the open-path measurement. Both of them happened after the water vapour concentration dropped sharply. Hence, the dents might be explained by desorption processes inside of the TGI, its piping and the measurement cell. This idea is backed by the 1.4 μm to 2.6 μm closed-path comparison (red). The same behaviour is also visible at 13:44, but here the 2.6 μm close path cell added less desorption water to the gas steam than the 1.4 μm one. Another influence leading to that deviation could be a temperature measurement on HALO affected by fast changes of humidity. The total temperature influence in Equation 2 is at this temperature and pressure approximately 0.6%/K caused by the temperature influence via the ideal gas law, the line strength temperature dependence and the Gaussian width. This brief insight in a detailed data discussion demonstrates the unique opportunities offered by HAI, which allow a deep analysis to identify and eventually classify different measurement sections of a flight in order to distinguish instrumental artefacts from real atmospheric situations.

On the second part of the flight shown in figure Figure 10 , the aircraft passes through ice clouds (Cirrus, temperature < -40 °C). The evaporation of the ice crystals captured by the gas inlet leads to a clear enhancement of the amount of water vapor in the closed-path cells. The relative comparison shows a ± 5% noisy structure, which is caused by the low time resolution of just 1 Hz. H_2O gradients in this particular part of the flight are in the range of several 1000 ppmv/s. Due to SSD space limitations in the first campaign, only parts of the flights where sampled with the highest time resolution of 240 Hz, in the other parts the

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data where online pre-averaged. Like in the clear sky region, the average deviation between both closed-path cells during cirrus cloud transects remains at 2%. To further check the accuracy of the open-path channel, we use two methods: The first is based on the physical argument that the gas-phase measurement always has to be lower than the total water measurement, which consists of gas-phase plus evaporated ice/liquid phase. This requirement is entirely fulfilled. The second check compares the absolute gas-phase measurement with the saturation mixing ration (SMR). In case of a fully equilibrated cloud, the gas-phase has to be saturated at temperature T, meaning that the relative humidity is rH = 100%, from which we can also derive a SMR with the help of the water vapour partial pressure curve. This is a weaker check, since it is a well-known fact that super saturation can occur during strong air updrafts.

The transition region at the fringe of the cloud is also very interesting, where the closed-path cell sees an enhancement while being still below the SMR. This means that the fringe of the cloud was not saturated but had sublimating ice particles in it.

These first detailed close-ups in HAIs multi-phase measurements show the potential to investigate many interesting questions in the following campaigns focusing on Cirrus clouds which are strongly linked to a very reliable accuracy study for the open-path cell in clouds. To really investigate and fully understand all effects, HAI needs more data in Cirrus, mixed-phase and other clouds in a multi-phase configuration as well as a validation campaign in which closed- and open-path cells measure the pure gas-phase via a backward measurement. The latter is necessary to distinguish between sampling-effects and optical/spectral-effects in and outside of clouds. This knowledge, especially in combination with the mentioned CFD model, will then allow to reevaluate HAI's raw data which are always saved during flights and extent statements even for this short cirrus transect.

6. Conclusion and outlook

The novel *Hygrometer for Atmospheric Investigation, HAI,* realizes in a unique concept a simultaneous gas-phase and total water measurement. Based on calibration-free direct tuneable diode laser absorption spectroscopy, HAI allows accurate, precise (0.065 ppmv at 4.8 Hz, channel depended) and very fast (up to 240 Hz) measurements with a metrologically defined uncertainty (4.3%, channel dependent). HAI contains four measurement channels, grouped as two completely independent dual-channel spectrometers, one at 1.4 µm and one at 2.6 µm to cover the entire H_2O concentration range of the atmosphere. Each spectrometer feeds light in a wavelength-individual extractive, closed-path cell with an optical absorption path length of 1.5 m for total water measurements. Additionally, both spectrometers couple their light in a common open-path cell (optical path of 4.8 m) located outside of the aircraft fuselage, for a sampling-free and contactless determination of the gas-phase water content. These four spectroscopic channels plus three additional supplementary spectroscopic channels allow multiple self-validation strategies inside and outside of clouds and therefore solve the current lack of an integrated approach to validate open-path sensors in flight. HAI's complex control software minimizes maintenance at ground and ensures almost entirely autonomous operation. In addition, instrument health is permanently supervised by permanent storage of a set of more than

120 housekeeping data. This enables a novel, holistic quality management and a sophisticated signal cross check, which guarantees a high trust level of the final H_2O values.

