University of Leipzig
Institute for Meteorology

Water Vapor Retrieval in the Upper Troposphere and Lower Stratosphere Using Airborne Measurements of Spectral Solar Irradiance

Author: Peter Stammer

Reviewer: Prof. Dr. Manfred Wendisch,
Second Reviewer: Prof. Dr. Andreas Macke

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1 Introduction

Water vapor is one of the major trace gases of the Earth’s atmosphere and has a substantial impact on different processes in the atmosphere. It plays a key role in weather processes, as it transports latent heat and forms clouds. Clouds strongly interact with shortwave and longwave radiation fluxes. Water vapor also strongly influences the Earth’s radiative energy budget, because it is a strong greenhouse gas and absorbs large fractions of longwave radiation in the atmosphere (Jacob, 2001). The increase of tropospheric water vapor with a warming climate and the related climate feedback are well understood and simulated in climate models. In contrast, processes controlling the distribution and variability of stratospheric water vapor are represented in climate models only with limited accuracy (Solomon et al., 2010).

Water vapor has a relatively short lifetime in the atmosphere, due to the strong temperature dependency of the saturation water vapor pressure. The water vapor mixing ratio \( w \) of different standard atmospheres defined by Anderson et al. (1986) are shown in Figure 1.1. At the surface \( w \) can vary by an order of magnitude between different latitudes and seasons. The water vapor mixing ratio quickly reduces with increasing altitude and has a local minimum at the tropopause that is three to four orders of magnitude lower than at the ground. In the stratosphere water vapor is distributed much more uniformly than it is in the troposphere. The differences between the tropospheric and the stratospheric water vapor illustrate, that the exchange of water vapor through the tropopause is limited.

The troposphere and the stratosphere are two distinctly different regions in the atmosphere with regard to radiative and dynamical processes. Traditionally, the two regions are thought as being separated clearly by the tropopause surface. From recent research, it becomes clear that the separation more realistically should be regarded as a gradual transition from the troposphere to the stratosphere within the upper troposphere and lower stratosphere (UTLS) (Gettelman et al., 2011). The UTLS extends approximately ±5 km around the tropopause (Gettelman et al., 2011). There is commonly a classification between the UTLS in the tropics, referred to as the tropical tropopause layer (TTL), and the extratropical UTLS (Ex-UTLS). The distinction arises, among other reasons, from differences in the driving processes, radiative-convective balance in the TTL (Gettelman et al., 2011).
Figure 1.1: Vertical profiles of water vapor mixing ratio in ppmv of model atmospheres for tropics, midlatitude summer, sub-arctic winter and the US standard atmosphere for the entire troposphere, stratosphere and mesosphere (a) and a close of the the tropospheric water vapor (b). Profiles are based on a study by Anderson et al. (1986).

and wave dynamics in the Ex-UTLS (Fueglistaler et al., 2009). The UTLS is characterized by a layer of enhanced static stability, the tropopause inversion layer (TIL), located just above the thermal tropopause.

Because of the enhanced stability in the UTLS, the distribution of water vapor in the stratosphere and mesosphere are largely decoupled from the troposphere, as described by the Brewer-Dobson circulation. The Brewer-Dobson circulation is a description of the general circulation in the stratosphere and mesosphere, and the exchange between the stratosphere and the troposphere, that was first proposed by Brewer (1949) and Dobson (1956). Figure 1.2 shows a pressure versus latitude cross-section of temperature and water vapor in the atmosphere and illustrates aspects of the Brewer-Dobson circulation. The circulation consists of a general flow of stratospheric air from the tropics to the poles, thus pulling air upwards over the tropics and pushing air downwards in the extratropics, as indicated by the arrows in the right panel of Figure 1.2. The driving force of the poleward flow is the dissipation of rossby and gravity waves originating in the extratropical troposphere (extratropical pumping) (Holton et al., 1995). The Brewer-Dobson model implies that air enters into the stratosphere from the troposphere exclusively through the tropical tropopause. Because of the upward motion of air over the tropics, the tropopause is caused to be particularly cold in this region compared with extratropical latitudes (see Figure 1.2). When air crosses the tropical tropopause it is “freeze-dried”, meaning it is strongly cooled down, water vapor condenses and is extracted, thereby reducing $w$ to the saturation limit $w_s$ at the tropopause. The minimum of $w$ is located over the tropics, while a gradual increase in moisture toward higher altitudes and latitudes is observed. The photochemical oxidation of methane and the small-scale diffusion of water vapor through the extratropical tropopause are two sources of water vapor, that cause a humidification of the
stratospheric air with time (Remsberg et al., 1984). In the literature the oxidation of methane is discussed to be the major source of stratospheric water vapor especially at higher altitudes, aside from variations in the intrusions of water vapor through the tropical tropopause (Solomon et al., 2010).

Holton et al. (1995) point out that dynamical, chemical, and radiative coupling between the stratosphere and the troposphere are important processes that must be understood for prediction of global climate, and the UTLS bares relevance as the exchange zone between both spheres. Water vapor has an influence on the UTLS. As an example, there is evidence that the vertical structure of water vapor, and particularly radiative cooling by water vapor near and immediately above the tropopause, maintain the TIL (Randel et al., 2007). This is underlined by the fact that local radiative cooling by water vapor is the primary cause for a particularly strong TIL in the polar summer (Randel and Wu, 2010). In the context of transport, specifically the TTL is a significant gateway for trace gases such as water vapor into the stratosphere. Therefore, understanding dynamic and radiative processes in the TTL is important for predicting and modeling the stratosphere. Many processes of the TTL are not well understood. Although the Brewer-Dobson circulation is generally accepted as the reason for low water vapor content in the stratosphere, there is still active debate on the details of the motion of tropical air into the stratosphere and the drying mechanism (Holton et al., 1995). E.g.
there is an open question which needs to be answered, as to whether the tropical tropopause is cold enough to explain the dryness observed in the stratosphere (Plumb, 2002). As dehydration and transport in the TTL are coupled processes, water vapor is an important tracer for examining the TTL. Observed increases in stratospheric water vapor still cannot be fully reconciled with current model calculations (Fueglistaler et al., 2009). Understanding all processes that control the TTL, and incorporating them in models, is an important prerequisite for reliable predictions of changes in the TTL in a changing climate and for predicting how the TTL feeds back on the climate (Fueglistaler et al., 2009).

1.1 Relevance of lower stratospheric water vapor

1.1.1 Observations and Climatology

Stratospheric water vapor is typically observed via methods of radio sonde, in-situ, or satellite measurements. The two most relevant time series of stratospheric water vapor are based on radiosonde measurements conducted in Boulder and on satellite measurements by the Halogen Occultation Experiment (HALOE). The radio soundings began in 1980 and are still operated (Scherer et al., 2008). It is the longest time series and the only continuous observation that spans multiple decades. The stratospheric water vapor measurements are done on the principal of the frost point hygrometer. Due to the low saturation water vapor pressure in the stratosphere even a small uncertainty in measuring the mirror temperature results in a relatively large uncertainty in \( w \). Vömel et al. (2007) obtain an uncertainty of 10% for \( w \) obtained in the lower stratosphere.

HALOE retrieved profiles of \( w \) through solar occultation between 15 and 80 km with a spatial coverage from 60° N to 60° S and a vertical resolution of 1.6 km. Occultation was performed at 6.61 \( \mu \)m. HALOE was conducted from 1991 to 2005 (Scherer et al., 2008). The retrieval of \( w \) has an uncertainty of 14% to 24% in the lower stratosphere (at altitudes up to 10 hPa) (Grooss and Russell III, 2005).

Rosenlof et al. (2001) give an overview of several observations made irregularly between 1954–2000. These include airborne in-situ measurements made by frostpoint hygrometry or Lyman-\( \alpha \) fluorescence. Examples of instruments used for airborne in-situ measurements are the Fast In-situ Stratospheric Hygrometer (FISH) and the Hygrometer for Atmospheric Investigation (HAI). Both have been operated on board the High Altitude and Long Range Research Aircraft (HALO). FISH is a closed-path hygrometer, which uses the principle of Lyman-\( \alpha \) photofragment fluorescence, as described by Meyer et al. (2015). Water molecules of the surrounding air are split into an excited OH molecule and a H atom inside a
chamber by Lyman-α radiation. While relaxing back to the ground state the OH molecules emit radiation in the shortwave range. From the magnitude of this radiation the water vapor mixing ratio is determined. HAI is an instrument that uses tunable diode laser absorption spectroscopy. Water vapor absorption is probed at two laser signals with the wavelengths 1.4 and 2.6 µm (Buchholz et al., 2017).

