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Fast in situ stratospheric hygrometers: A new family of balloon-borne and airborne Lyman α photofragment fluorescence hygrometers

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Abstract. A new set of hygrometers based on the Lyman α photofragment fluorescence technique has been developed for operation on aircraft and balloons in the stratosphere and upper troposphere. They combine technical details from existing fluorescence hygrometers with several improvements in order to achieve the highest data quality and to minimize maintenance and operational procedures. With these instruments, stratospheric H_2O measurements can be accomplished with a precision of < 0.2 ppmv at 1 s integration time, as has been demonstrated both in the laboratory and under field deployment. The design enables a rapid exchange of the air sample of the order of 1 s for fast measurements of small-scale variations of the H_2O mixing ratio in the atmosphere. The hygrometer is calibrated using a laboratory calibration bench with approximately 4% accuracy. Measurements made by the hygrometer are compared with a frost point hygrometer during an aircraft mission at H_2O mixing ratios from 280 to 8 ppmv, yielding an agreement between both techniques within the instrumental errors.

1. Introduction

Water vapor is one of the most important trace gases in the atmosphere. Its absorption and emission bands in the IR dominate the outgoing radiation budget of the atmosphere. H_2O is also the most abundant condensable trace gas in the atmosphere, even at stratospheric altitudes. Therefore it is a critical parameter for the existence of, for example, polar stratospheric clouds and aerosols and their impact on heterogeneous processing of air masses. Furthermore, H_2O is a major source for HO_x species that participate in fast photochemical processes. Measurements of H_2O may also serve to identify transport processes, in particular, the exchange of air masses between the stratosphere and troposphere.

The distribution of H_2O in the atmosphere shows a large gradient at the tropopause with mixing ratios of several hundred ppmv (part per million by volume) in the free troposphere to a minimum of about 4 ppmv in the lower stratosphere. At greater altitudes, the H_2O mixing ratio again increases up to approximately 7 ppmv due to oxidation of CH_4 [e.g., *Ehhalt*, 1974; *Kley et al.*, 1979]. Therefore measurements of H_2O in the stratosphere and the tropopause region require instruments with a large dynamical range and that are capable of measuring the low stratospheric as well as the much larger concentrations in the troposphere.

Besides ground-based and spaceborne remote sensing instruments, several in situ techniques have been developed to measure water vapor in the stratosphere: For many years, frost point hygrometers have been used on balloons [e.g., *Brewer et al.*, 1946; *Oltmans et al.*, 1985; *Ovarlez et al.*, 1989] and also in aircraft [*Foot*, 1984; *Ovarlez and van Velthoven*, 1996]. The typical time resolution of sophisticated frost point hygrometers is of the order of 10 s. The dynamical range allows for measurements from the troposphere up to the middle stratosphere. A few tunable diode laser (TDL) absorption instruments for airborne measurements of H_2O were developed in the past [e.g., *Silver and Hovde*, 1994], and recently a TDL instrument was used on board the ER-2 aircraft, validating this technique for stratospheric water vapor measurements [*May*, 1998]. For a 2 s integration time this instrument showed a measurement precision of ±0.05 ppmv in the stratosphere with an estimated total uncertainty of 5-10% depending on pressure.

For about two decades, Lyman α photofragment fluorescence hygrometers have been used from balloons [e.g., *Kley and Stone*, 1978; *Weinstock et al.*, 1990], for airborne measurements [e.g., *Kelly et al.*, 1989; *Weinstock et al.*, 1994] and on rockets [*Khaplanov et al.*, 1996]. The technique enables a fast measurement even under highly variable conditions as in the tropopause region or in clouds and contrails. A typical integration time of a few seconds allows a precision of the order of 0.2 ppmv at stratospheric mixing ratios. Another advantage of this technique is that the air in the instrument and its inlet tubes can be exchanged very rapidly. Similar to the frost point hygrometer, the measurements can be extended into the free troposphere due to the large dynamical range. Lyman α fluorescence

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instruments need to be absolutely calibrated and traced to a secondary standard technique.

2. Photofragment Fluorescence Technique

The method to measure H_2O in the stratosphere by photofragment fluorescence was developed in the 1970s by *Kley and Stone* [1978] and *Bertaux and Delannoy* [1978]. The photodissociation of H_2O molecules by radiation at wavelengths $\lambda < 137$ nm produces electronically excited OH:

$$H_2O + h\nu\ (\lambda < 137\ nm) \rightarrow OH\ (A^2\Sigma^+) + H(^2S) \quad (1)$$

with a quantum yield far less than 1. The electronically excited OH relaxes to the ground state both by fluorescence at $\lambda = 280\text{-}330$ nm and by collisions with air molecules M:

$$OH\ (A^2\Sigma^+) \rightarrow OH\ (X^2\Pi) + h\nu\ (\lambda = 305\text{-}325\ nm) \quad (2a)$$

$$OH\ (A^2\Sigma^+) + M \rightarrow OH\ (X^2\Pi) + M \quad (2b)$$

By measuring the fluorescence radiation the H_2O abundance can be determined. For stationary conditions, the number of fluorescence photons N_f is given by

$$N_f = A_0 \cdot \frac{[H_2O] \cdot J \cdot \Phi}{A_0 + k_q \cdot [air]} \quad (3)$$

$[H_2O]$ and [air] denote the concentration of H_2O and air molecules, J the photodissociation rate of reaction (1), Φ the quantum efficiency for the OH production via (1), A_0 the Einstein coefficient of reaction (2a), and k_q the quenching coefficient of the OH radical in air (reaction (2b)). Using $A_0 = 1.26 \times 10^6\ s^{-1}$ [*Crosley and Lengel*, 1975] and $k_q = 2.3 \times 10^{-11}\ cm^{-3}\ s^{-1}$ [*Kley and Stone*, 1978], $A_0 << k_q \cdot [air]$ in the lower stratosphere, that is, for altitudes below 20 km. For this case, equation (3) can be approximated by

$$N_f = C \cdot \frac{[H_2O]}{[air]} \quad (3')$$

The factor C in (3') summarizes molecular coefficients known from the literature as well as instrument specific quantities. If C is a constant, the number of detected fluorescence photons is proportional to the H_2O mixing ratio $[H_2O]/[air]$ for measurements in the troposphere and lower stratosphere. For measurements at higher altitudes, equation (3) has to be used to obtain correct water vapor mixing ratios.