HAI was operated for the first time during the TACTS/ESMVal flight campaign for more than 120 operation hours without any malfunction. The entirely independent, never calibrated first principles evaluation of the closed-path spectrometer channels reduced in-flight deviations to only 1.9% over a large concentration (75 to 2000 ppmv) and pressure range (175 to 450 hPa). The deviation between the open-path and the closed-path measurements in the same flight segment was just 2%. Despite measuring with a single evaluation concept, over a very broad range of conditions (i.e. temperatures (-70°C to 30°C), gas speeds (cm/s vs 100 m/s), optical disturbances (no background light vs sunlight, clean mirrors vs. dirty scratched mirrors outside), HAI provided a high trust level of the data over extensive science missions. Laboratory evaluations demonstrated the lowest achievable precision of 0.18 ppmv (at 3.8 Hz) for the 1.4 µm closed-path cell and 0.055 ppmv (at 13 Hz) for the 2.6 µm closed-path cell.

In conclusion, HAI proved during its first deployment a novel, highly complex and demanding set of capabilities. This will enable in the future a much more accurate and stringent evaluation of atmospheric multiphase water vapor data inside and outside of clouds and foster in further HALO missions the investigation of new scientific questions in the atmospheric water cycle. HAI can serve in the future as a major, powerful tool for cutting-edge atmospheric water vapor measurements.

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Figures:

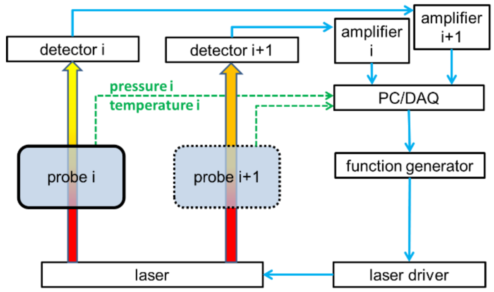

Figure 1: Simplified schematic of a standard dTDLAS setup with several spectroscopic channels: HAI combines two lasers (one at 1.4 µm, one at 2.6µm for high and low concentrations respectively), two with measurement cells (open- and closed-path), and two supplementary spectroscopic channels (parasitic water detector (Buchholz and Ebert, 2014b) and reference cell for spectral stabilization). Completely independent from the 1.4 µm system is HAI's 2.6 µm system with two other channels (open- and closed-path) and one additional channel for the spectral stabilization. Hence, HAI has in total seven spectroscopic channels.

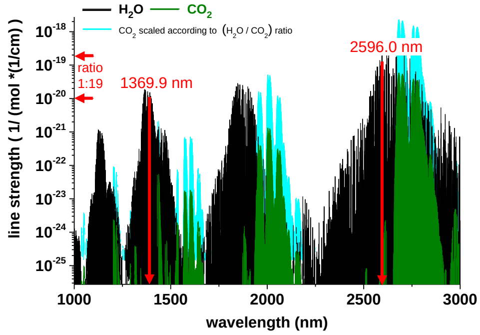

Figure 2: HITRAN 2008 line strength plot (Rothman et al., 2009) for CO_2 (green) and H_2O (green). The turquoise spectrum in the back shows the scaled (factor 39) CO_2 line strength for visualization of the stratospheric situation with a low level of water vapor (10 ppmv) and standard CO_2 (390 ppmv) level.