Hurst et al. (2011) evaluated the observations from the radio soundings for the time period 1980–2010 and for an altitude coverage of 16 to 26 km. During this period, w showed a variability within the range of 3.0 − 5.5 ppmv. The general trend for the three decades was an increase of 1.0 ± 0.2 ppmv (or 27 ± 6 %). This increase was interrupted by a significant drop between 2001-2005 by -0.35 ± 0.04 ppmv on average. Rosenlof et al. (2001) found a general increase in stratospheric water vapor by 2 ppmv for 1954–2000, or 1% per year. This is in line with the trend found by Hurst et al. (2011).

The radio sonde and HALOE observations have been compared for the overlap period 1991–2005 and several studies have noted that there is a significant disagreement between the two studies (Randel et al., 2004; Scherer et al., 2008). Figure 1.3 shows a time series of the radio sonde measurements between 1980–2004 and of the HALOE retrieval between 1991–2004, both for the average w for the altitude range 17–22 km. Randel et al. (2004) notice reasonable agreement for 1992–1997 and for the relative drop after 2001, but there is disagreement after 1997. While the radio sonde data shows a general rise in water vapor concentration, the HALOE retrieval shows a constant or decreasing trend (Figure 1.3). The study could not state the reason for this discrepancy. Scherer et al. (2008) concluded that the differences between both data sets made a quantification of water vapor entering into the stratosphere impossible.

The mechanisms behind the observed variations and long-term trends of lower stratospheric water vapor are a matter of ongoing debate (Hurst et al., 2011; Kindel et al., 2015). Randel et al. (2006) give evidence that the drop after 2001 was connected to a prolonged period of enhanced tropical upwelling and a colder tropical tropopause. There is a strong correlation between seasonal variations in tropical tropopause temperatures and stratospheric water vapor (Randel et al., 2004). However, the same is not true in the case of the long-term trend suggested by the Boulder data. The observed long-term trend of the cold-point temperatures at the tropical tropopause shows cooling instead of warming, which should contribute to a decrease in stratospheric water vapor (Randel et al., 2004). There is an increase in tropospheric methane entering the stratosphere due to industrial activity. However, Hurst et al. (2011) found that increasing methane could explain only up to 28% of the overall increase in stratospheric water vapor between 1980–2010.
As a result of these uncertainties, it is still unclear whether changes in stratospheric water vapor are a long term trend and possibly represent a source of climate feedback or climate forcing, or whether they caused by unforced internal climate variability (Maycock et al., 2013). Dessler et al. (2013) support and give evidence for the idea that a stratospheric water vapor feedback exists, by which stratospheric water vapor increases when temperatures in the troposphere warm. However they point out that there is insufficient understanding of processes that regulate water vapor in the lower stratosphere and that more work is needed to improve our understanding. There are projections for a significant increase of the stratospheric water vapor content over the next century in response to increasing greenhouse gases. However, these projections are considered to be highly uncertain (Maycock et al., 2013).

1.1.2 Radiative, dynamical and climatological impacts

While there are limited observations of the content of water vapor in the lower stratosphere and limited understanding of the factors that drive it, several studies...
give evidence that, despite its sparse occurrence, stratospheric water vapor has important impacts on the stratosphere’s radiative budget and dynamical processes, and on the temperature in the troposphere and at the surface. Already small changes in stratospheric water vapor potentially have a large impact on these factors.

Water vapor impacts the local temperature in the atmosphere via absorption and emission of longwave radiation. This impact depends on the balance between absorption and emission. Stratospheric water vapor produces a local cooling (Forster and Shine, 1999) because the additional emission following an increase in water vapor outweighs the additional absorption of radiation from the ground, which to a large extent is already absorbed by the lower atmospheric layers. Maycock et al. (2013) simulated the radiative temperature change in the stratosphere after a doubling of the water vapor from a uniform vertical distribution above the tropopause of 3 ppmv to 6 ppmv. The result was a general cooling throughout the stratosphere, with a maximum cooling of 5-6 K in the polar lower stratosphere and 2-3 K in the tropical lower stratosphere.

The strengthened radiative cooling by stratospheric water vapor toward higher latitudes is an energy source for dynamical processes. An increase in stratospheric water vapor is generally expected to strengthen the stratospheric circulation. The doubling of stratospheric water vapor by Maycock et al. (2013) strengthens the Brewer-Dobson circulation by ~10%. The same study also found a significant effect on the jetstreams, with e.g. a more westerly flow in the subtropical jets. Dynamical effects by a change of total water vapor were caused almost entirely by water vapor in the lower stratosphere, while water vapor changes in higher levels of the stratosphere are less effective.

Solomon et al. (2010) performed calculations of the radiative forcing and the relative implications of water vapor for the 30-year time span after 1980. For an observed -0.4 ppmv drop after the year 2000 (~10%) the study simulated a radiative forcing of -0.098 W m⁻². In comparison the radiative forcing due to the growth in CO₂ from 1996 to 2005 was +0.26 W m⁻². The water vapor decrease slowed warming from the increase in well-mixed greenhouse gases and aerosols from 1996 to 2005 by 25%. For an increase of 1 ppmv between 1980-2000 the simulated forcing was +0.24 W m⁻². The resulting warming is 30% of the warming caused by well-mixed greenhouse gases during 1980-2000. Solomon et al. (2010) conclude that stratospheric water vapor is an important driver of decadal global surface climate change. Similar results are given by Forster and Shine (2002).
1.2 Objectives of the thesis

There is clearly demand for more observations of water vapor in the UTLS. To resolve the fine horizontal and vertical scales in the UTLS it is important to obtain measurements of water vapor with a spatial resolution of at least 100 km in the horizontal and 1 km in the vertical (Shepherd, 2007). Measurements of water vapor content in the UTLS are complicated. At these high altitudes, measurements with a high spatial and temporal coverage are challenging. Furthermore, the low water vapor concentration of only several parts per million requires different measurement approaches compared to the troposphere.

Satellites have limited vertical resolution, while radio sondes have a low spatial coverage. High-flying aircraft that can reach into the lower stratosphere enable the observation of the UTLS with a high spatial resolution, both in the vertical and the horizontal, and a large spatial coverage. Aircraft measurements are an important tool for evaluating the representation of the UTLS in chemistry-climate models (Hegglin et al., 2010) and for satellite validation (Gettelman et al., 2011). Besides in-situ measurements, airborne solar irradiance measurements provide another approach for remote sensing of the integrated water vapor (IWV) of the atmospheric air column above the UTLS region. This remote sensing deploys the absorption bands of water vapor in the solar spectral range.

1.3 Outline

Chapter 2 will give a description of the principle of differential optical absorption spectroscopy, upon which the retrieval is based. It will also give a brief overview of past applications of the method for retrieving the IWV from high-flying aircraft. The algorithm developed in the scope of this work will be introduced and questions concerning the sensitivity of the method will be discussed.

In chapter 3 the HALO and its onboard spectrometer system will be described. Two measurement campaigns which HALO performed in 2016 will be introduced.

In chapter 4 solutions for applying the retrieval to the HALO measurements will be presented.

Chapter 4.3 will deal with components that go into measurement uncertainty of the spectral irradiance and the resulting total uncertainty of the IWV retrieval.

Chapter 5 will display time series of retrieved IWV for several flight periods. These flight periods are from two different flights and were made under different flight altitudes and latitudes, so that the retrieval is applied to a range of conditions with regard to IWV.
2 Retrieval of Water Vapor from Solar Radiation

Measurements of downward solar radiation have reportedly been applied for remote sensing the water vapor column of the atmosphere in several instances (Houghton and Seeley, 1960; Kindel et al., 2015). This form of remote sensing generally relies on the principle of differential optical absorption spectroscopy (DOAS), which will be introduced here.

2.1 Differential optical absorption spectroscopy

The principles of differential optical absorption spectroscopy are thoroughly discussed by Platt and Stutz (2008).

The retrieval of water vapor by means of DOAS relies on the absorption of solar radiation by atmospheric water vapor. Figure 2.1 illustrates the radiative transfer through the atmosphere. The incident solar radiation at the top of the atmosphere (TOA) is quantified to the initial spectral irradiance $F_{\lambda,0}$ with respect to a unit area that is perpendicular to the direction of propagation. The radiation propagates through the atmosphere by a certain optical path length $s$. Depending on the solar zenith angle $\theta$ it reaches a specific altitude $z = TOA - dz$, with $dz = s/\mu$ and $\mu = |\cos \theta|$. Over this length the irradiance is attenuated from $F_{\lambda,0}$ to $F_{\lambda}(s)$ through absorption and Rayleigh and Mie scattering by various atmospheric gases.

