Differential absorption lidar for water vapor isotopologues in the 1.98-_µm spectral region: sensitivity analysis with respect to regional atmospheric variability

- Jonas Hamperl¹, Clément Capitaine², Jean-Baptiste Dherbecourt¹, Myriam Raybaut¹, Patrick Chazette³, Julien Totems³, Bruno Grouiez², Laurence Régalia², Rosa Santagata¹, Corinne Evesque⁴, Jean-Michel Melkonian¹, Antoine Godard¹, Andrew Seidl⁵, Harald Sodemann⁵, Cyrille Flamant⁶
- 10 ¹DPHY, ONERA, Université Paris Saclay, F-91123 Palaiseau, France
 - ²Groupe de Spectrométrie moléculaire et atmosphérique (GSMA) UMR 7331, URCA, France
 - ³Laboratoire des Sciences du Climat et de l'Environnement (LSCE), UMR 1572, CEA-CNRS-UVSQ, Gif-sur-Yvette, France ⁴Institut Pierre-Simon Laplace (IPSL), FR636, Guyancourt, France
 - ⁵Geophysical Institute, University of Bergen, and Bjerknes Centre for Climate Research, Bergen, Norway
- 15 ⁶Laboratoire Atmosphères Milieux et Observations Spatiales (LATMOS), UMR 8190, CNRS-SU-UVSQ, Paris, France

Correspondence to: Jonas Hamperl (jonas.hamperl@onera.fr)

Abstract. Laser active remote sensing of tropospheric water vapor is a promising technology to complement passive observational means in order to enhance our understanding of processes governing the global hydrological cycle. In such context, we investigate the potential of monitoring both water vapor H₂¹⁶O and its isotopologue HD¹⁶O using a differential absorption lidar (DIAL) allowing for ground-based remote measurements at high spatio-temporal resolution (150 m and 10 min) in the lower troposphere. This paper presents a sensitivity analysis and an error budget for a DIAL system under development which will operate in the two-micrometer spectral region. Using a performance simulator, the sensitivity of the DIAL-retrieved mixing ratios to instrument-specific and environmental parameters is investigated. This numerical study uses different atmospheric conditions ranging from tropical to polar latitudes with realistic aerosol loads. Our simulations show that the measurement of the main isotopologue $H_2^{16}O$ is possible over the first 1.5 km of atmosphere with a relative precision in the water vapor mixing ratio of <1% in a mid-latitude or tropical environment. For the measurement of HD¹⁶O mixing ratios under the same conditions, relative precision is shownfound to be of similar order, thus allowing slightly lower, but still sufficient for the retrieval of range-resolved isotopic ratios with precisions in δD of a few per mil. We also show that expected precisions vary by an order of magnitude between tropical and polar conditions, the latter giving rise to reduced precision poorer sensitivity due to low water vapor content and low aerosol load. Such values have been obtained for a commercial InGaAs PIN photodiode, as well as temporal and line-of-sight resolutions of 10 min and 150 m, respectively. Additionally, using vertical isotopologue profiles derived from a previous field campaign, precision estimates for the HD¹⁶O isotopic abundance are provided for that specific case.

35 1 Introduction

40

In many important aspects, climate and weather depend on the distribution of water vapor in the atmosphere. Water vapor leads to the largest climate change feedback, as it more than doubles the surface warming from atmospheric carbon dioxide (Stevens et al., 2009). Knowing exactly how water vapor is distributed in the vertical is of paramount importance for understanding the lower-tropospheric circulation, deep convection, the distribution of radiative heating, surface fluxes magnitude and patterns, among other processes. Conventional radio-sounding or passive remote sensors, such as microwave radiometers or infrared spectrometers, are well established tools used for water vapor profile retrieval in the atmosphere. However, apart from balloon-borne soundings, most of these instruments do not allow for determining how water vapor is distributed along the vertical in the 0–3 km above the surface which contains 80% of the water vapor amount of the atmosphere. Additionally, passive remote sensors will generally require ancillary measurements such as aerosols, temperature, or cloud heights to limit the errors on retrieved concentrations from radiance measurements. To complement these methods, active remote sensing techniques are expected to provide higher resolution measurement capabilities especially in the vertical direction where the different layers of the atmosphere are directly probed with a high power-laser-transmitter. Among these active remote sensing techniques, Raman lidar is a powerful way to probe the atmosphere as it can give access to several atmospheric state parameters within a single line of sight such as temperature, aerosols, and water vapor mixing ratio (WVMR) (Whiteman et al., 1992). Benefiting from widely commercially available high-energy visible or UV lasers, as well as highly sensitive detectors, they allow high accuracy, long range measurements despite the small Raman scattering cross-section. WVMR retrieval from Raman Lidar signal is however typically limited by parasitic daytime sky radiance and requires instrument constant and overlap function calibration (Whiteman et al., 1992; Wandiger and Raman, 2005). Conversely, the differential absorption lidar (DIAL) technique is in principle calibration free since the targeted molecule mixing ratio can be directly retrieved from the attenuation of the lidar signals at two different wavelengths, knowing the specific differential absorption cross-section of the targeted molecule (Bösenberg, 2005). However, this benefit must be balanced with higher instrumental constraints especially on the laser source which is required to provide high power as well as high frequency agility and stability at the same time. For water vapor this method has been successfully demonstrated essentially using pulsed laser sources emitting in the visible or near infrared (Bruneau et al., 2001; Wirth et al., 2009; Wagner and Plusquellic, 2018), and recent progress in the fabrication and integration of tapered semiconductor optical amplifiers has enabled the development of small-footprint field-deployable instrumentation (Spuler et al., 2015). The infrared region between 1.5 µm and 2.0 µm has also attracted interest for water vapor DIAL sounding, especially in the context of co-located methane and carbon dioxide monitoring (Wagner and Plusquellic, 2018, Cadiou et al., 2016). One of the potential benefits of co-located multiple species measurement would be to reduce the uncertainties related to the retrieval of dry-air volume mixing ratios for the greenhouse gas (GHG) of interest. This aspect has particularly been studied in the field of space-borne integrated path differential absorption (IPDA) lidar for carbon dioxide (CO₂) monitoring in the 2.05 µm region where water vapor absorption lines may affect the measurement (Refaat et al., 2015). One of the great potentials of these multiple-wavelengths and multiple-species approaches would be their adaptability to isotopologue measurements with the DIAL technique since isotopic ratio estimation is equivalent to multiple species measurement provided the targeted isotopologues display similarly suitable and well separated absorption lines in a sufficiently narrow spectral window.