In reality, C is a function of J and thus depends on the photon flux in the fluorescence volume, which in turn depends on variations of the lamp intensity and absorption by atmospheric gases. In the vacuum UV spectral region (VUV), absorption by oxygen and water vapor has to be taken in account. The attenuation and variation of the VUV radiation have to be monitored when determining H_2O mixing ratios by measuring the fluorescence signal (see section 3.1). At tropospheric H_2O concentrations, VUV radiation is strongly absorbed. The Lyman α line at $\lambda = 121.6$ nm coincides with a narrow deep minimum of the oxygen absorption cross section [e.g., *Watanabe et al.*, 1953]. Using a Lyman α radiation source with high spectral purity at $\lambda < 200$ nm, the oxygen absorption is small enough to enable measurements down to the middle troposphere. In the past, krypton line sources ($\lambda = 123.6$ nm) have also been used in fluorescence hygrometers for which, however, a path length of a few centimeters is optically thick and which are therefore only suited for higher altitude measurements.

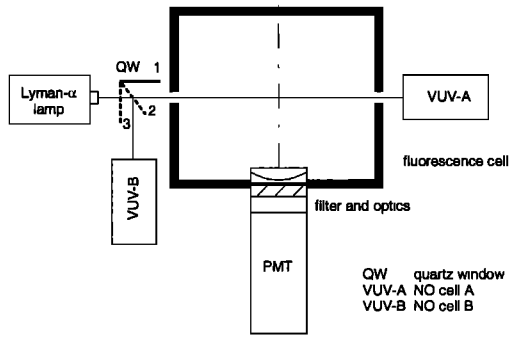

Figure 1. Sketch of the fluorescence cell and the major components as Lyman α radiation source, photomultiplier tube, vacuum UV (VUV) detectors and the mirror drive.

3. Fast in Situ Stratospheric Hygrometer (FISH)

The program to develop and to use the fast in situ stratospheric hygrometer (FISH), based on the Lyman α photofragment fluorescence technique, started several years ago at the Forschungszentrum Jülich. A prototype instrument was designed for balloon-borne operation, and its first flight was on November 10, 1990 [*Mörschel et al.*, 1991; *Mörschel*, 1992]. In the following years we developed a hygrometer for use on commercial and high-altitude aircraft based on a completely new design with major improvement of several components. The first measurement and test campaign of this instrument was conducted in March 1994 using the German research aircraft Falcon-E [*Zöger*, 1996; *Zöger et al.*, 1997]. Based on the good performance of the aircraft instrument, we redesigned the balloon instrument recently in order to improve its time resolution and precision.

The method of Lyman α photofragment fluorescence has been used and described by several groups in the past. Although most of the existing instruments show good performance, their usage is often limited, for example, by sensitivity to ambient solar radiation which usually exceeds the fluorescence radiation to be detected, exchange time of the air in the fluorescence cell, contamination due to desorption of water vapor from walls, signal-to-noise ratio, or size and weight. The design of FISH combines the advantages and performance of existing instruments, avoiding the aforementioned limitations. Besides other technical details, this was mainly achieved by a compact cell design, well-selected use of materials (e.g., gold-plated surfaces), a patented lamp design and extensive laboratory testing. For calibration and laboratory characterization a novel approach is used. In this paper we describe the design of FISH, emphasizing its characteristics and performances as well as results from intercomparison with other H_2O measurement techniques.

3.1. The Concept of FISH

FISH is a closed, vacuum tight instrument based on a modular design that allows use of most of the components both in the aircraft and the balloon instrument. Furthermore, the modular design allows for a flexible deployment on

different aircraft under different conditions (e.g., pressurized and nonpressurized environment) with only a few modifications.

The core of FISH is the fluorescence cell with the Lyman α radiation source, the photomultiplier tube (PMT) including an interference filter and projecting optics, two VUV radiation detectors and a mirror drive. Figure 1 shows a sketch of the arrangement of the different components. The mirror drive allows monitoring of the intensity of the Lyman α radiation and a determination of the background and dark count rates. Thus calibration factors can be determined which are nearly independent of changes of the output of the radiation source. The moving mirror design avoids additional errors in the water vapour mixing ratio by degradation of a fixed magnesium fluoride beam splitter to measure the intensity as used by, for example, *Kelly et al.* [1989]. Laboratory and field measurements showed that the reflectivity of the mirror does not change significantly over several flights.

The mirror in Figure 1 is a quartz window that is transparent for wavelengths at which the fluorescence occurs but opaque for VUV radiation. The three positions of the quartz window allow for the following measuring cycle: In position 1, both VUV radiation and radiation with longer wavelength that is also emitted by the Lyman α radiation source (see 3.2.1) penetrates into the fluorescence cell. So the count rate of the PMT is a combination of the fluorescence signal, the background from the Lyman α radiation source and the dark counts of the PMT. The detector VUV-A, which is mounted approximately 150 mm opposite to the Lyman α radiation source, allows determination of the absorption by H_2O along this light path at high H_2O concentrations, which may serve as an additional calibration of the fluorescence signal (see section 4). The quartz window in position 2 reflects the radiation onto detector VUV-B, which is mounted the same distance from the Lyman α radiation source as the fluorescence volume. This geometry, first used by *Kley and Stone* [1978], allows the detector to monitor the changes in the intensity of the Lyman α source both due to changes of its output and due to absorption along the distance from the radiation source to the fluorescence volume by, for example, H_2O and O_2. In position 3 of the quartz window, only the background radiation of the lamp penetrates into the fluorescence chamber, while Lyman α radiation is absorbed. Hence the contribution of the background radiation to the fluorescence signal can be determined. The relative count rate N_f^* is defined by the difference in the count rate of the PMT with the quartz window in position 1 (N_1) and in position 3 (N_3) and normalized to changes of the intensity, expressed by the signal UB of the detector VUV-B at quartz window position 2:

$$N_f^* = \frac{N_1 - f_u N_3}{U_B} \quad (4)$$

f_u is a parameter that mainly considers the transmission of the quartz window for UV radiation and thus is slightly larger than 1. Using the relative count rate N_f^*, equation (3)' is transformed to

$$N_f^* = C^* \cdot \mu_{H2O} \quad (3'')$$

with the H_2O mixing ratio $\mu_{H2O} = [H_2O]/[air]$. The parameters C^* (units: cps ppmv^{-1} V^{-1}) and f_u have to be determined during the calibration procedure (see section 4).

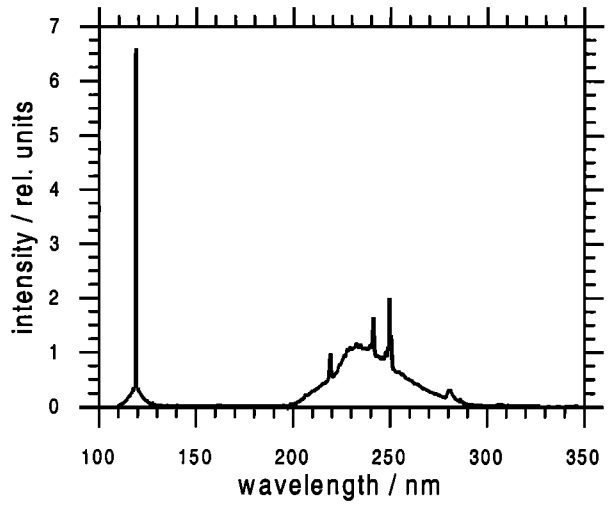

Figure 2. Typical emission spectrum of a Lyman α radiation source. At 250 nm the spectral sensitivity of the monochromator is larger by approximately 2 orders of magnitude than at the Lyman α line.

3.2. Experimental Details

3.2.1. The Lyman α radiation sources. For the Lyman α radiation source, RF-excited discharge lamps are produced in our laboratory based on a similar technique first developed by *Dieke and Cunningham* [1952]. They consist of a sealed off bulb filled with a hydrogen reservoir and an argon filling to maintain the discharge. The lamps are powered with approximately 15 W.

Typical photon fluxes at the Lyman α line are several 10^{14} photons sr^{-1} s^{-1}, which are required to obtain the signal-to-noise ratios given below. The spectral purity of our Lyman α radiation sources in the VUV spectral region is sufficient to dispense with the oxygen filter that is used in other Lyman α hygrometers as well as in our prototype instrument [*Mörschel et al.*, 1991, and references therein]. Figure 2 shows a typical spectrum of our lamps in relative units. Since the sensitivity of the monochromator strongly decreases for wavelengths less than 150 nm, the molecular hydrogen continuum and the much more intense Lyman α line are displayed simultaneously. For an optimum lamp the tail of this continuum around 310 nm has to be as low as possible in order to keep the background of the fluorescence as low as possible.

Most of the Lyman α radiation sources used by other groups are equipped with uranium hydride as the hydrogen reservoir and getter for impurities [e.g., *Kley and Stone*, 1978; *Kelly et al.*, 1989; *Zuber*, 1989; *Weinstock et al.*, 1990]. In our lamps we successfully replaced uranium by a ZrCo alloy [*Woyke et al.*, 1995; *Woyke and Schober*, 1995] that simplifies the production and handling of the lamps significantly. Since the ZrCo hydride has similar thermodynamical properties as UH_3, the operational conditions are comparable to optimize the H_2 partial pressures to a few Pa at an argon filling of 2-7 hPa.

The degrading of the MgF_2 window transmission limits the lifetime of the sealed getter lamps to several tens of operating hours. Laboratory experiments showed an exponential decrease of the intensity within the first 10 hours and a much

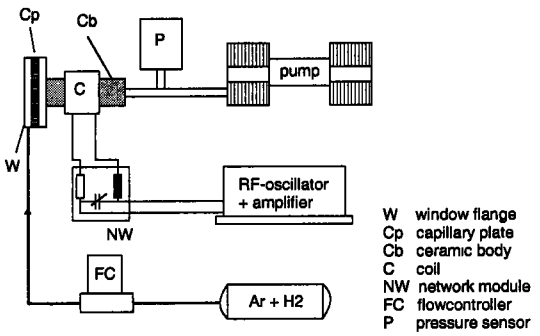

Figure 3. Schematic flow and controlling of the purged Lyman α radiation source.

slower decrease within the next 20 hours. XPS (X ray photoelectron spectroscopy) and SNMS (secondary neutral mass spectrometry) analysis of single MgF_2 windows from the lamps used showed traces of SiO and C. Therefore plasma deposition of Si and C at the inner side of the MgF_2 window is believed to cause the intensity decrease.