![Figure 2.1: Outline of quantities of Lambert-Beer’s law.](image-url)
and particles. In the following, the focus will be solely on absorption by water vapor. The resulting attenuation through water vapor absorption is, as described by Lambert-Beer’s,

$$ F_{\lambda}(s) = F_{\lambda,0} \cdot e^{-\tau} \quad (2.1) $$

For the optical thickness $\tau$ the following holds,

$$ \tau = \int -b_{\text{abs}}(s) ds = \int -k_{\text{abs}} \cdot \rho_{\text{wv}}(s) ds = -k_{\text{abs}} \cdot IWV \quad (2.2) $$

The optical thickness $\tau$ thus equals the integral of the water vapor absorption coefficient $b_{\text{abs}}(s)$ over $s$. $b_{\text{abs}}(s) = k_{\text{abs}} \cdot \rho_{\text{wv}}(s)$ is the product of the mass absorption coefficient $k_{\text{abs}}$ of WV and the absolute humidity $\rho_{\text{wv}}(s)$.

The integrated water vapor (IWV) results when integrating the absolute humidity over the entire column,

$$ \int \rho_{\text{wv}}(s) ds = IWV \quad (2.3) $$

and it has the units kg m$^{-2}$.

Based on Eq. (2.2), IWV can be derived from the ratio between the irradiance at the TOA and the irradiance at $z$, i.e. before and after absorption has occurred, at a wavelength $\lambda_{wv}$ at which water vapor absorbs radiation in an absorption band.

$$ IWV = -\frac{1}{k_{\text{abs}}} \ln \left( \frac{F_{\lambda}(s)}{F_{\lambda,0}} \right) \quad (2.4) $$

Figure 2.2 illustrates the various water vapor absorption bands in the visible and near-infrared (NIR). The visible spectrum has only relatively weak absorption, the strongest absorption band being at 0.72 $\mu$m. The NIR radiation is more strongly affected by water vapor absorption in several absorption bands. The bands in this range are located at 0.82 $\mu$m, 0.94 $\mu$m, 1.1 $\mu$m, 1.37 $\mu$m, 1.87 $\mu$m, and 2.6 $\mu$m. The absorption bands in the NIR are not suitable for ground-based remote sensing, Figure 2.2: Spectral transmissivity of the water vapor component of the US standard atmosphere under a zenith angle of 0° (Wendisch, 2017).
because they are saturated at the surface, meaning that all radiation has been absorbed at the center of the respective absorption band before the radiation reaches the surface. This is not the case for the weaker bands in the visible range, making them suitable for ground-based remote sensing. At higher altitudes, such as in the UTLS, the stronger bands in the NIR are no longer saturated and are better suited than the weaker bands in the visible given the low water vapor content that is encountered there. The 1.37 µm and 1.87 µm bands will both be considered for retrieving the range of IWV at the UTLS.

In absorption spectrometry a measurement of the irradiance $F_{0,\lambda}$ without the absorbing matter is compared with a measurement of the irradiance $F_{\lambda}$ with the absorbing matter. However, for atmospheric measurements using the Sun as the source of radiation, as in the situation described above, it is not possible to directly measure $F_0$. Even if the irradiance $F_0$ that is incident at the TOA is known, there are other processes than the absorption by the trace gas that contribute to an extinction of measured radiation. These processes, as given by Platt and Stutz (2008), are absorption and scattering by the various other components in the atmosphere besides water vapor, effects of the measuring instrument that reduce transmissivity, and light beam widening due to fluctuations in the refractive index caused by turbulence in the atmosphere. In order to apply Lambert-Beers law as in Eq. (2.1) and Eq. (2.2) to retrieve the IWV, it would therefore be necessary to quantify all the other processes that attenuate the solar radiation, which is not possible. Differential optical absorption spectroscopy solves this problem by

![Figure 2.3: a) Initial intensity ($I_0$), new initial intensity after broadband extinction has taken place ($I'_0$), and differential optical density of the trace gas ($D'$) b) Broad band absorption cross section ($\sigma_b$) and differential absorption cross section ($\sigma'$) which together add up to the total absorption cross section of the atmospheric column (Platt and Stutz, 2008)](image-url)
observing the difference absorption occurring at several wavelengths. Figure 2.3 illustrates the two components of total extinction, namely the trace gas absorption and the broadband extinction, as well as the process of distinguishing the two. While absorption by the trace gas occurs only in a narrow band, the other processes that cause extinction generally act spectrally neutral. Therefore, the narrow band trace gas absorption can be isolated from the broad band extinction from observations outside the trace gas absorption band. Instead of $F_0$, the broadband extinction spectrum is used as the new intensity spectrum of reference $F_0'$.

Figure 2.4 shows the spectral transmissivity of the US standard atmosphere at 10 km in the NIR spectral range. The blue line is the simulated transmittance of a model atmosphere considering all atmospheric gases besides water vapor, while the black line includes water vapor. In the regions where both lines differ absorption by water vapor is pronounced. In this spectral range also several absorption bands by O$_2$ and CO$_2$ are located, which are denoted by grey and blue shading. However, at the centers of both water vapor absorption bands, denoted by red lines, practically all molecular absorption is due to water vapor and not affected by O$_2$ or CO$_2$. These absorption band centers are located at $\lambda_{wv1} = 1363$ nm and $\lambda_{wv2} = 1873$ nm.

The green lines in Figure 2.4 denote examples of wavelengths that are close to the absorption bands, at which the atmosphere is practically free of molecular absorption from water vapor, as well as all other atmospheric gases. These wave-
lengths are 1235 nm, 1553 nm, and 1704 nm. Because they are used to scale the spectral irradiance they are referred to as $\lambda_{sc1}$, $\lambda_{sc2}$, and $\lambda_{sc3}$. The incident spectral irradiance $F_{0,\lambda}$ at the TOA is known theoretically and a spectrometer measurement results in $F_{meas,\lambda}(s)$, as in Figure 2.5(a). The narrow band water vapor absorption in the $F_{meas,\lambda}(s)$ can be isolated from broadband extinction by scaling $F_{0,\lambda}$ to $F_{meas,\lambda}(s)$ at the scaling wavelengths. Therefore, the scaled spectrum $F'_{0,\lambda}$ is obtained. As a result, the spectral transmissivity

$$T_\lambda = \frac{F_{meas,\lambda}(s)}{F_{0,\lambda}}$$

must equal unity at $\lambda_{sc1}$, $\lambda_{sc2}$, and $\lambda_{sc3}$, as in Figure 2.5(b).

### 2.2 Previous work

Houghton and Seeley (1960) performed an early study with DOAS for remote sensing of the IWV above an aircraft flying over England between 12 and 13 km altitude. The retrieval was done from observations in the water vapor absorption band at 2.7 $\mu$m. They determined an average IWV of 0.0045 kg m$^{-2}$ for 13 individual spectroscopic measurements. Assuming $w$ is uniform throughout the vertical column, this corresponded to a water vapor mixing ratio of $\sim$3 ppmv, which is about the climatological value of stratospheric water vapor from Hurst et al. (2011) or Remsberg et al. (1984). The uncertainty of $w$ in this study was about ±1 ppmv.

![Figure 2.5: Modeled irradiances for the near infrared (left panel) and inferred transmittance spectra (right panel). Vertical green lines denote water vapor absorption-free wavelengths. (after Kindel et al., 2015)](image)
Kaufman and Gao (1992) proposed a method for retrieving the IWV of the total atmospheric column from measurements with the Moderate Resolution Imaging Spectrometer (MODIS) instrument aboard the Earth Observing System (EOS). They used upward solar irradiance that has passed through the atmosphere, has been reflected on the Earth’s surface and passed up through the atmosphere again. The retrieval used a ratio of the irradiance measured in the 940 nm channel, located in a water vapor absorption band, to the irradiance measured in the 865 nm channel, which is free of water vapor absorption. The study deployed the LOWTRAN 7 radiation model code. Sources of uncertainty included uncertainties in the reflectance of the Earth’s surface, temperature and moisture profile, and several instrument technicalities, including sensor calibration, pixel registration and shifts in the channel location. These technical aspects are also relevant for this work and will be elaborated in later sections. Error analyses revealed a total uncertainty between $\pm 7\%$ and $\pm 13\%$. They thereby concluded that the operation is feasible for remote sensing of the IWV of the total atmospheric column.

Kindel et al. (2015) used measurements by the solar spectral flux radiometer (SSFR) in the 1.4 and 1.9 $\mu$m absorption bands to retrieve the water vapor in the tropical tropopause region off the coast of Southern California. The instrument’s uncertainty is between 1–3% (Pilewskie et al., 2003), without taking errors from aircraft movement into consideration. The NASA Global Hawk performed several profiles between $\sim 14$ and 18 km altitude, during which the SSFR measured downwelling solar irradiance. Simultaneously, in-situ measurements of $w$ were taken during the profiles.