70

75

Humidity observations alone are not sufficient for identifying the variety of processes accounting for the proportions and history of tropospheric air masses (Galewsky et al., 2016). Stable water isotopologues, mainly H₂¹⁶O, HD¹⁶O and H₂¹⁸O differ by their mass and molecular symmetry. As a result, during water phase transitions, they have slightly different behaviors. The heavier molecules prefer to stay in the liquid or solid phase while the lighter ones tend to evaporate more easily, or prefer to stay in the vapor phase. This unique characteristic makes water isotopologues the ideal tracers for processes in the global hydrological cycle. Water isotopologues are independent quantities depending on many climate factors, such as vapor source, atmospheric circulation, precipitation and droplet evaporation, and ambient temperature. So far, no lidar system has been investigated for the measurement of water vapor isotopologues other than H₂¹⁶O (hereafter referred to as H₂O). Here, in the framework of the Water Vapor Isotope Lidar (WaVIL) project (Wavil, 2021), we investigate the possibility of a transportable differential absorption lidar to measure the concentration of both water vapor H₂O and the isotopologue HD¹⁶O (hereafter referred to as HDO) at high spatio-temporal resolution in the lower troposphere (Hamperl et al., 2020). The proposed lidar will operate in the two-micrometer spectral region where water vapor isotopologues display close but distinct absorption lines. Such an innovative remote sensing instrument would allow for the first time the simultaneous monitoring of water vapor and HDO isotopic abundance profiles with a single setup, enabling the improvement of knowledge on the water cycle at scales relevant for meteorological and climate studies.

The purpose of this paper is to assess the expected performances of a DIAL instrument for probing of H_2O and HDO in the lower troposphere. In section 2, the choice of the sensing spectral range is substantiated, and the performance model is outlined. The approach for modelling transmitter, detection, and environmental parameters is detailed. The sensitivity analysis is based on representative average columns of arctic, mid-latitude, and tropic environments. The simulation results and an extensive error analysis are presented in section 3. To assess the random uncertainty in the retrieved isotopologue mixing ratio, major detection noise contributions are analyzed for a commercial InGaAs PIN and a state-of-the-art HgCdTe avalanche photodiode. Instrument- and atmosphere-specific systematic errors are discussed for different model environments. Finally, performance calculations were applied to vertical profiles retrieved from a past experimental campaign where a Raman lidar for water vapor measurements along with in-situ sensors for the HDO isotopologue measurements were deployed. A conclusion and perspectives for forthcoming calibration and validation field campaigns are given in section 4.