Therefore we recently developed a Lyman α radiation source that is operated purging the bulb with a H_2/Ar mixture [*Tuitjer*, 1997, *Zöger and Tuitjer*, 1998]. Figure 3 shows a flow diagram of these lamps. Varying the flow through the flow controller (FC) controls the gas pressure in the discharge tube. The compact four-head diaphragm pump (KNF Neuberger) maintains a pressure of a few hPa at typical flow rates in the range of a few mL/min. In order to avoid plasma deposition at the inner side of the MgF_2 window, a capillary plate (Hamamatsu) is mounted a few millimeters in front of the window. The capillary plate transmits and focuses the light but prevents most of the plasma particles from reaching the window since they recombine on the large surface of the plate. Its transmission is given by the area ratio between capillaries and wall material of 7:3 and by its focusing properties which are adventageous for use in a fluorescence instrument. Degrading of the transmission due to plasma recombination is slow: one plate can be used for several hundred operating hours. The housing is made of an Al_2O_3 tube. The plasma is maintained by radio frequency excitation at ~140 MHz with an output power of approximately 30 W. The radio frequency is coupled into the plasma via a copper coil. Two variable capacitors are used for proper tuning of the coil to the amplifier. Laboratory tests as well as recent aircraft campaigns showed a nearly constant Lyman α intensity of the flow lamp over several tens of hours. The additional components such as the gas bottle, pump and flow and pressure sensor yield an additional weight of approximately 3 kg. The purged lamps show similar spectral characteristics to the sealed getter lamps. Since the procedure for preparing a sealed getter bulb is more time consuming than operating the purged Lyman α radiation source, we recently replaced the sealed getter lamps in the aircraft instruments for frequent measurements.

3.2.2. Detectors. For the detection of the fluorescence radiation, a photomultiplier tube is used in photon counting mode. Most recently, the hygrometer is equipped with a Hamamatsu photon counting unit H6240, which includes PMT R4220 selected for dark counts less than 30 c/s. In order to confine the sensitivity to the spectral interval of the fluorescence radiation, an interference filter is placed between the PMT and the fluorescence volume. The filter is centered at 310 nm with a full width at half maximum of 12 nm and a transmission of 80%.

For the detection of the Lyman α radiation (detectors VUV-A and VUV-B in Figure 2), ionization cells filled with NO are used (distributor Buck Research, Boulder, Colorado). The energy required to ionize NO corresponds to a threshold wavelength of 134 nm. Therefore NO filled ionization cells are a highly selective radiation sensor for the VUV spectral region [*Samson*, 1967].

3.2.3. The fluorescence cell. The design of the fluorescence cell is crucial for the performance of the instrument. Different approaches have been used in the past: The balloon instrument of *Kley and Stone* [1978] is an open structure to avoid contamination by desorption of H_2O from walls, but is thus highly sensitive to ambient solar radiation. The instrument of *Bertaux and Delannoy* [1978] suffered from severe contamination problems. *Weinstock et al.* [1990, 1994] put great effort into avoiding contamination, but their instrument is also sensitive to solar radiation. For aircraft use, *Kelly et al.* [1989] successfully designed a closed cell, but its large volume needs high purge rates to become free of contamination and memory effects. Thus for use both on aircraft and balloons, the fluorescence cell of FISH is technically based on a similar design as that developed by *Kelly et al.* [1989] optimized with respect to a mimimal volume of the cell. It consists of an inner cube made of black glass (Schott NG1) providing surfaces with low adsorptivity of water. In order to avoid H_2O contamination by dead volumes the edges are not sealed. This cube is mounted in an electropolished vacuum-tight stainless steel housing. The glass plates are fixed 5 mm from the walls of the stainless steel housing. Figure 4 illustrates a cross section through the cell. Air from the corners of the inner cell is pumped through the rifts between the glass plates into the stainless steel cell and the exhaust in order to minimize contamination.

In order to minimize the background radiation from the Lyman α radiation source, the fluorescence cell has to trap the stray light in the field of view of the PMT. *Kelly et al.* [1989] therefore placed the PMT opposite to one of the corners of the cell. As stated above a goal of our design was to minimize the volume of the cell in order to minimize both the weight and the volume of air to be exchanged. The latter is particularly

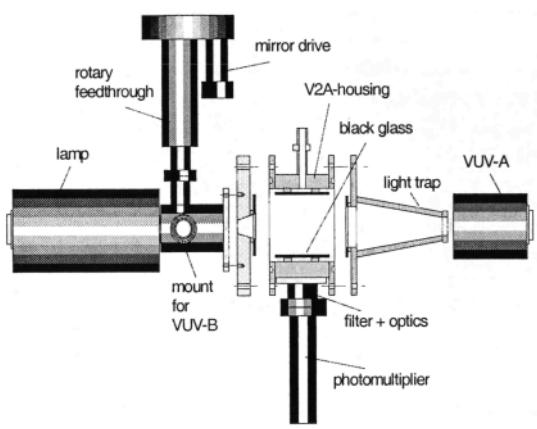

Figure 4. Cross section of the fluorescence cell equipped with the Lyman α radiation source, the photomultiplier tube (PMT) and both VUV detectors.

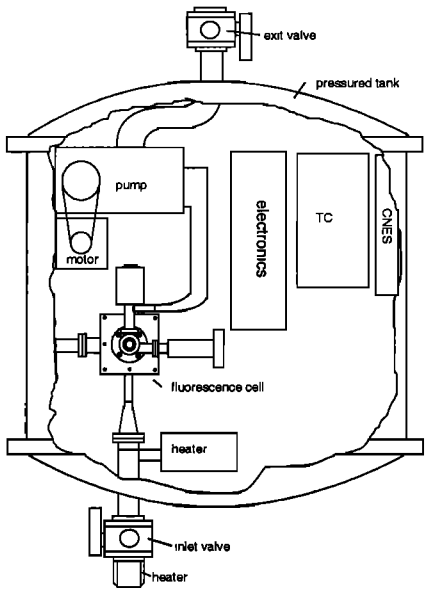

Figure 5. Cross section of the FISH balloon instrument.

advantageous when large gradients of H_2O abundances are measured as, for example, for studies of stratosphere-troposphere exchange. From a ray-tracing analysis and empirical tests we found that a cube with 50 mm length and a cone as a light trap opposite the Lyman α lamp are sufficient to reduce the background radiation to the contribution of scattering by air molecules and aerosol. The Lyman α radiation source and the PMT are mounted as indicated in Figure 4.