Directly before and after each profile the aircraft performed several minutes of horizontal flight, during which 5–10 minutes of flight, or several hundred irradiance spectra, were averaged to obtain the average downwelling irradiance. Although the study reports the overall uncertainty of the absolute spectrometer measurement at 5%, it reports a drastic reduction to only 0.1% due to this temporal averaging. From the averaged spectral irradiances at the low and high end of the profiles, transmittance spectra of the atmospheric layer were created from the ratio of both irradiances ($T = F_{14km}/F_{18km}$). The water vapor profiles from the in-situ measurements were used as input for model simulations of the layer spectral transmittance with the radiative transfer model MODTRAN5. The simulated and measured transmittance were then compared for validation of the measured transmittance, in order to validate the capability of the spectrometer to detect the water vapor absorption signal within the layer.

An example of a comparison of measured and simulated transmissivity is given in Figure 2.6. One can see that there is noise in the measured signal and that the simulation shows higher absorptivity in the 1.4 $\mu$m and the 1.9 $\mu$m band compared
to the measured spectra. The amount of water vapor within the profiles is quite low and on average the resulting absorptance was less than 2% at the center of both absorption bands. Despite this small signal the study found that the agreement of the measured and simulated transmittances was within 0.002 for the 1.4 $\mu$m band and between 0.003 and 0.004 for the 1.9 $\mu$m. These differences were translated into an uncertainty of the IWV throughout the layer of 7–27% for the 1.4 $\mu$m band and 12–44% for the 1.9 $\mu$m band. The study thus concluded that it is possible to accurately determine the IWV within the profiled layers.

Kindel et al. (2015) also consider the potential of retrieving the IWV of the entire overlying atmospheric column between the aircraft and the TOA from a theoretical point of view. The 0.002 mean difference between measurements and simulations was applied to the retrieval of total IWV at altitudes of 14–20 km in a model atmosphere. The resulting uncertainty of IWV was $\sim 10^{-4}$ kg m$^{-2}$. For comparison, this is 1% of the total water vapor amount of the US standard atmosphere above 12 km as provided by Anderson et al. (1986). They conclude that the application of the DOAS method to the entire atmospheric column above the UTLS is also within the technical capabilities of the irradiance measurement.

### 2.3 General algorithm

The IWV retrieval uses simulations of the downward solar irradiance, which are performed with the library of radiative transfer routines libRadtran, version 2.0.2 (Emde et al., 2016). Calculation of radiative transfer in LibRadtran is based pri-
arily on the DIScrete ORdinate Radiative Transfer solver. To derive the IWV from an irradiance measurement, simulations need to be performed for identical atmospheric and geometric conditions. Therefore all relevant quantities, including the solar zenith angle and the flight altitude are adjusted according to the observations. Also, LibRadtran incorporates several model atmospheres for a range of latitudes and seasonal conditions, that include temperature and water vapor profiles among other quantities (see Emde et al. (2016)). Of these, a model atmosphere is chosen that resembles the conditions of the measurement.

The unknown quantity in the simulation is the IWV of the overlying atmospheric column above the altitude at which the measurement is made. The IWV in the model is adjusted so that the simulated irradiance $F_{\text{sim},\lambda}$ results in the same transmissivity in the water vapor absorption band at $\lambda_{\text{wv}_1}$ or $\lambda_{\text{wv}_2}$ as the measured irradiance $F_{\text{meas},\lambda}$. When adjusting the model IWV, the water vapor profile above the aircraft is scaled to a new IWV value while the behavior of its vertical distribution is left as it is defined by the model atmosphere. Before adjusting the IWV, the scaling procedure mentioned in Section 2.1 is realized by scaling the $F_{\text{meas},\lambda}$ to $F_{\text{sim},\lambda}$. This is done at the scaling wavelength nearest the concerning water vapor band being used for the retrieval. The difference between both spectra at the scaling wavelength, $F_{\text{sim}}(\lambda_{\text{sc}})$ and $F_{\text{sim}}(\lambda_{\text{sc}})$, is added uniformly to the measured irradiance. Due to the scaling, the quantity that is directly the input for the retrieval is the relative irradiance, i.e. the retrieval compares the difference in the simulated irradiance between the absorption band and the scaling wavelength with the difference in the measured irradiance between both wavelengths. This difference in irradiances can also be seen as the absorptance of the water vapor band, with units W m$^{-2}$, because the scaling wavelength is virtually free from extinction (see Figure 2.4).

The IWV is then retrieved by iteratively adjusting the input IWV of the model. With the measured irradiance $F_{\text{meas}}(\lambda_{\text{wv}})$ and the simulated irradiance $F_{\text{sim}}(\lambda_{\text{wv}}, IWV_n)$ the iteration is performed by:

$$IWV_{n+1} = \frac{F_{\text{sim}}(\lambda_{\text{wl}}, IWV_n)}{F_{\text{meas}}(\lambda_{\text{wl}})} \cdot IWV_n \quad (2.5)$$

After a sufficient amount of iterations the iteration factor becomes close to one and the IWV converges. The iteration is stopped when the difference between the measured and the simulated irradiance at $\lambda_{\text{wv}_1}$ or $\lambda_{\text{wv}_2}$ is below 0.001% of $F_{\text{meas}}(\lambda_{\text{wv}})$ at the concerning wavelength.

Each iteration step requires a costly simulation of the spectral radiative transfer. Therefore, it is important to keep the number of iteration steps until convergence low, especially when retrieving the water vapor for a longer flight interval.
vergence of the iteration is accelerated by taking the iteration factor to the power of an exponent that is a whole multiple of 2, e.g. 4, 8, or 16. The exponent is dynamically divided in half if the convergence would otherwise not happen stability.

2.4 Sensitivity study

The strength of the water vapor absorption band can decrease significantly for increasing flight altitude and decreasing water vapor content above the aircraft, to below 1% as was seen in Figure 2.6. Therefore, the accuracy of the irradiance measurements needs to be sufficiently high in order to allow a reliable retrieval of IWV. Figure 2.7 shows the simulated spectral transmissivity at 10 km and 15 km in the model US standard atmosphere. The tropopause in this model atmosphere

![Figure 2.7: Spectral transmissivity (black line) of model US-standard atmosphere at 10 km (a) and 15 km (b). Red lines represent a hypothetical measurement uncertainty of ±2%.](image-url)
is located at 11 km. The uncertainty of the measured irradiance is estimated with ±2 %. The uncertainty is considered by adding or subtracting ±2 % of the irradiance to the measured value (as represented by the black plots) and then applying the IWV retrieval to these modified irradiance spectra to determine the uncertainty range of IWV. The boundaries of this uncertainty range then produce new irradiance spectra, that are displayed as the red plots, thus representing the resulting uncertainty of the IWV retrieval. There is a large difference in the strength of the water vapor absorption bands between both altitudes. At 10 km the IWV is 0.031 kg m$^{-2}$.

Figure 2.8: IWV as a function of the transmissivity of the atmospheric column from 10 km to the TOA according to simulations (solid lines). Dashed lines illustrate uncertainty of IWV as a result of the +/−2 % uncertainty of the irradiance.
is about as large as the actual change of transmissivity corresponding to the variability in IWV. As the transmissivity is generally high for these low values of IWV even an uncertainty of 2% for the absolute irradiance causes a large uncertainty in transmissivity. The resulting uncertainty of IWV is between ±50% and ±100%.

The absolute and the relative uncertainty of the IWV retrieval both vary depending on the actual amount of IWV. The absolute uncertainty increases and the relative uncertainty decreases the higher the actual IWV is. For the simulations shown in Figure 2.7 an uncertainty of the IWV of +0.008/-0.011 kg m\(^{-2}\) at 10 km and +0.014/-0.004 kg m\(^{-2}\) on the upper bound at 15 km is obtained assuming a measurement uncertainty of 2%.

When performing calculations in this way and assuming a 2% uncertainty in the irradiance measurement it can be found that, in order to obtain an IWV uncertainty of at most 5% one needs an IWV of at least around 1 kg m\(^{-2}\). For comparison, this is the IWV one finds in the US standard atmosphere by Anderson et al. (1986) at an altitude of approximately 6 km.
3 Measurement with Research Aircraft

3.1 HALO research aircraft

The High Altitude and Long Range Research Aircraft (HALO) is a modified business jet that has operated as a German research aircraft since 2009. An image of the aircraft is displayed in Figure 3.1. HALO is able to carry a large payload and it is capable of long flight ranges and durations, and high flight altitudes of up to 15 km. Depending on the latitude HALO can thus reach into the stratosphere and is well suited for studying the UTLS.

HALO is permanently equipped with the Basis HALO Measurement and Sensor System (BAHAMAS) for in-situ measurements of basic meteorological quantities and data concerning the geographical location, altitude and attitude of the aircraft. It also includes an instrument that measures the in-situ WVMR in the range 20-60,000 ppmv with an uncertainty of less than 10 % (Wendisch et al., 2016).

So far, measurements of the water vapor content in the overlying atmospheric column have not been available from HALO. Retrieving the IWV from air-borne measurements of the downwelling solar irradiance would enable this.