2 DIAL method and performance model for water vapor isotopologue measurement

2.1 Choice of the sensing spectral range

100 Remote sensing by DIAL relies on the alternate emission of at least two-two closely spaced laser wavelengths, named λ_{en} and λ_{off} , respectively in coincidence one coinciding with and out of a gas an absorption feature, line of the molecule of interest (λ_{on}) and the other tuned to the wing of the absorption line (λ_{off}), to retrieve a given species concentration. The key to independently measure HDO and H₂O abundances with a single instrument lies thus in the proper selection of a spectral region where: i) the two molecules display well separated, significant absorption lines while minimizing the interference from other atmospheric 105 species, and ii) the selected lines should preserve relatively equal lidar signal dynamic and relative precision ranges for both isotopologues. This makes the line selection rather limited. Using spectroscopic data from the HITRAN 2016 database (Gordon et al., 2017), we investigated the possibilities for HDO sounding up to 4 µm, where robust pulsed nanosecond lasers or optical parametric oscillator sources based on mature lasers or nonlinear crystals components can be developed (Godard, 2007). Figure 1a shows that HDO lines are strong in the 2.7 µm region but overlap with an even more dominant H₂O absorption band. 110 Considering the state of possible commercial photodetector technologies, we chose to limit the range of investigation to 2.6 µm, corresponding to the possibilities offered by InGaAs photodiodes. In the telecom wavelength range, which offers both mature laser sources and photodetectors, HDO absorption lines are too weak to be exploited for DIAL measurements over 1-3 km. The same argumentation holds for wavelengths towards 2.05 µm (see Fig. 1b) which have been extensively studied for space-borne CO₂ IPDA lidar sensing (Singh et al., 2017; Ehret et al., 2008). However, the 2 µm region seems to offer an 115 interesting possibility in terms of absorption strength as well as technical feasibility of pulsed, high-energy, single-frequency laser sources (Geng et al., 2014). The spectral window between 1982-1985 nm is well suited to meet the mentioned requirements as illustrated in Fig. 1c. In this paper we will focus on the linesH₂¹⁶O line at the positions-5043.0475 cm⁻¹ (1982.93 nm) and the HD¹⁶O lines at 5044.2277 cm⁻¹ (1982.47 nm) and 5040.4937 cm⁻¹ (1983.93 nm), respectively for H₂O and HDO (hereafter referred to as option 1HDO options (1) and (2), allowing for a sufficiently high absorption over several 120 kilometers with negligible interference from other gas species. Additionally, a second option for H₂O slightly detuned from the absorption peak at 1982.97 nm will be discussed as a possibility to reduce the temperature sensitivity of the DIAL measurement (hereafter referred to as H₂O option (2)-)). Wavelength switching will be realized on a shot-to-shot basis to consecutively address the chosen on-line wavelengths and the off-line wavelength at 1982.25 nm and the on line wavelengths for H₂O (1/2) and HDO₇ (1) or the off-line wavelength at 1983.72 nm for HDO (2). As shown in Fig.- 1c, the HDO absorption 125 line at 1982.47 nm is accompanied by a non-negligible H₂O absorption which has to be corrected for when retrieving the volume mixing ratio and thus adding a bias dependent on the accuracy of the H₂O measurement at 1982.93 nm. Alternatively, measurement within the spectral window between 1983.5 nm and 1984.5 nm is also possible for simultaneous H₂O and HDO probing, however with weaker absorption giving Furthermore, the interfering H₂O line has a ground-state energy of 2756 cm⁻¹ 1 (see Table 1) which makes it highly temperature sensitive. Probing HDO at 1982.47 nm thus requires highly accurate 130 knowledge of the H₂O and temperature profile through auxiliary measurements (lidar, radio sounding). The alternative second option for HDO at 1983.93 nm avoids any H₂O interference, however with slightly weaker absorption optical depth it gives rise to smaller signal-to-noise ratios and consequently increased measurement statistical uncertainty. In any of the proposed cases, addressing the on-line and off-line spectral features requires a tuning capability larger than 0.5 nm which can be offered, for instance, by an optical parametric oscillator source (Cadiou et al., 2016; Barrientos Barria et al., 2014), which is envisioned to be used for the WaVIL system. It should be noted that the chosen absorption lines do not fulfil the criterion of temperature insensitivity as outlined by Browell et al. which imposes the strict knowledge of the temperature profile along the lidar line of sight from auxiliary measurements for an accurate isotopologue retrieval.

2.2 DIAL performance model

150

155

The objective of the presented performance model is to elaborate the precision achievable with the proposed DIAL instrument of the volume mixing ratios of the water isotopologues H_2O and HDO and thus of the precision on the measurement of HDO abundance (noted δD) which expresses the excess (or defect) of the deuterated isotope compared to a reference value of 311.5·10⁻⁶ (one HDO molecule for 3115 H_2O molecules) (Craig, 1961). Following the convention, the HDO abundance (in permil, ‰) is expressed as the deviation from that of the standard mean ocean water (SMOW) in the so-called notation:

$$\delta D = 1000 \times \left[\frac{[\text{HDO}]_{\text{sample}} / [\text{H}_2\text{O}]_{\text{sample}}}{[\text{HDO}]_{\text{SMOW}} / [\text{H}_2\text{O}]_{\text{SMOW}}} - 1 \right]$$
(1)

where [] represents the concentration of H_2O and HDO.