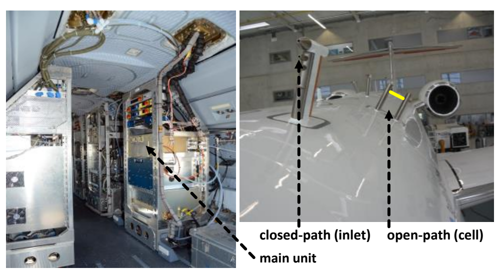

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Figure 3: Photos from the cabin layout of HALO with payload of the TACTS/ESMVal campaign (left). Photo of the fuselage of HALO showing the trace gas inlet (TGI) for the closed-path HAI cells (for total water detection) as well as the open-path HAI cell (for gas phase water detection; yellow marked are the open laser beams)

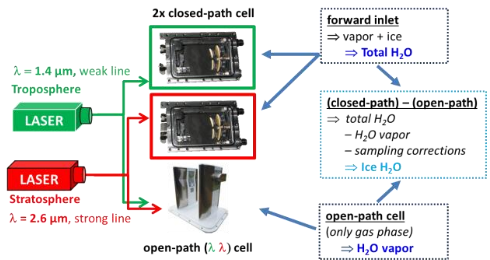

Figure 4: Schematic of HAI's working principle in the multi-phase configuration. By combining selective open-path gas phase measurements with total water measurements in the closed-path, extractive cell (ice is evaporated before the cell by heated inlet lines), it is possible to derive the ice water content from the difference between closed-path and open-path readings.

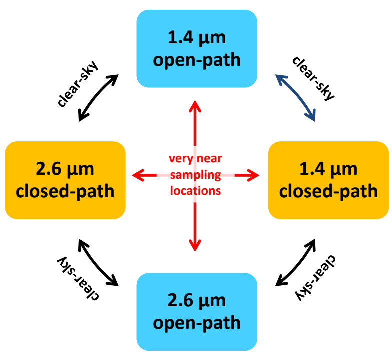

Figure 5: Self-validation possibilities of HAI under clear-sky conditions (black) and redundancies under all atmospheric conditions (red). For details see text.

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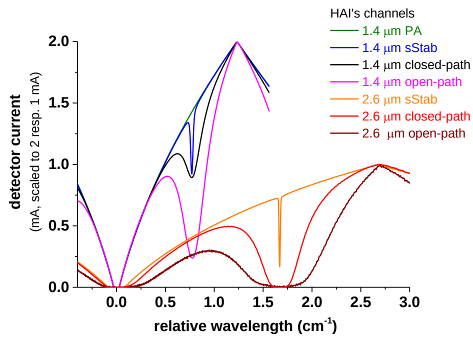

Figure 6: dTDLAS raw signals (spectra) of all seven spectroscopic channels permanently acquired and saved by HAI. The x-axis is already converted to the wavelength axis via the dynamic tuning rate of the laser. Each spectrum contains approx. 1700 16-bit values. (PA: parasitic absorption, sStab: spectral stabilization)

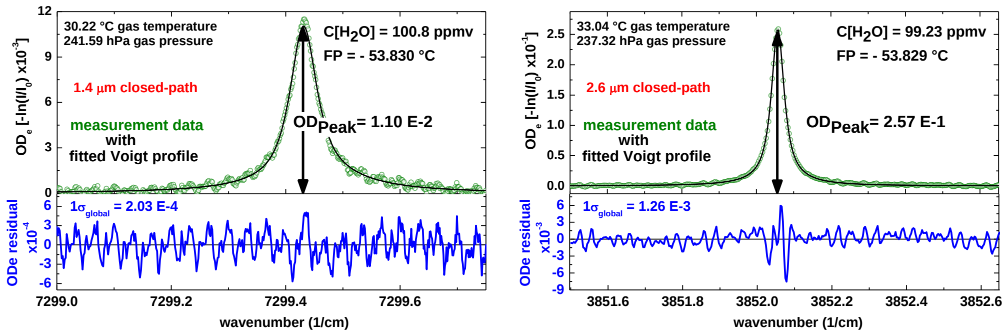

Figure 7: Typical pre-processed absorption signals (after baseline, offset and transmission correction) for both closed-path spectrometer channels (right 1.4 µm left: 2.6 µm) during flight. The laser modulation frequency was 240 Hz. 50 individual raw scans are pre-averaged yielding 4.8 H_2O measurements per second with 69 ms total integration time. This corresponds to a spatial resolution of 15 m at 800 km/h cursing speed. Without averaging, HAI achieves a maximum time resolution of 1.3 ms, respectively a spatial resolution of 30 cm at 800 km/h cursing speed. (It should be noted that the four vertical axes have different scales).