3.2 Irradiance measurements

The Spectral Modular Airborne Measurement System (SMART)-Albedometer installed on HALO measures the spectral upward and downward solar irradiance ($F_{\lambda}^\uparrow$ and $F_{\lambda}^\downarrow$) with two optical inlets. These inlets are mounted to the upper and lower fuselage. A third inlet in the lower fuselage with a 2° field of view measures the upward solar radiance ($I_{\lambda}^\uparrow$). Each of the optical inlets are connected with two grating photodiode array spectrometers by optical fibers. After entering into the spectrometer through a slit, the incoming radiation is spectrally dispersed by a reflective grating and is detected by an array of photo-diodes. The diodes are ar-
ranged with a certain spacing to one another, so each diode measures a pixel in the spectrum according to its location with respect to the dispersed signal. For a single measurement of the spectrum the diodes count photons over an integration time of 0.5 s. Together, both spectrometers cover a large portion of the solar spectrum, spanning the spectral wavelength range of 300–2000 nm. The first spectrometer, for the visible range, covers 300–1000 nm with 1024 pixels. The second spectrometer, which is assigned to the NIR, covers 950–2200 nm with 256 pixels. A more detailed description of SMART can by found e.g. in Bierwirth et al. (2009) and in Ehrlich et al. (2008).

Another key component of the SMART-Albedometer is an active horizontal stabilization system, which reduces the system’s measurement uncertainty. For proper measurements the sensors must be aligned with respect to the horizontal plane, and even small deviations from a horizontal alignment can cause significant measurement errors. Misalignment easily results during flight, from the movements of the aircraft. Even during horizontal flight it is impossible for the airplane to maintain a perfectly stable horizontal position. With a firm installation of the sensor to the aircraft fuselage an accuracy of ±5 % or better would not be possible (Wendisch et al., 2001). In order to solve this problem Wendisch et al. (2001) developed an active stabilization system, which maintains a horizontal alignment of the optical inlets during flight with respect to the earth-fixed coordinate system. The inertial navigation system (INS) determines the aircraft attitude and position. The measured changes in attitude are then instantaneously compensated by an active horizontal adjustment system. With the active stabilization, an accuracy of the sensor’s horizontal alignment of better than ±0.2° for pitch and roll angles of up to ±6° is obtained. For the measured absolute irradiance the stabilization system thereby ensures an accuracy of better than ±1 % (Wendisch et al., 2001).

Several components go into the total uncertainty of the measurement of irradiance. The photodiodes in the spectrometer give off electronic noise by producing a signal even when there is no incoming radiation. This signal is temperature dependant. It is known as the “dark current”. Thus, the measured signal consists of the sum of the actually incident photons and the dark current. During operation the dark current is measured in regular, close intervals in dark measurements, whereby a shutter darkens the spectrometer for a few measurements. The measurement is then corrected for the registered dark current. The instrument undergoes an absolute calibration in the laboratory. Here it is calibrated spectrally, i.e. with respect to the pixel-wavelength assignment, with spectral lamps that have well defined, known emission lines. The absolute calibration also includes a calibration for absolute irradiance using a certificated 1000-W halogen lamp. The spectrometer also undergoes a transfer calibration when it is moved into the
field. During measurement campaigns the temporal stability of the calibration is verified by using secondary calibration standards. Brückner et al. (2014) estimate the total uncertainty from the uncertainty of the spectrometer signal, the absolute calibration, and the transfer calibration, to be 8.3% and 9.4% in the NIR. The largest fraction of the total uncertainty is on account of the total calibration. As the IWV retrieval uses the ratio of irradiance at two different wavelengths, the uncertainty of the spectrometer and transfer calibration can be ignored and only the uncertainty of the spectrometer signal remains relevant. This reduces the uncertainty in the NIR to 1.8% to 2.2% (Brückner et al., 2014).

Together, the spectrometer and the horizontal stabilization make up a total uncertainty of 2.8–3.2%.

### 3.3 Measurement campaigns

Two measurement campaigns that HALO performed in 2016 were the Next-generation Aircraft Remote-sensing for VALidation studies (NARVAL-II) and the North Atlantic Waveguide and Downstream impact EXperiment (NAWDEX). NARVAL-II took place from 20 June to 31 August and was located near Barbados. NAWDEX took place shortly afterwards from 12 September to 16 October. This campaign was focused on the North Atlantic region and was based on Iceland. The flight tracks of both campaigns are displayed in Figure 3.2. The general cruise altitude during both campaigns was above 8 km and up to approximately 14-15 km. With both of these campaigns, measurements are available from the sub-tropics and midlatitude region. Depending on the geographical location, the maximum flight altitude was in the upper troposphere or lower stratosphere.

![Flight tracks of the NARVAL-II campaign near Barbados (left) and the NAWDEX campaign over the north Atlantic (right)](Provided by German Aerospace Center.)
4 Realisation of Water Vapor Retrieval

The first step of the retrieval algorithm filters data when clouds above the aircraft affected the measurements. Therefore, the time series of downward irradiance at a visible wavelength was analyzed. Periods in which the downward irradiance was significantly reduced or showed high frequency noise were identified as being affected by overlying clouds. Additionally, the spectral pattern of the irradiance was used to identify clouds. The decline of the irradiance with wavelength is less steep under cloud cover than it is for a clear sky. If the transmissivity $T_\lambda = F_{\text{meas,}\lambda}/F_{\text{TOA,}\lambda}$ is larger at 2002 nm than at 1750 nm the measurement is excluded.

The spectral irradiance was smoothed with a moving average over three pixels, whereby each point $F_{\text{meas}}(i)$ in the spectrum is replaced by the average of the original value and the values of the neighboring point left and right, $F_{\text{meas}}(i - 1)$ and $F_{\text{meas}}(i + 1)$. This smoothed some of the high frequency structure in the signal caused by electronic noise or differences in sensitivity between neighboring pixels. Performing a moving average may decrease the uncertainty from pixel registration (Kaufman and Gao, 1992).

The iteration is stopped when the simulated and measured water vapor absorption band agree to better than 0.001% in relation to the measured irradiance in the absorption band. The possible gap between the retrieved and actual value of IWV becomes so small with this threshold it becomes negligible in comparison to the spectrometer uncertainty.

4.1 Spectral resolution

A technical aspect is introduced by the finite resolution of the spectrometer measurements. Each of the spectrometer’s photo diodes is centered at a specific wavelength in the spectrum of the dispersed radiation. To begin with, there is a degree of uncertainty regarding the pixel-wavelength alignment. But also, even when a pixel is clearly assigned to a specific wavelength, this pixel does not only receive photons from the infinitesimal spectral range of this wavelength, but also from wavelengths nearby. The photon registration of the pixel has a maximum at the central wavelength and decreases away from this wavelength symmetrically with
the shape of a gauss curve.

In order to apply simulations to the measurements, the simulations need to be convoluted with a spectral slit function. This slit function is applied to each point of the simulated irradiance spectrum. It is a Gaussian function with a FWHM equal to that of the measurement’s pixel. The FWHM varies throughout the spectrum. Choosing a suitable value for the FWHM that goes into the retrieval becomes a trade-off between reproducing the irradiance at the center of an absorption band the most accurately and reproducing irradiance throughout the whole absorption band the most accurately.

The retrieval was performed for the scan using a range of values for the FWHM, resulting in several simulated irradiance spectra from different retrieved values of IWV. The simulated spectra were then compared with the measured scan. An ideal value was determined from a combination of reducing the standard deviation in the difference between measured and simulated irradiance in the absorption band on the one hand, and on the other hand a simple estimation by eyesight. The specific values of FWHM for the absorption bands will be named below.

4.2 Retrieval Method

4.2.1 Retrieval using center wavelength of absorption band

For the retrieval of IWV using the center wavelengths of the absorption bands $\lambda_{wv1}=1363\,\text{nm}$ and $\lambda_{wv2}=1873\,\text{nm}$, where maximum absorption occurs for the respective band, the finite resolution of the spectrometer becomes relevant. It must be taken into account that the spectrometer has no pixel that is centered at the precise location of the absorption band center and instead measures at wavelengths that are close to the center. When looking at the 1.4 $\mu$m absorption band (e. g. in Figure 2.4) one notices a steep drop in absorption when moving along the spectrum away from the point of maximum absorption. As a result, varying the wavelength in the retrieval slightly can result in differing values of the detected IWV.