As schematically depicted in Fig. 2, the DIAL simulator consists of three sub-models describing atmospheric properties, lidar instrument parameters, and detector properties. Each model will be explained in a more details in the following paragraphs. The atmosphere model is based on a set of standard profiles of temperature, pressure, and humidity representative of different climate regions along with aerosol optical depth data of the AERONET database. Those data are exploited to calculate the atmospheric transmission using absorption cross-sections computed with the HITRAN2016 spectroscopic database (Gordon et al., 2017). Together with the model describing the lidar instrument, the calculated transmission data are used to feed the lidar equation in order to calculate the received power at each selected on-line and off-line wavelength. In a subsequent step, noise contributions arising from the detection unit are taken into account to estimate the signal-to-noise ratio. Then, we use an analytical approach based on an error propagation calculation to estimate the random error on the measured isotopic mixing ratios and thus the uncertainty of the δD retrieval obtained with the simulated instrumental parameters.

Starting from the lidar equation (Collis and Russell, 1976), the calculated received power as a function of distance r writes as:

$$P_{r}(r) = T_{r} \frac{A}{r^{2}} \beta_{\pi}(r) O(r) \frac{c}{2} T_{\text{atm}}^{2}(r) E_{p}$$
 (2)

where T_r is the receiver transmission, (assumed value of 0.5 for all calculations), A is the effective area of the receiving telescope, $\beta_{\pi}(r)$ is the backscatter coefficient, O(r) is the overlap function between the laser beam and the field of view of the receiving telescope, c is the speed of light, $T_{\text{atm}}(r)$ is the one-way atmospheric transmission and E_p the laser pulse energy. The DIAL technique is based on the emission of two wavelengths, one at or close to the peak of an absorption line (λ_{on}) and another tuned to the absorption line's wing (λ_{off}) in (λ_{on}) and out of (λ_{off}) coincidence with a target gas absorption line. Provided that the two laser pulses are emitted sufficiently close in wavelength and in time for the atmospheric aerosol content to be equivalent, the two wavelengthsthey experience the same backscattering along the line of sight, and the differential optical depth $\Delta \tau$ as the difference of on- and off-line optical depth at a measurement range r can be retrieved by:

$$\Delta \tau(r) = \frac{1}{2} \ln \left(\frac{P_{\text{off}}(r)}{P_{\text{on}}(r)} \right) \tag{3}$$

with P_{on} and P_{off} as the backscattered power signals for λ_{on} and λ_{off} , respectively. Using the optical depth measurement, the gas concentration can be retrieved at a remote range r within a range cell $\Delta r = r_2 - r_1$. Assuming Δr is sufficiently small, the water vapor content expressed as volume mixing ratio, which is assumed as constant within Δr , can then be derived by:

$$X_{\rm H2O}(r_1 \to r_2) = \frac{\Delta \tau(r_2) - \Delta \tau(r_1)}{\int_{r_1}^{r_2} WF(r) dr}$$
(4)

with WF(r) representing a weighting function defined as:

$$WF(r) = \left(\sigma_{\text{on}}(r) - \sigma_{\text{off}}(r)\right) \rho_{\text{sir}}(r) \tag{5}$$

where ρ_{air} is the total air number density and σ_{on} and σ_{off} are the on-line and off-line absorption cross-sections calculated with the HITRAN 2016 spectroscopic database assuming a Voigt profile. The given formulas are representation of the form:

$$\sigma(v) = \sigma_0 \frac{y}{\pi} \int_{-\infty}^{\infty} \frac{\exp(-t^2)}{v^2 + (x - t)^2} dt$$
 (6)

with

160

170

$$\sigma_0 = \frac{S}{\gamma_D} \left(\frac{\ln 2}{\pi}\right)^{1/2}$$

$$y = \frac{\gamma}{\gamma_D} (\ln 2)^{1/2}$$

$$x = \frac{\nu - \nu_0}{\nu_D} (\ln 2)^{1/2}$$

where S is the line strength, γ_D is the Doppler width, γ is the pressure broadened linewidth, and v_0 is the line center position. The temperature T dependence of the line strength is determined by the energy of the lower molecular state E" according to:

$$S = S_0 \left(\frac{T_0}{T}\right)^{3/2} \exp\left[\frac{E''hc}{k} \left(\frac{1}{T_0} - \frac{1}{T}\right)\right] \tag{7}$$

where T_0 is the reference temperature, k is the Boltzmann constant, and c is the speed of light.

Equation (3) is valid for the detection of the main isotopologue H₂O. For HDO however, the presence of H₂O absorption at the on-line wavelength of HDO (see Fig. 1c) necessitates an additional consideration of that bias for the inversion. Taking this into account, Eq. (3) changes to:

$$\Delta \tau_{\text{HDO}}(r) = \frac{1}{2} \ln \left(\frac{P_{\text{off}}(r)}{P_{\text{on}}(r)} \right) - \Delta \tau_{\text{H2O}}(r)$$
 (68)

where $\Delta \tau_{\rm H2O}$ represents the H₂O differential optical depth at the HDO on-line wavelength $\lambda_{\rm HDO}$ which can be calculated with the knowledge of the volume mixing ratio $X_{\rm H2O}$ measured at $\lambda_{\rm H2O}$.