The air inlet of the cell is a ½ inch tube mounted directly at the cone to which the detector VUV-A is mounted. The ½ inch outlet tube is placed close to detector VUV-B. Thus the cell can be purged completely with fresh air, close to the detectors, which is essential to avoid measurement artefacts due to contamination. The total volume of the cell including the volumes in front of the detectors is approximately 0.3 L. Typical purge rates of FISH are of the order of 1 L(STP)/s (see below) so that the air in the cell is exchanged in less than 1 s. The inlet tubing of both the balloon and the aircraft instrument is heated with 100 W in order to avoid memory effects of H_2O contamination on the walls and to vaporize H_2O-containing particles.

The quartz window selecting the different measuring modes is driven by a dc motor via a rotary feedthrough that is mounted between the Lyman α radiation source and the fluorescence volume (see Figures 1 and 4). Its position is controlled via an angle encoder. The air in the cell is exchanged in less than 1 s which, as will be shown below, is also the proper integration time to obtain a sufficient signal-to-noise ratio in order to detect small-scale variations of H_2O in the atmosphere. Therefore the complete measurement cycle is chosen to consist of 12 steps each of 1 s duration: 10 steps with the quartz window in position 1 (measurement of fluorescence) are followed by one step in position 2 (monitoring the Lyman α intensity) and by one step in position 3 (measurement of background radiation).

3.2.4. Electronics. The instrument is controlled by a 386 or 486 processor unit driving several I/O interfaces to convert and store the analog and digital signals of the sensors and to control the mirror drive, heaters of the Lyman α radiation source and the inlet tubing, the purging of the Lyman α radiation source (aircraft instrument only), valves and the pump (balloon instrument only). The instrument is operated exclusively at 28 V dc and requires approximately 60 W plus the power for the heater for the inlet tubing and the pump for the balloon instrument.

For the balloon instrument, an additional interface transmits the data at 4096 bits/s via telemetry to be displayed in real time at the ground. A keyboard enables remote control of the instrument, for example, opening of the valves and adjustment of the frequency of the pump motor.

3.3. The Aircraft Instrument

Up to now, FISH has been used for airborne measurements on a Falcon-E aircraft, a Cessna Citation and a Lear jet. In these aircraft, FISH is mounted in the pressurized cabin in 19 inch type racks. The inlet on these aircraft is a forward directed ½ inch tube that allows for measurement of total water, that is, sampling H_2O in both the gaseous and the condensed phase. Determination of enhancement factors for particle measurement due to anisokinetic probing are discussed by C. Schiller et al. (manuscript in preparation, 1998). The ram pressure during flights with these aircraft is sufficient to maintain a flow of at least 0.5 L(STP)/s through the instrument even at the maximum altitude, that is, corresponding to 200 - 150 hPa. During a recent mission a rear facing probe in combination with a pump has also been used for gas phase measurements. During ascent and descent the flow through the instrument is shut down in the lower troposphere by closing valves in order to avoid major contamination.

Only a few components have to be added to the basic instrument described above: pressure and temperature sensors in the flow tube close to the fluorescence cell, and a flowmeter (MKS) to monitor the mass flow through the system. When using the purged lamp, the total weight of FISH for the use in aircraft is ~30 kg.

3.4. The Balloon Instrument

The balloon hygrometer is a more complex instrument. All components are mounted in a pressure-tight container of 750 mm diameter and 850 mm height made of aluminum. Thus the produced heat is transferred to the walls, avoiding thermal problems at ambient pressures down to 5 hPa during a balloon flight. The total weight of the balloon instrument FISH is about 80 kg without batteries. In Figure 5 a cross section of the balloon instrument is shown.

Since the balloon instrument is not used as intensively as the aircraft hygrometer, the sealed getter lamps are used as the Lyman α radiation source. Temperature and pressure of the air in the fluorescence cell are measured, the latter using a pressure sensor with a range from 1000 to 1 hPa and an accuracy better than 0.1 hPa (ParoScientific).

FISH measurements from a balloon are performed during a valve-controlled descent with typical rates of 2-5 m/s. H_2O measurements during ascent are contaminated by the water cloud of the balloon which is 120 m above the payload. For example, a contamination of up to 50 ppmv was observed during the first flight of the prototype of FISH during ascent

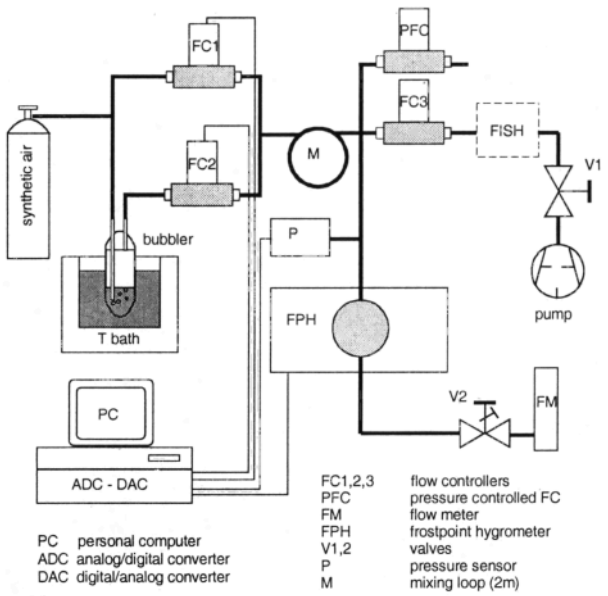

Figure 6. Sketch of the laboratory calibration bench for FISH. Via the flow controller FC1 and FC2, dry and wet air are mixed together within the mixing loop (M). The pressure-controlled flow controller (PFC) keeps the pressure constant inside the frost point hygrometer (FPH) which serves as a reference instrument. A constant flow of the mixed air is sent to FISH by FC3. By regulating the pumping speed with valve V1, the pressure inside FISH is varied. Valve V2 and the flowmeter (FM) ensure a constant air flow through the frost point hygrometer.

[*Mörschel et al.*, 1991]. With two valves at the inlet and the outlet the instrument can be kept closed during the ascent. Before a balloon launch, the instrument is purged with dry nitrogen at the ground. Thus the measurements can be started immediately at the ceiling altitude without a longer degassing period.