Figure 4.1 shows the measured and the simulated transmissivity. The simulated transmissivity is retrieved using the 1.4 $\mu$m absorption band for the top plot and the 1.9 $\mu$m absorption band for the bottom plot. In one case the retrieval wavelength is the band center given by the simulation and in two other cases the retrieval wavelength was the two pixels of the measurement that were closest to this band center. For the 1.4 $\mu$m band the proposed wavelength for performing the retrieval is 1365 nm. The retrieved IWV is 0.084 kg m$^{-2}$. The absolute standard deviation in retrieved IWV between this and the two adjacent wavelengths is $5.5 \cdot 10^{-3}$ kg m$^{-2}$.
or approximately 7% of the retrieved IWV. In the case of the 1.9 μm the proposed wavelength for retrieval is 1873 nm. The retrieved IWV is 0.057 kg m$^{-2}$. Here, the absolute standard deviation between the three adjacent retrieval wavelengths is $3 \cdot 10^{-3}$ kg m$^{-2}$ or approximately 5% of the retrieved IWV.

This retrieval method is sensitive to the FWHM of the slit function. The ideal FWHM was found to be 18.0 nm for the 1.4 μm and 10.8 nm for the 1.9 μm band. A measure for this sensitivity is to observe the degree to which the retrieved IWV changes after changing the FWHM by 1 nm. For the retrieval at 1365 nm the change in retrieved IWV is 0.003 kg m$^{-2}$ nm$^{-1}$ or approximately 3% of the retrieved IWV and at 1873 nm this is 0.0044 kg m$^{-2}$ nm$^{-1}$ or 6%.

![Graph](image)

Figure 4.1: Measurement from NAWDEX at 8 km (red plot) and simulation with retrieved IWV (blue plot). Top: retrieval for 1.4 μm band at 1363, 1365 and 1368 nm. Bottom: retrieval for 1.9 μm band at 1871, 1873 and 1877 nm.
4.2.2 Retrieval using integrated absorption band

To become less sensitive to the exact spectral location of measured and simulated water vapor absorption bands, the absorptivity of the entire absorption band was integrated for both measurements and simulations. The integrated signal between 1300–1500 nm for the 1.4 \( \mu \text{m} \) and between 1750–1950 nm for the 1.9 \( \mu \text{m} \) band was used in the retrieval to estimate the IWV. This is approximately the entire span of the respective band.

The FWHM of the slit function that was applied to the simulation was varied for these two retrieval methods as well and the ideal values are found to be 15.6 nm for the 1.4 \( \mu \text{m} \) band and 10.8 nm for the 1.9 \( \mu \text{m} \) band. Comparing both retrieval methods the integral method is found to be less sensitive to the assumed slit function than the retrieval based on the irradiance at the center wavelength only. The sensitivity of the retrieved IWV for a variation of the FWHM by 1 nm is 0.00019 kg m\(^{-2}\) nm\(^{-1}\) or 0.2 % for the 1.4 \( \mu \text{m} \) band and 0.00037 kg m\(^{-2}\) nm\(^{-1}\) or 0.5 % for the 1.9 \( \mu \text{m} \) band.

Figure 4.2 shows an example of transmissivity measured at 8 km altitude and the simulations of the best fit obtained by the retrieval. The top plot is for the retrieval with the 1.9 \( \mu \text{m} \) band with a retrieved IWV of 0.073 kg m\(^{-2}\). The bottom is for the retrieval with the 1.4 \( \mu \text{m} \) band and a retrieved IWV of 0.089 kg m\(^{-2}\).

4.2.3 Comparison of different retrieval approaches

The retrieved values for IWV tend to be lower when retrieving with the 1900 nm band than with the 1400 nm band, both for the integral and for the wavelength method. On average the disagreement between both retrieval bands is smaller for the wavelength method than the integral method, with an average deviation between both wavelengths of 0.007 mm versus 0.014 mm throughout the FWHM range 10.8–15.6 nm.

With the slit functions determined above, the agreement becomes twice as good for the integral method compared to the wavelength method. This makes sense because the FHWM has a larger influence on the wavelength method than on the integral method. The disagreement for the central wavelength makes up 36 % of the mean IWV from both bands versus 19 % for the integral method.

It can be expected that the 1.4 \( \mu \text{m} \) band has a better signal-to-noise ratio than the 1.9 \( \mu \text{m} \) band, because the solar irradiance during a cloudless sky is stronger at a shorter wavelength within the NIR. Therefore, the retrieval method of integration of the 1.4 \( \mu \text{m} \) band between 1.3 \( \mu \text{m} \) and 1.5 \( \mu \text{m} \) is used for the IWV retrieval in the following.
Figure 4.2: Measurement from NAWDEX at 8 km (red plot) and simulation with retrieved IWV using integral method of retrieval (black plot). Top: retrieval for 1.4 $\mu$m band and FWHM of 15.6 nm. Bottom: retrieval for 1.9 $\mu$m and FWHM of 10.8 nm.

4.3 Uncertainty Estimation

The uncertainty of the IWV retrieval results from several factors. One of these is the uncertainty of the temperature and water vapor profile throughout the atmosphere above the aircraft. Several processes take place in the atmosphere that lead to a decrease in the absorption in the center wavelength of an absorption band and an increase in the absorption in the wings of the absorption band, i.e. at wavelengths near the center wavelength. The result is a broadening and flattening of the absorption band. The processes behind such broadening are doppler broadening and pressure broadening. Both processes depend on local conditions in the atmosphere, including the temperature. Therefore, a lack of knowledge of the temperature profile or the vertical distribution of water vapor both contribute to
uncertainties in the interpretation of a transmissivity spectrum. Kaufman and Gao (1992) quantify this effect for the Earth Observing System (EOS) measurements and conclude that the temperature and water vapor profile can cause a significant error of 2% to 5% for retrieved IWV of the entire atmospheric column. They also found that this error can be reduced to \( \sim 1\% \) by assuming a realistic temperature profile that fits the conditions of the measurement, or by using a temperature profile derived simultaneously by the EOS. These estimates were calculated for solar radiation that has passed through the entire atmosphere twice. For retrieval analyzing the downward solar radiation at the UTLS, the uncertainties will decrease.

To summarize the factors of the irradiance measurement that contribute to uncertainty of the IWV retrieval, these are the uncertainty of the horizontal alignment of the optical inlets during flight, as well as several aspects in the uncertainty from the spectrometer, which can be summarized as noise. Noise sources, as stated by Platt and Stutz (2008) are photon statistics, electronic detector noise in the instrument and further, unexplained, random spectral structures in the signal. A reason for the latter factor can for example be pixel-to-pixel variations in sensitivity.

Kindel et al. (2015) achieve a very high accuracy in their measurements of the solar irradiance by averaging their spectra over 5 to 10 minutes, which comprised 300-600 spectra. In doing so they average out signal noise, as well as effects from attitude changes during flight. The uncertainty of the spectrometer after averaging is stated at 0.1%.

The red plot in Figure 4.3 shows an example of the transmittance spectrum from a single measurement taken during a cruise episode at 12.3 km during a NAWDEX flight. In this situation there is a low IWV and water vapor absorption is not clearly distinguishable above the signal noise. After averaging irradiance spectra over five minutes or approximately 600 spectra around the same measurement the signal noise remains apparently as high as the non-averaged spectrum and shows a similar structure. There must be a noise component that is due to spectral structures of the instrument.

The signal noise can be considered quantitatively by observing the dispersion of the signal around the stochastic expectation. The expectation is the theoretical irradiance as given by a simulation. Water vapor is an unknown variable for the radiative transfer, but can be avoided by considering a spectral range in which water vapor exerts no influence, such as the range 1530 nm to 1700 nm. Within this range the standard deviation of the spread between measurement and simulation is calculated to quantify the noise.

Despite the averaged spectra having a larger slant towards large wavelengths than the single spectrum, the dispersion of the two is about the same. In both cases the standard deviation is at about \( \sim 0.002 \text{ W m}^{-2} \) or 2% relative to the mean
of the simulated irradiance within the stated spectral range. The running average over three pixels reduces the relative standard deviation to 1.2%. A running average over five pixels further reduces this to 0.0012 W m\(^{-2}\) or slightly over 1%, but using a generous running average may also eliminate information out of the signal. These dispersions are in good agreement with the spectrometer uncertainty that is adopted from Brückner et al. (2014).

When applying this dispersion to the entire spectrum, error bars such as those in Figure 4.4 are obtained. The error bars in the plot denote the uncertainty resulting solely from the investigated noise. The figure shows two cases at different altitudes, namely at 7 km and at 12 km. Both spectra are now smoothed over three pixels. The absolute standard deviation of the spectral measurement is approximately 0.001 W m\(^{-2}\) for both cases. This accounts to about 1% of the irradiance. Depending on the altitude of the measurement and the water vapor content in the atmospheric column, this noise uncertainty may be more or less strong. In the case of the low measurement, where there is a strong water vapor signal, noise uncertainty results in a relative uncertainty in the IWV detection of only 10%. For the high measurement this uncertainty is in the order of 100%. This confirms the qualitative insight of the plot, which is that at a certain altitude the expected water vapor content becomes so low, that the resulting absorption signal becomes too weak to reliably retrieve the IWV.