To obtain an analytical expression for the random error in the concentration measurement, an error propagation calculation can be applied to Eqs. (3) and (4) assuming that the range cell interval Δr is sufficiently small and that the range cell resolution of the receiver is sufficiently high to consider $\Delta \tau(r_1)$ and $\Delta \tau(r_2)$ as uncorrelated. The absolute uncertainty in the volume mixing ratio X expressed as standard deviation $\sigma(X)$ can be calculated from the signal-to-noise ratios of the on- and off-line power signals as follows:

190
$$\sigma(X) = \frac{\Delta f}{\sqrt{2} \ WF \ c} \left(\frac{1}{SNR_{\text{on}}^2} + \frac{1}{SNR_{\text{off}}^2} \right)^{1/2}$$
 (79)

where Δf is the measurement bandwidth which is the same for the on- and off-line pulses since they are measured sequentially by the same detector. Finally, with both uncertainties in the volume mixing ratios $X_{\rm H2O}$ and $X_{\rm H2O}$ known, an estimation of the uncertainty in δD is obtained by applying an error propagation calculation to Eq. (1) in order to get the expected uncertainty expressed as variance:

195
$$\sigma(\delta D) = \left(\delta D + 1\right) \left\{ \left(\frac{\sigma(X_{\text{H2O}})}{X_{\text{H2O}}}\right)^2 + \left(\frac{\sigma(X_{\text{HDO}})}{X_{\text{HDO}}}\right)^2 \right\}^{1/2}$$
 (810)

2.3 Instrument and detector model

200

205

In order to estimate the feasibility of a DIAL measurement, calculations were performed for the transmitter and receiver parameters summarized in Table 12. The emitter of the DIAL system will be based on a generic optical parametric oscillator/optical parametric amplifier (OPO/OPA) architecture as the one developed in (Barrientos Barria et al., 2014). The combination of a doubly-resonant Nested Cavity OPO (NesCOPO) and an OPA pumped by a 1064 nm Nd:YAG commercial laser with 150 Hz repetition rate allows for single-frequency, high-energy pulses with adequate tunability. From this system we expect an extracted signal energy of up to 20 mJ at 1983 nm. For a more conservative estimate, we will also consider a lower-limit pulse energy of 10 mJ for our simulations. The receiver part consists of a Cassegrain-type telescope with a primary mirror of 40 cm in diameter. For the detection part, calculations were performed in a direct-detection setup for i) a commercial InGaAs PIN photodiode and ii) a HgCdTe avalanche photodiode (APD) specifically developed for DIAL applications in the 2 µm range, presented in (Gibert et al., 2018). The telescope field of view is determined by an aperture in the telescope's focal

plane. For better comparability, we assume the same aperture diameter of 1.2 mm for both the PIN photodiode and the APD. Given the small active area of the APD, aligning the optics and the imaging of the field of view on the detector might prove extremely challenging in practice. However, for our simulations we do not take this into account and assume the same imaging optics for both the PIN photodiode and the APD except for a reduced diaphragm diameter for the APD, thus resulting in different field of view angles however. The measurement bandwidth of the DIAL system is effectively determined by an electronic low-pass filter in the detection chain. In the simulation we use a bandwidth setting of 1 MHz corresponding to a spatial resolution of the retrieved isotopologue concentrations of 150 m. For all our calculations we assume signal averaging over an integration time of 10 min (3045) 000 laser shots for eachon- and off-line wavelength).

210

215

220

225

235

In order to quantify the measurement uncertainty in the retrieved isotope mixing ratios, random and systematic sources of errors are taken into account. Random errors in measuring the differential optical depth, and thus the species mixing ratio, are related to different noise contributions arising from the detection setups. For a single return-signal pulse, the associated noise power P_n consists of a constant detector and amplifier noise expressed as noise equivalent power NEP, shot noise due to background radiation P_{sky} , shot noise dependent on the pulse power $P(\lambda)$, as well as speckle noise $P_{sp}(\lambda)$:

$$P_{n} = \sqrt{\left(NEP^{2} + 2 \cdot e \cdot \left[P_{\text{sky}} + P(\lambda)\right] \cdot F / R\right) \cdot \Delta f + P_{\text{sp}}^{2}(\lambda)}$$
 (911)

where e is the elementary charge, F the excess noise factor (in case of the APD), R the detector responsivity (depending on quantum efficiency in case of the APD) and Δf the measurement bandwidth. The NEP of 600 fW Hz^{-1/2} for configuration i) featuring the InGaAs PIN photodiode is a conservative estimate by calculations based on the specifications of the photodiode and amplifier manufacturer (G12182-003K InGaAs PIN photodiode from Hamamatsu combined with a gain adjustable DHPCA-100 current amplifier from FEMTO). The background power $P_{\rm sky}$ depends on the background irradiance $S_{\rm sky}$ and the receiver geometry according to:

$$P_{\text{sky}} = \frac{\pi}{4} \cdot S_{\text{sky}} \cdot \Delta \lambda_f \cdot A_{\text{eff}} \cdot \theta_{\text{FOV}}^2$$
 (4012)