An Eaton roots blower driven by a brushless dc motor is used to maintain a sufficient airflow through the instrument so that contamination effects can be neglected. During the flight the frequency of this pump is adapted to the ambient pressure to allow for a quasi-constant mass flow of several L(STP)/min through the instrument. The diameter of the inlet tube is 40 mm and is reduced to the ½ inch system of the fluorescence cell through a conical diverter. The inlet line is designed to trap backscattered solar UV radiation that might interfere with the fluorescence signal. The complete flow channel is made of electropolished stainless steel. The inner surfaces are partially gold plated and heated, since H_2O desorbs completely from gold surfaces at temperatures of 50° [e.g., *Tam and Schroeder*, 1988].

The design of the prototype instrument described by *Mörschel et al.* [1991] is different in important details, in particular the complete flow system including the pump and the fluorescence cell. The high background of the old fluorescence cell yielded a very poor signal-to-noise ratio and subsequently long integration times. Using the new cell, which required a complete redesign of the entire instrument, improved the performance of the hygrometer significantly.

The balloon hygrometer is mounted in a gondola for protection against damage at touchdown of the payload. The gondola is an open frame structure made of aluminum tubes enabling a free flow to the FISH inlet from the bottom. Thus the inlet does not have to be extended below the gondola. The hexagonal structure of the frame allows us to combine several gondolas into a joint payload of several instruments. Currently, we combine FISH together with a cryogenic whole air sampler and a ClO/BrO resonance fluorescence instrument to form a joint payload called "Triple" for investigations in the lower and middle stratosphere.

4. Calibrations

4.1. Laboratory Calibrations

In order to calibrate the fluorescence hygrometer, a laboratory calibration facility with the capability to simulate realistic atmospheric conditions was developed. In particular, a wide range of H_2O mixing ratios from several hundred to a few ppmv and pressure from 1000 to approximately 10 hPa has to be provided. The absolute accuracy of the calibration should be of the order of 5%, that is, 0.2 ppmv or less for stratospheric mixing ratios.

The calibration bench (Figure 6) is purged with synthetic air (20.5% O_2) from a pressure bottle that typically contains 2-5 ppmv H_2O. This dry airflow is divided into two branches of which one is moistened in a temperature-stabilized H_2O bubbler. Via two flow controllers the airflows of both branches are mixed together in a mixing loop of 2 m length, producing variable H_2O mixing ratios in the desired range. At its end the H_2O mixing ratio is determined using a commercial dew point hygrometer (General Eastern 1311 DRX). The pressure is measured close to the dew point hygrometer with a temperature-stabilized baratron and electronically regulated to 2000 hPa. This pressure was chosen to extend the range of measurable mixing ratios of the dew point hygrometer down to 1-2 ppmv. The uncertainty of the dew point is 0.1 K, corresponding to an error of 3% in the H_2O partial pressure at 210 K. This error includes the uncertainty of the temperature dependence of the partial pressure itself as determined by *Marti and Mauersberger* [1993]. Assuming the error of the pressure determination to be 1%, the H_2O mixing ratio can be determined with an accuracy better than 4%.

FISH is flanged to the calibration facility via a flow controller (FC3 in Figure 6). Together with FC3 a pump mounted behind FISH enables the pressure in the FISH fluorescence cell to be reduced between 1000 and 15 hPa. The flow rate through FISH has to be kept at 5-10 L/min in order to obtain stationary conditions in the complete system within a couple of minutes.

The calibration system is controlled completely by a PC. After purging the system with dry synthetic air for 1 to 2 hours, the calibration is started with the lowest H_2O mixing ratio, which is then increased stepwise. At each mixing ratio level, the calibration is carried out for different pressures. Depending on the number of calibration points, a complete calibration cycle takes 3 to 5 hours. As an example, Figure 7 shows the calibration performed before the aircraft mission on March 10, 1997 using the calibration bench (solid symbols and regression line; open symbols are discussed below).

Using the laboratory calibration facility, FISH is calibrated usually before and after each flight. However, the calibration factor C^* in (3") does not change significantly from flight to flight; for example, during a test campaign of FISH on the Falcon aircraft for a period of two flights with together 8

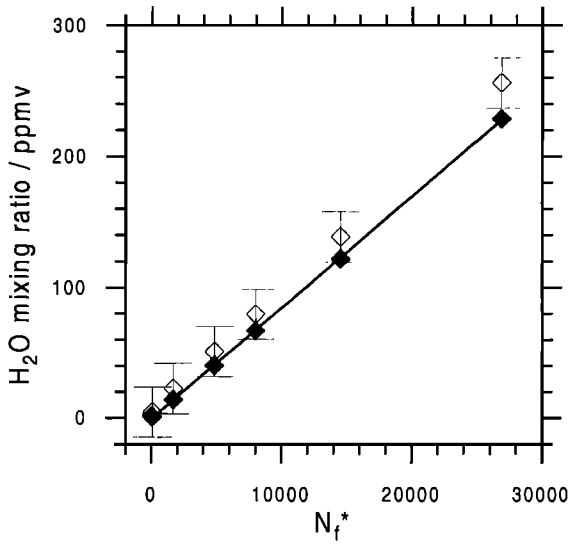

Figure 7. Relative count rate N_f^* as a function of the H_2O mixing ratio for the FISH calibration for the aircraft mission on March 10, 1997: Solid symbols and the corresponding regression line show the laboratory calibration before the flight using the frost point hygrometer as reference. Open symbols denote calibration points obtained in the same calibration cycle, but the H_2O mixing ratio is calculated from the simultaneous absorption measurement of FISH (see text), yielding approximately 15% higher mixing ratios.

operation hours of the Lyman α radiation source, C* varied by less than 3%, which corresponds to the accuracy of the dew point hygrometer. Therefore we are confident that the calibration factor does not change over the duration of a flight, and all short-term variations as those of the background and the Lyman α radiation are considered by the full measuring cycle. As a result of several field campaigns, the calibration factors have been proven to be almost constant for the duration of a campaign even when a new Lyman α radiation source has been used.