Therefore, the uncertainty of the irradiance as explained in Section 3.2 remains

![Figure 4.3: Spectral transmissivity measured at 12.3 km during a NAWDEX flight (red) and the spectral transmissivity averaged over 600 spectra (black). The blue plot is the result of a moving average over three pixels of the red plot.](image)
accurate. The stabilization uncertainty may actually be smaller for the same reason that the spectrometer uncertainty is only 2\%, namely that we are looking at relative irradiance instead of the absolute irradiance. A total uncertainty of ±3\% appears to be a reasonable upper limit for the irradiance measurement and will be inserted into the discussion regarding the uncertainty of the retrieval.

4.3.1 Total uncertainty of the IWV retrieval

The uncertainty of IWV obtained by the retrieval is estimated for a ±3\% uncertainty of the measured transmissivity. For each measurement an uncertainty of IWV is estimated by propagating the uncertainty of the measurements into the retrieval algorithm. The calculated retrieval uncertainty results in a range of IWV

![Figure 4.4: Spectral transmissivity measured at 12 km (left) and 7 km (right) and retrieved transmissivity with noise. Vertical line denotes scaling wavelength.](image)

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which is not symmetrically with respect to the main retrieval result, which is in line with the physics of Lambert-Beer’s law. For the measurement at 7 km from the bottom panel in Figure 4.4 the calculated uncertainty is \( +0.059/-0.069 \text{ kg m}^{-2} \) or \( +32/-38 \% \). This measurement was geographically in the North Atlantic and the flight altitude was thus roughly at the tropopause. At 8 km during the same flight the uncertainties grow to \( +0.049/-0.038 \text{ kg m}^{-2} \) or \( +188/-92 \% \). Even a relatively small uncertainty in the irradiance measurement lead to a large uncertainty in the retrieved IWV in the UTLS, due to the scarce water vapor content encountered in the overlying atmosphere.
5 Integrated Water Vapor Retrieved from
SMART-Measurements

As an example of a longer measurement period from the NAWDEX campaign, the second flight of the campaign, which took place on 21 September, is presented here. The flight took place in the vicinity of Iceland, staying within 350 km of the island. The latitude ranged from 62° N to 68° N. A timeseries of the flight altitude as well as the measured downward irradiance at the visible wavelength 640 nm is shown in Figure 5.1. The flight consisted of a steady ascent after take-off shortly before 14:00 UTC (and 14:00 local time) and thereafter a period of cruise at ∼12,400 m that lasted for 4 1/2 hours until 19:00 UTC. From the noisy behavior in the measured 640 nm signal at the beginning of the ascent between the ground and 6 km one can see the influence of overlying cloud cover. Otherwise, the flight was apparently free from overlying clouds, which makes it suitable for the retrieval of IWV. The further irregularities in the 640 nm signal that occur sporadically during the cruise arise from deviations in the alignment of the SMART optical inlet during changes in flight direction.

The presented example flight from NARVAL-II is the fifth campaign flight, which took place on 19 August 2016. HALO flew between 13° N and 19° N. A time series of the flight altitude as well as the measured downward irradiance at the visible

![Figure 5.1](image)

Figure 5.1: Timeseries of the flight altitude (yellow) and the measured (black) and simulated (red) downward irradiance at 640 nm in the visible spectrum.
wavelength 640 nm is shown in Figure 5.2. After take-off at 8:30 local time the aircraft ascended to approximately 9,700 m and cruised for six hours. Then it ascended farther and cruised at 14,400 m for an hour from 15:10 to 16:10 local time.

In the first following section, the focus will be on periods of cruise flight. The retrieval for the cruise periods was performed with a temporal resolution of 1 minute. Thereafter, the focus will be on periods of ascent, which will be used to derive vertical profiles of \( w \). The IWV was retrieved with a resolution of 10 seconds for the ascent periods.

### 5.1 Horizontal variability

#### 5.1.1 NAWDEX

The cruise altitude during NAWDEX of 12,400 m is slightly higher than the altitude one would expect for the local tropopause, so the overlying atmospheric column is characterized by dry stratospheric conditions. Using the predefined model-atmosphere from libRadtran for subarctic summer leads to an IWV of 6 g m\(^{-2}\) at 12.3 km. The average value of the IWV retrieved for this flight period is 5.6 g m\(^{-2}\). The time series of the retrieved IWV is shown in Figure 5.3. As can be seen quantitatively in Figure 5.3, there is very large variability in the time series of the IWV. The relative standard deviation is 148%. Many retrieved values result in practically zero IWV, with values that are ten orders of magnitude below the average, while many outliers are several orders of magnitude larger than the average. The uncertainty of the retrieval from \( \pm 3\% \) spectrometer uncertainty under these conditions is -100%/+200%. It is not possible to make reliable statements
Figure 5.3: Time series of retrieved integrated water vapor (IWV) at an average flight altitude of 12400 m for 21 September 2016. Uncertainty is indicated by red error bars.

on the IWV for this flight period, because the water vapor signal becomes too weak to be distinguishable in the spectral irradiance measurement from the spectrometer. Variability in retrieved IWV is completely dominated by differences between measurements due to noise. This example illustrates that water vapor content is too low when flying above the tropopause to retrieve IWV with the method and the irradiance measurements applied in this work.

### 5.1.2 NARVAL-II

In this section the consideration will move from the stratosphere to the troposphere, where IWV is larger. NARVAL-II took place in the tropical region, Barbados is located at $13\degree$ N. Because the tropopause is higher at lower latitudes the flight altitudes performed during the campaign are generally farther below the tropopause than during NAWDEX.

The time series of the retrieved IWV for the first cruise period, at 9,700 m altitude, is displayed in Figure 5.4. This cruise period was well below the local tropopause. The model atmosphere for the tropics has an IWV of $0.12 \text{ kg m}^{-2}$ at this altitude. The retrieved IWV is slightly higher, with an average value over the measurement period of $0.17 \text{ kg m}^{-2}$, but is in the same order of magnitude. The retrieved IWV shows significant variation, with a short period of decrease at the beginning and thereafter strong variation around a more constant trend. When excluding the outliers, the time series has a variability with a relative standard
deviation of 32%. Throughout the time series the flight altitude \( z \) varies by 27 m between the highest and the lowest altitude. The correlation coefficient between the altitude and IWV is -0.11, which is negligible. Thus it can be assumed that the variability in flight altitude was not a significant factor in the IWV variability. The time series shows evidence of horizontal heterogeneity of the vertical water vapor profile, or at least of the IWV, above the aircraft along the flight path. With an average flight speed of 220 m s\(^{-1}\) the distance that HALO travels in an hour is approximately 800 km. The retrieval uncertainty is a limitation when interpreting the observed variability, because the error bars mostly cover the range of the IWV variability. The uncertainty of the retrieval is +40/-34 %.

The time series of the retrieved IWV for the second cruise period, at 14,400 m altitude, is shown in Figure 5.5. This period can be assumed to be slightly below the tropopause, as the general height of the tropopause in the tropics is placed at 18 km. However, the tropical tropopause region is generally dryer than the extratropical tropopause. The model atmosphere for the tropics gives an IWV of 0.004 kg m\(^{-2}\) for 14.4 km. The average retrieved IWV was 0.005 kg m\(^{-2}\). The variability is less than the NAWDEX time series. The relative standard deviation is 45%. This is not that much less than the lower cruise. The lower cruise was longer in duration however, and when calculating the standard deviation of a similarly long period the relative standard deviation reduces from 32% to 16% for the lower cruise. This may be evidence that other factors than retrieval uncertainty played a role in the variability of the time series of the lower cruise. As a whole,
one encounters the same problem as for the NAWDEX cruise, which is that the water vapor signal is too weak for a reliable retrieval. The upper bound of the uncertainty calculated from ±3% measurement uncertainty for this flight period, as can be seen in Figure 5.5, is in the order of +1000%.

5.2 Vertical profile

From flight periods of ascent or descent it is possible to retrieve the IWV within a certain vertical section of the atmosphere, as was done by Kindel et al. (2015). In a similar manner, a vertical profile of water vapor concentration can be obtained by retrieving the IWV within many small segments of the total vertical section. Performing either of the above tasks assumes that there is negligible horizontal variability in water vapor, because one must assume that a difference in the IWV detected at two different altitudes results solely from the change in altitude and not from a horizontal change in the IWV over the distance that the aircraft required to change its altitude.

5.2.1 NAWDEX

After take-off for the flight on 21 September during NAWDEX the ascent of the aircraft to 12.3 km took 24 minutes. On average, the altitude difference between two retrieval points was 77 m. The retrieval was not performed starting at ground level directly at take-off, but at an altitude of 6 km. This way measurements were
avoided that were inhibited by cloud cover (see Figure 5.1). The resulting profile of the absolute humidity $\rho_{wv}$ is displayed in the bottom panel of Figure 5.6. Included is also the profile from in-situ measurements of water vapor from the BAHAMAS system and measurements from a radio sonde launched 2 hours before the take-off of HALO. In the top panel the time series of retrieved IWV, from which the profile was created, and of the flight altitude are shown.