where $\Delta \lambda_f$, A_{eff} and θ_{FOV} are the optical filter bandwidth, effective receiver telescope area and field of view angle, respectively. A constant background irradiance of 1 W/m²/ μ m/sr and an optical filter bandwidth of 50 nm are used for all calculations. Assuming Gaussian beam characteristics, the speckle-related noise power is approximately given by (Ehret et al., 2008):

$$P_{\rm sp} = P(\lambda) \cdot \frac{\lambda \cdot 2\sqrt{\Delta f \cdot \tau_c}}{\pi \cdot R_{\rm tel} \cdot \theta_{\rm FOV}}$$
 (4113)

where R_{tel} denotes the telescope radius and τ_c the coherence time of the laser pulse corresponding to the pulse duration for a Fourier-transform-limited pulse. Finally, the overall time-averaged signal-to-noise ratio is given as the ratio of received power from Eq. (2) and total noise power from Eq. (911) multiplied by the square root of the number of laser shots N:

$$SNR = \frac{P_r}{P_r} \sqrt{N}$$
 (1214)

2.4 Atmosphere model

240

245

255

260

265

We constructed different atmospheric models for mid-latitude, arctic, and tropical locations to study the sensitivity of the DIAL measurement to environmental factors. The atmosphere model consists of vertical profiles of pressure, temperature and humidity (see appendix for origin of sounding data) which serve as input to calculate altitude-dependent absorption cross-sections using the HITRAN 2016 spectroscopic database. For the sake of simplicity, HDO mixing ratios were obtained from H_2O profiles simply by considering their natural abundance of 3.11 10^{-4} , i.e., variability in terms of the isotopic ratio δD is not assumed in our model. For each location, a baseline model was constructed by using the columns of pressure, temperature and volume mixing ratios averaged over the year of 2019. To reflect seasonal variations in our sensitivity analysis, we use profiles with the lowest and highest monthly averages of temperature and humidity (Fig. 3 a–c). To complement the atmospheric model, data of level 2 aerosol optical depth (AOD) from the AERONET database (https://aeronet.gsfc.nasa.gov/) were used. AERONET sun photometer products are usually available for wavelengths between 340 nm and 1640 nm. For extrapolation to the 2 μ m spectral region, we used the wavelength dependence of the AOD described by a power law of the form (Angström, 1929):

$$250 \quad \frac{AOD(\lambda)}{AOD(\lambda_{\rm p})} = \left(\frac{\lambda}{\lambda_{\rm p}}\right)^{-\alpha} \tag{1315}$$

where $AOD(\lambda)$ is the optical depth at wavelength λ , $AOD(\lambda_0)$ is the optical depth at a reference wavelength, and α represents the Angstrom exponent. The Angstrom exponent was obtained by fitting Eq. (4315) to the available AOD data in the above-mentioned spectral range in order to extrapolate further to 1.98 μ m. Histograms of the yearly distribution of the extrapolated AOD at 1.98 μ m are shown in the right column of Fig. 3 (g–i). Median values of the AOD are used for the baseline model. The lowest (AOD₁₀) and highest (AOD₉₀) decile values serve as input for the sensitivity analysis to model conditions of low and high aerosol charge, respectively. As a next step, vertical profiles of aerosol extinction are constructed by making basic assumptions about their shape and constraining their values by the extrapolated AOD. In our baseline model, the vertical distribution of aerosols is represented by an altitude-dependent Gaussian profile of the extinction coefficient with varying half-width depending on the location (Fig. 3 d–f). This type of profile roughly corresponds to the ESA Aerosol Reference Model of the Atmosphere (ARMA) (ARMA, 1999) which is plotted for each region normalized to the AOD₉₀-derived extinction profile maximum.

However, the distribution of tropospheric aerosols varies widely from region to region (Winker et al., 2013). To broadly reflect the different boundary layer characteristics for each environment, the extinction profile was adapted accordingly. In midlatitude regions, vertical aerosol distributions vary widely due regional and seasonal factors (Chazette and Royer, 2017). The PBL height can range from a few hundred meters up to 3 km (Matthias et al., 2004; Chazette et al., 2017). Assuming that