If FISH is used for measurements at altitudes above 20 km when the approximation $A_0 << k_q \cdot [air]$ is no longer valid, either the pressure dependence of C^* has to be determined during calibration or the data obtained at low ambient pressure have to be analyzed via equation (3).

4.2. In-Flight Calibrations

In addition to the laboratory calibration, FISH can be calibrated during in-flight operation by measuring the absorption of Lyman α radiation whenever the optical depth of H_2O is sufficient. For this purpose the detector VUV-A is mounted 150 mm from the Lyman α radiation source. In the upper troposphere at H_2O mixing ratios of the order of 100 ppmv, the absorption becomes measurable and provides an independent calibration of the fluorescence signal. *Kelly et al.* [1989] injected water vapor from a bottle for in-flight calibration at stratospheric altitudes via absorption measurement. However, due to our precise laboratory calibration and the proven stability of the calibration factors during a flight, such an option is currently not considered.

Taking into account the absorption by H_2O and O_2, the H_2O mixing ratio can be determined according to Beer's law

$$\mu_{H2O} \cdot [air] \cdot \sigma_{H2O} \cdot l = \ln \frac{C_A}{C_B} - \ln \frac{U_A}{U_B} - \sigma_{O2} \cdot l \cdot [O_2] \qquad (5)$$

with l the absorption length, σ_{H2O} and σ_{O2} the absorption cross sections of H_2O and O_2, respectively, C_A and C_B detector constants of the detectors VUV-A and VUV-B (the latter including mirror reflectivity as well), and $[O_2]$ the concentration of molecular oxygen. The absorption of other species as ozone, carbon dioxide and methane are not significant for this approach.

As an independent reference during a laboratory calibration, H_2O mixing ratios were calculated from the simultaneous absorption measurement of FISH using equation 5 (open symbols in Figure 7) and from the frost point hygrometer of the calibration bench (solid symbols). The calibration was performed at six different water vapor mixing ratios between 2 and 230 ppmv at 400 hPa. Here, σ_{H2O} and the pressure dependent σ_{O2} of *Kley* [1984] have been used, yielding deviations in the mixing ratio around 15% compared to the calibration bench. The error bars are determined only by the uncertainty of σ_{O2}; in fact, they become even larger considering the uncertainties of σ_{H2O} and of the instrumental constant $\ln(C_A/C_B)$.

A problem for the calibration by absorption measurement is the determination and uncertainty of the pressure dependent $\sigma_{O2}(\lambda \approx 121.6\ nm)$, which has been determined in the past by, for example, *Watanabe et al.* [1953] and *Kley* [1984]. The quantity required is an integral absorption cross section, that is, a convolution of the line shape of the Lyman α radiation source and the highly structured $\sigma_{O2}(\lambda)$ which has to be determined here specifically and may be different from values published by other groups. Unfortunately, the quantities σ_{H2O} (if not taken from the literature), σ_{O2}, and $\ln(C_A/C_B)$ have to be found simultaneously. For measurements under stratospheric conditions, the left-hand side of equation (5) becomes $<10^{-3}$ and is thus negligible. Measuring U_A and U_B as a function of $[O_2]$ determines $\ln(C_A/C_B)$ and σ_{O2}. In order to determine the concentrations [air] and $[O_2]$ both the pressure and the temperature in the cell are measured. The specific cross sections for our instrument σ_{O2} and σ_{H2O} that we determined from a set of calibration measurements under different conditions are approximately 10% higher than those of *Kley* [1984].

5. Performance and Technical Tests of FISH

In this section we briefly discuss measurements obtained in the laboratory and during the first field campaigns of FISH from balloons and aircraft in order to show its performance. For the scientific interpretation of the field data we refer to recent and forthcoming papers [e.g. *Zöger et al.*, 1996; *Zöger et al.*, 1997; *Zöger et al.*, this issue; also C. Schiller et al., manuscript in preparation, 1998].

5.1. Technical Performance Under Field Operation

The design of FISH aims to measure H_2O at stratospheric conditions with at least 5% precision (0.2 ppmv) within 1 s. For airborne measurements and a typical aircraft speed the hygrometer resolves horizontal structures of the H_2O mixing ratio of 200 m. Measurements from a balloon at valve-controlled descent rates from 2 to 5 m/s enable us to resolve

very detailed vertical structure; even during parachute descent, FISH is still sensitive to small-scale changes.

As described above, the exchange time of air in the cell and the detection steps are set to 1 s. A statistical analysis of laboratory measurements yields the following characteristics of the hygrometer. For an integration time of 1 s, the noise equivalent mixing ratio at 3 ppmv for typical light sources is determined to be 0.20-0.15 ppmv, and the detection limit is 0.18-0.13 ppmv. However, the signal-to-noise ratio and the detection limit critically depend on the quality of the lamps and the subsequent count rates of the fluorescence and the background ratio; these criteria finally define the selection of the Lyman α radiation sources.

During measurements of FISH from the Falcon aircraft in March 1993 over Germany we determined the noise of the instrument for field deployment. Via an autocovariance analysis the atmospheric variability was separated from the instrumental noise [e.g., *Giez et al.*, 1998]. At different flight levels the noise was found to be 0.22 ppmv at an average H_2O mixing ratio of 8.6 ppmv, 0.19 at 7.2 ppmv, and 0.16 ppmv at 5.9 ppmv. This result is in good agreement with the values obtained in the laboratory. These noise levels are further reduced using the new purge lamps.