The top panel shows a steady increase in the altitude. Accordingly, the IWV

![Figure 5.6: Ascent after take-off on 21 September on second flight day of NAWDEX from ground level to 12.3 km. Top panel: times series of flight altitude (blue) and of the retrieved IWV (black). Bottom panel: vertical profile of WVMR from the IWV retrieved from SMART measurements (black) and the vertical profile of WVMR from BAHAMAS (red) and from radiosonde (blue). Uncertainty of the SMART profile exceeds the range of the plot. The value for the uncertainty is $\pm 1.5 \text{ g m}^{-3}$.](image_url)
generally drops throughout the ascent. The BAHAMAS profile and the radio sonde measurement show an apparent gap. For the retrieved profile one can perceive a behavior that follows that of the BAHAMAS profile for the most part, but with significantly more variation. This is especially the case at the bottom of the profile. There is also a large occurrence of negative values, which is physically impossible. This happens when a retrieved value for the IWV is lower than the IWV retrieved for the previous time step, at a lower altitude. From observing the variability of the IWV in Figure 5.4 horizontal inhomogeneities are possibly a cause for the vibrations in the time series of the retrieved IWV. However, uncertainties in the IWV retrieval are likely a larger factor.

5.2.2 NARVAL-II

After take-off for the flight on 19 August during NARVAL-II the ascent of the aircraft to 9.7 km took 16 minutes. The average altitude difference between two points of retrieval was 130 m. The time series of the flight altitude and retrieved IWV, as well as the resulting profile of $w$, a BAHAMAS profile, and a radio sonde measurement are displayed in Figure 5.7. The radio sonde was launched at 12 UTC, a half hour before take-off. Many of the findings for the profile from the NAWDEX flight hold for this profile as well. There is an evident structure in the retrieved profile, with a decrease towards larger altitudes, that is similar to the BAHAMAS profile. However the retrieved profile shows large vibration and many negative values.

From the difference in the IWV from the SMART measurements at 4 km and 9.7 km, the IWV between both altitudes is $2.43 \pm 0.32/-0.36 \text{ kg m}^{-2}$. In comparison, the BAHAMAS measurements result in $3.21 \text{ kg m}^{-2}$ and the radio sonde results in $2.74 \text{ kg m}^{-2}$ for the same layer.

In the higher section between 9.7 and 14.4 km $w$ decreases much less than it did throughout the layer below, as is confirmed by the BAHAMAS and radio sonde data (Figure 5.8). The vertical resolution resulting from the 10 second resolution of the retrieval was approximately 90 m. In the SMART profile the vertical development of the absolute humidity $\rho_w$ is barely distinguishable over the noise. Although the top of the profile is likely below the tropopause, as with the profile from NAWDEX, the IWV here has dropped to below $0.01 \text{ kg m}^{-2}$. 
Figure 5.7: Ascent after take-off on 19 August on fifth flight day of NARVAL-II from ground level to 9.7 km. Top panel: times series of flight altitude (blue) and of the retrieved IWV (black). Bottom panel: vertical profile of WVMR from the IWV retrieved from SMART measurements (black) and the vertical profile of WVMR from BAHAMAS (red) and from radiosonde (blue). The thin dashed line denotes the uncertainty of the SMART profile. The value for the uncertainty is $\pm 1.5 \, \text{g m}^{-3}$.
Figure 5.8: Ascent after take-off on 19 August on fifth flight day of NARVAL-II after cruise at 9.7 km up to 14.4 km. Top panel: times series of flight altitude (blue) and of the retrieved IWV (black). Bottom panel: vertical profile of absolute humidity from the IWV retrieved from SMART measurements (black) and the vertical profile of absolute humidity from BAHAMAS (red) and from radiosonde (blue). Uncertainty of the SMART profile exceeds the range of the plot. The value for the uncertainty is $\pm 1.4 \text{ g m}^{-3}$. 
6 Summary and Conclusion

Airborne spectral solar radiation measurements are used to investigate the fine spatial structure of water vapor distribution in the upper troposphere and lower stratosphere (UTLS). Measuring water vapor in the UTLS is a difficult task, and requires high accuracy measurements due to the conditions of low pressure and temperature and the exceedingly low water vapor mixing ratio. This work analyzes the potential of retrieving the integrated water vapor (IWV) at altitudes in the UTLS from measurements of the spectral solar irradiance using the principle of differential optical absorption spectroscopy. For this purpose, two water vapor absorption bands at 1.37 and 1.87 \( \mu \text{m} \) in the NIR were considered. An algorithm was developed for retrieving the IWV from measured spectral irradiance.

The irradiance in the respective absorption band was transformed into the difference between the irradiance at the center of the absorption band and the irradiance at a wavelength free of trace gas absorption. This absorptance is less sensitive for measurement uncertainties. The IWV was estimated by iteratively adjusting simulations by changing the IWV until the water vapor signal in the simulation and the measurement agreed.

A sensitivity study showed that, for the weaker water vapor concentrations in the UTLS, uncertainties in the radiation measurement lead to a large uncertainty in the retrieval. Estimates were deduced for the measurement certainty of the spectral irradiance required to achieve an acceptable uncertainty of the retrieved IWV. Measurements with a large signal-to-noise ratio are required to detect the weak water vapor signal found under the dry circumstanced in the UTLS. Assuming an uncertainty of \( \pm 2 \% \) for the irradiance measurements, the retrieval uncertainty exceeds 5 \% for the IWV below 1 kg m\(^{-2}\). In the US standard atmosphere this is value is encountered at about 6 km. For a better performance of the retrieval a much higher certainty of the spectrometer is necessary. With a \( \pm 0.1 \% \) uncertainty the altitude with \( \pm 5 \% \) uncertainty is raised to 10 km in the US standard atmosphere. This is still below the tropopause, which is located at 11 km in this model atmosphere.

The retrieval algorithm is applied to observations from one measurement flight from two HALO campaigns each, NAWDEX and NARVAL-II, providing measurements in different altitudes and atmospheric conditions. Various factors for
measurement uncertainty from the radiation measurement system SMART have been considered and a total uncertainty of ±3% was concluded. Particularly, the signal noise is a strongly restricting factor in gaining reliable values of IWV. Kindel et al. (2015) were able to assume a very high certainty of the spectrometer measurements used in their study. After averaging spectra over several minutes they achieve an accuracy of 0.1%. This work was not able to achieve similar results through temporal averaging of spectral measurements. Reasons for this may be differences in the deployed instrument. It was illustrated that for measurements at high altitudes, in the lower stratosphere or at the tropopause, the water vapor signal becomes indistinguishable over the spectral noise. Time series of the retrieved IWV at such high altitudes are dominated by measurement noise, as the noise leads to large variations in the retrieved IWV. The retrieval uncertainty in these cases exceeds 100%.

Measurements from NARVAL-II showed that the method can be used for flights farther below the tropopause, e.g. at ~10 km in the tropics. Thus, the uncertainty of the retrieval was in the range of ±30-40%. In comparison, this is more than the uncertainty the Boulder balloon measurements and the HALOE measurements achieve in the stratosphere, which is 10% and 14-24% respectively.

Also, the retrieval of vertical profiles of water vapor concentration is possible in principal from flight periods with ascent or descent. This was attempted for several flight periods from both flights. Apart from the the retrieved profiles being constrained with large uncertainty, beyond 100%, they show reasonable agreement with water vapor profiles from radio sonde and airborne in-situ measurements. The IWV retrieved for a layer between 4 km and 9.7 km from one ascent period during NARVAL-II resulted in a value that was close to the values obtained from radio sonde and in-situ measurements, namely 2.43 ±0.32/-0.36 kg m⁻² compared with 2.74 kg m⁻² from the radio sonde and 3.21 kg m⁻² from the in-situ measurements.

In summary, both for flight periods of cruise as well as ascent, the results for the IWV are strongly affected by noise from the spectrometer and have a high uncertainty. The examples investigated within this work show that a retrieval of IWV from airborne spectral solar radiation measurements is possible for IWV exceeding 0.1 kg m⁻². For lower IWV the measurement uncertainty of SMART is to high. Therefore, IWV estimates in the UTLS, where IWV ranges below 0.05 kg m⁻² are not possible with the current measurement setup. A significant improvement of the measurement uncertainties needs to be achieved in order to obtain IWV retrieval in these altitudes with only low amount of WV.


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Declaration of Authorship

I hereby certify that this thesis has been composed by me and is based on my own work, unless stated otherwise. All direct or indirect sources used are acknowledged as references.

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Peter Stammer