aerosols are mostly confined to the PBL and that the free-tropospheric contribution to aerosol extinction is weak, the half-Gaussian-shaped baseline model used for the simulations gives rise to 85% of AOD within the first 1.5 km. Since high aerosol loads in the free troposphere due to long-range dust transport are not uncommon over Western Europe (Ansmann et al., 2003), a dust scenario profile constrained by the highest-decile AOD was also investigated. Dust aerosols are represented by a Gaussian profile above the PBL extending well up to a height of 5 km. For this case, aerosol extinction in the PBL below 1.5 km accounts for half of the total AOD, while dust in the free troposphere accounts for the other half. At high latitudes, the boundary layer tends to be stable and extends from a few meters to a few hundred meters above ground. Our baseline Arctic extinction profile thus contains 95% of the AOD within the first 1.5 km since most aerosols are confined within the first kilometer of the troposphere as observed by space-born lidar during long-term studies of the global aerosol distribution (Di Pierro et al., 2013). The occurrence histogram in Fig. 3h shows very low values of AOD for most of the time in the available photometer products from February to September. The long-tailed wing of the asymmetric distribution towards higher values can be explained by seasonally occurring episodes of arctic haze due to anthropogenic aerosols transported from mid-latitude regions (winter to spring) and boreal forest fire smoke during the summer season (Tomasi et al., 2015; Chazette et al., 2018). Similar to the dust scenario for the mid-latitude model, haze and smoke events are modelled by an additional Gaussian profile in the free troposphere constrained by the highest-decile AOD. Extinction profiles representing the tropical environment of La Réunion Island, where sea salt aerosols can be assumed to be the dominant aerosol species, are chosen such that 90% of the AOD is contributed to the first 1.5 km.

Vertical profiles of the aerosol backscatter coefficient were calculated assuming, for the sake of simplicity, a constant extinction-to-backscatter ratio (lidar ratio) of 40 sr throughout all sets of extinction profiles.

3 Simulation results and discussion

3.1 Instrument random error

270

275

280

285

290

This section aims to quantify the random error on the mixing ratio measurement depending on instrument settings such as laser pulse energy and the type of detector employed. All calculations are based on the mid-latitude baseline atmosphere model assuming vertical sounding of the lower troposphere with aerosols confined to the lowest 2-km. Considering a simple calculation of random errors we will discuss their implications on the precision of the measurement of range-resolved δD profiles. Given the instrument parameters presented in Table $\frac{1}{2}$, the dominant noise contributions are estimated which are shown for a single on-line pulse in Fig. 4 for both detector configurations.

As expected, the <u>overallelectronic</u> noise level is significantly reduced by roughly one order of magnitude for the HgCdTe APD combined with a transimpedance amplifier due to a low combined NEP of 75 fW Hz^{-1/2} compared to 600 fW Hz^{-1/2} for the amplifier of the InGaAs PIN detector. In fact, shot noise and <u>electronic noisespeckle</u> are <u>at the same level predominant</u> for the APD for <u>a height up to 1 kmthe first kilometer of range</u> whereas the electronic noise of the transimpedance amplifier is the predominant contribution over the entire range for the commercial PIN detector. Signal-to-noise ratios up to 10^2 are obtained

for a single measurement pulse within the first kilometer. Integrating over $\frac{3045}{4}$ 000 laser shots (equivalent to 10 min averaging time if the two-on-line and one off-line wavelengths are addressed sequentially) would increase the signal-to-noise ratios to over 10^4 in the first kilometer for both detectors and to values around 10^2 at a 2 km range for the commercial PIN detector and 10^3 for the HgCdTe APD.

300

305

310

315

320

325

330

The expected relative random errors on the mixing ratios of H₂O and HDO are shown separately in Figs. 5a and 5b-for each detector in the upper and lower panels of Fig. 5. We examined two scenarios with different laser pulse energies of 10 mJ and 20 mJ, a measurement bandwidth of 1 MHz (150 m range cell resolution) and an integrating time of 10 minutes for a repetition rate (on-off rate) of 150 Hz. The simulation based on the 20 mJ configuration gives an estimation of the best-case precision limit of the DIAL system. The second configuration with 10 mJ pulse energy can be understood as a lower limit on the precision of measuring mixing ratios of H₂O and HDO, and finally δD . As shown in Fig. 5a5, a relative random error of well under 1% on the mixing ratio of both H₂O and HDO can be achieved within the first kilometer for both detectors and 20 mJ pulse energy. The degraded precision for measuring HDO is due to its lower differential absorption compared to H₂O. The slight difference in optical depth for the two HDO options leads only to a small loss in precision for wavelength option 2. For the low-noise APD shown in the bottom panel of Fig. 555, the simulations show that even for the conservative assumption of 10 mJ pulse energy, the relative error stays below 1% for both H₂O and HDO over a range of 1.5 km corresponding to typical heights of the planetary boundary layer. H₂O uncertainties were calculated for sounding at the peak of the absorption line (option 1). The simulation results also reveal a sharp rise in the random uncertainty towards longer distances which is attributed to the drastic decline of aerosol backscatter in the free troposphere in our model. The sharp fall of the random error within the first 200-300 m is due to the increasing overlap between laser beam and telescope field of view imaged onto the detector described by the overlap function O(r) in Eq. (2). This overlap term is zero right in front of the lidar instrument and reaches unity after $\frac{1}{2}$ few hundred meters. It should be noted that around 450 m for the here described configuration. It should be noted that for the range zone of non-uniform overlap, slight differences between the on- and off-line overlap, for example due to laser beam pointing, can induce significant systematic errors. From a practical point of view, the expected lowest instrument range is thus closer to 0.5 km than the distance suggested by the location of the random error minima around 250 m. H₂O uncertainties were ealculated for sounding at the peak of the absorption line (option 1).