5.2. Intercomparison With Other H_2O Measuring Techniques

As described in section 4.1, the accuracy of the laboratory calibration is better than 4%, corresponding to approximately 0.2 ppmv at stratospheric mixing ratios. However, in the past sometimes unexpected discrepancies between different hygrometers during field operation were found that are of the order of 0.5 ppmv and therefore exceed the assumed accuracy of each individual instrument [e.g. World Meteorological Organization (*WMO*), 1986; *Dessler et al.*, 1994; *Vömel et al.*, 1995]. The reason for these striking discrepancies is still unknown, whether they are caused by instrumental or sampling artefacts. Therefore intercomparison of H_2O measurements in the atmosphere is essential to detect potential larger uncertanties. The improvement of the accuracy of H_2O measurements hence has to be a major issue of future activities.

The balloon flight of the prototype instrument of FISH in September 1993 allowed for intercomparison with a nearby overpass of the Halogen Occultation Experiment (HALOE) on the Upper Atmospheric Research Satellite. H_2O mixing ratios measured by HALOE (version 18) are only slightly lower by 3% than our in situ data, a discrepancy that is not significant within the combined error bars. A detailed discussion of this flight is given by *Zöger et al.* [this issue]. Furthermore, the quantity $2{\cdot}CH_4 + H_2O$ obtained during this flight is compared with data measured during the European Arctic Stratospheric Ozone Experiment (EASOE) using a frost point hygrometer and the same whole air sampler to measure CH_4 [*Engel et al.*, 1996]: From both data sets, $2{\cdot}CH_4 + H_2O$ agrees within 5%. Assuming $2{\cdot}CH_4 + H_2O$ to be constant for both periods and locations provides indirect evidence that both hygrometer measurements are in good agreement.

A direct intercomparison with a frost point hygrometer [*Busen and Buck*, 1995] on board the aircraft was performed during the aircraft mission in March 1995. FISH and the frost point hygrometer were probed by the same air inlet in order to avoid discrepancies caused by potential sampling artefacts. In Figure 8, both data sets are plotted as a function of flight time. Some oscillations of the frost point hygrometer occur that have been observed previously [*Ström et al.*, 1994] and can be explained by electronic control problems of the instrument. Despite these oscillations the agreement of both measurements is 5% over the whole range of H_2O mixing ratios from 8 to 280 ppmv when the frost point hygrometer was operational. The range of mixing ratios below 15 ppmv is expanded in Figure 8 to show the intercomparison in this critical and important range more clearly: The frost point hygrometer signal oscillates with a minimum amplitude of 1 ppmv and a period of the order of 1 min around the signal of FISH, which is able to resolve true atmospheric changes on sub-ppmv levels. It has to be stated here that the electronic control of the frost point hygrometer was not optimized for stratospheric measurements.

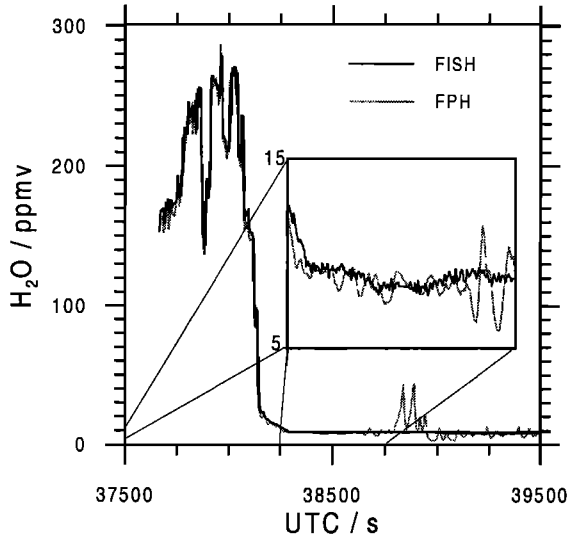

Figure 8. Intercomparison of simultaneous H_2O measurements of FISH and a frost point hygrometer on the Falcon aircraft (R. Busen, private communication, 1995), showing very good agreement within 5% despite some oscillations of the frost point hygrometer below 15 ppmv.

6. Discussion / Summary

FISH, the fast in situ stratospheric hygrometer developed at the Forschungszentrum Jülich, is a modern version of a hygrometer based on the Lyman α photofragment fluorescence technique. The design allows for flexible use on balloons and different aircraft. The measurements can be accomplished with a precision < 0.2 ppmv at 1 s integration time as demonstrated both in the laboratory and under field operation. The fast purging through the instrument and the choice of the materials enable a measurement free of contamination even in the region of large gradients of the H_2O mixing ratio close to the tropopause. Compared to other advanced fluorescence hygrometers with similar performance, the dimensions and the weight of FISH were further reduced. Several improvements of individual components have been developed, in particular the technology of the Lyman α radiation sources, which has been described elsewhere in detail [*Woyke et al.*, 1995; *Tuitjer*, 1997]. The sophisticated

VUV radiation techniques, the problem of contamination when measuring low H_2O mixing ratios and the required accurate calibration still require high scientific and engineering effort. However, the instruments now allow reliable and frequent operation during field campaigns without major maintenance. Data processing including actual calibration constants can usually be accomplished within a few hours after a flight.

The novel laboratory calibration bench allows for calibration of FISH measurements with approximately 4% accuracy. The excellent agreement in the first field intercomparisons with other measurements confirmed this value, even at low stratospheric mixing ratios of a few to 10 ppmv. Since the calibration is carried out at realistic stratospheric conditions, the hygrometer can be characterized in detail before field operation.

Up to now, only the advanced TDL techniques as used in the hygrometer by *May* [1998] have proven that similar performance for stratospheric H_2O measurements can be achieved. These techniques might be advantageous for specific applications, for example, when even higher time resolution (i.e., << 1 s) is required. However, the technique of Lyman α photofragment fluorerescence that has been used for about two decades by different groups and improved several times will in the future also provide a powerful tool to measure water vapor over a large range from the free troposphere to the stratosphere. It enables highly precise and accurate process studies with sufficiently high time resolution for most applications. The existence of several advanced and well-characterized hygrometers based on different independent in situ techniques is essential for a successful investigation of those scientific questions involving atmospheric water vapor.

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