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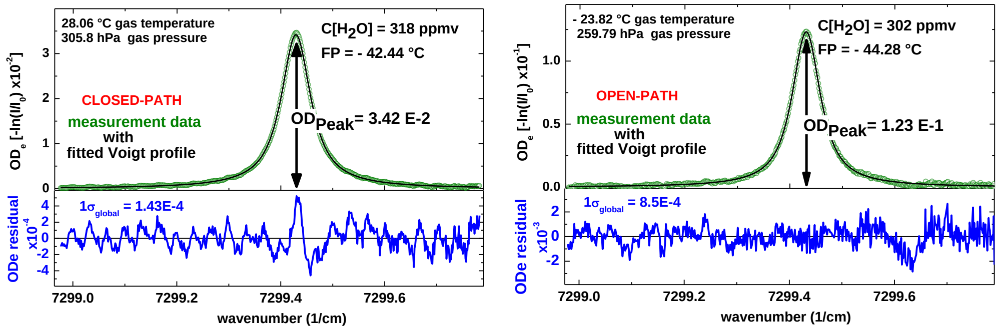

Figure 8: Comparison of pre-processed TDLAS scans (similar to Figure 7) of HAI's 1.4 µm closed-path (left) and the 1.4 µm open-path cell (right) during a HALO flight. Despite the entirely different measurement conditions such as wind speed (cm/s to 100 m/s) or temperature (+28 °C to -23 °C), both channels are evaluated calibration-free with the exact same methods and model.

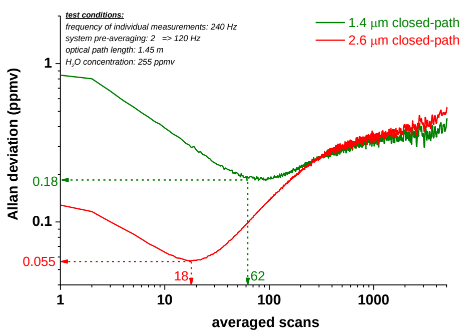

Figure 9: Allan variance plots for both closed-path spectrometer channels. The measurements were done by analysing gas from a big buffer vessel with a H_2O concentration of 255 ppmv. The optimal precision from this measurement is 0.22 ppmv for the 1.4 µm closed-path cell at 4.8 Hz (same as in Figure 7) and 0.065 ppmv for the 2.6 µm closed-path cell. The best i.e. highest precision of 0.18 ppmv is achieved at 3.8 Hz for the 1.4 µm cell and 0.055 ppmv at 13 Hz for the 2.6 µm cell.

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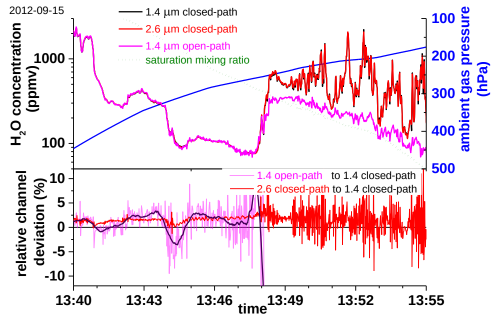

Figure 10: Typical HAI measurement on board HALO during the TACTS/ESMVal campaign. The flight profile shows a 15 min section of a slow ascent from the lower troposphere to the upper troposphere. The first part (under clear sky condition) can be used to perform an absolute in-flight accuracy validation of both closed-path channels of HAI; the second part on the right side demonstrates a HAI multi-phase H_2O measurement. For further explanations of all signals and visible effects see text.