Figure Figures 5c shows and 5f show the expected precision in δD which depends on the relative random errors of the volume mixing ratios for H₂O and HDO (see Eq. (79)). For the commercial InGaAs PIN photodiode we find for the limiting best-case of high measurement precision configuration (20 mJ pulse energy, 1 MHz bandwidth) that the absolute value of uncertainty in δD is below 3‰ within a range of 1 km. The 10 mJ configuration also allows for measurement of δD , however with deteriorated absolute precision up to 10‰ within the first kilometer. Simulations with For greater ranges, the precision levels decline rapidly and are not sufficient to resolve variations of δD in the order of a few tens of permil. The use of a HgCdTe APD detector can overcome this limitation where calculations indicate that an absolute precision level lower better than 10‰ within the first 1.5a range of close to 2 km can be achievable with 20 mJ laser energy.

3.2 Sensitivity to atmospheric variability

335

340

345

350

The sensitivity of the DIAL instrument to the variability in temperature, humidity, and aerosol load was investigated for the mid-latitude, arctic and tropical atmosphere models. In the following analysis, the relative random error (precision) is used to compare the influence of each atmospheric parameter under investigation. Simulation results are summarized in Fig. 6 for targeting H₂O at 1982.93 nm (blue) and HDO at 1983.93 nm (red). Here again, we consider a measurement bandwidth of 1 MHz (150 m range cell resolution), and an integrating time of 10 minutes for a repetition rate of 150 Hz. All calculations have been performed with the InGaAs PIN detector and assuming a laser pulse energy of 20 mJ.

Starting with temperature, no effect on the measurement random error was found when simulating under conditions of lower and higher temperature compared to the average atmospheric columns. Comparing the three baseline models of mid-latitude, tropical and arctic environments, the performance simulations find that highest precision measurements can be achieved under tropical conditions due to high humidity levels and favourable aerosol backscattering. Relative random errors lower than 0.1% for H₂O are achievable within the first kilometer. The precision for H₂O degrades faster than for HDO with increasing range due to strong absorption leading to low return signals. On the contrary, random uncertainties for the arctic environment are almost one order of magnitude higher due to rather dry conditions in terms of WVMR and low aerosol content observed at the Eastern Greenland AERONET station of Ittoqqortoormiit. A high sensitivity to seasonal variability of the humidity profile was observed for the arctic model, whereas variations of humidity in the tropics throughout the year are small and thus only slightly affect the expected measurement precision. The simulations also clearly show the influence of aerosols on the performance of DIAL measurements. For all three locations, the precision gain between the low-charge (lowest-decile AOD) and high-charge (highest-decile AOD) aerosol model is roughly one order of magnitude. The presence of free-tropospheric aerosols, for example due to long-range dust transport in the mid-latitudes and arctic haze or boreal forest fire smoke in the Arctic, leads to significant improvements in the precision at altitudes beyond the atmospheric boundary layer. Adapting the measurement bandwidth, along the line of sight for instance, and of course adapting the integrating time could be envisioned to retrieve nominal performances under these different atmospheric conditions.

355 3.3 Systematic errors

Systematic errors are associated with an uncertainty in the knowledge of atmospheric, spectroscopic, and instrument-related parameters when obtaining the VMR from the measured differential optical depth according to Eq. (4). Expressed in a general form, errors were estimated by calculating the VMR retrieval sensitivity to a deviation δY from a reference parameter Y:

$$\varepsilon_s = \max \left\{ \frac{\left| X(Y) - X(Y \pm \delta Y) \right|}{X(Y)} \right\}$$
 (1416)

For the case of atmospheric systematic errors, the reference parameter Y used for the VMR retrieval stands for either the vertical pressure or temperature profile of the baseline atmospheric model. The systematic error due to an uncertainty in the knowledge of the temperature profile was calculated for temperature deviations δT from the reference profile ranging from

 ± 0.5 K to ± 2 K. This range of accuracy can be obtained by in situ sensors or an additional lidar instrument for the temperature profile which is necessary to calculate the temperature-dependent absorption cross sections for the concentration retrieval. As shown in Fig. 7, this kind of error can lead to a significant contribution to the error budget. The analysis shows that sounding H_2O at the absorption peak is especially sensitive to temperature uncertainties and that a measurement with the on-line wavelength shifted off the absorption peak (eption 2) significantly reduces this bias. H_2O option 2) significantly reduces this bias. Table 3 gives an overview of the calculated biases comparing the three different atmospheric models. Note the significant temperature bias for HDO option 1 in regions with high water vapor content due to the highly temperature sensitive H_2O interference line. Similarly, a pressure deviation δp ranging from 0.5 hPa to 2 hPa was used to estimate the error due to an uncertainty in the pressure profile. In this case, H_2O wavelength option 2 is more sensitive to such an uncertainty. The resulting bias on the measurement of HDO is found to be negligible. Note the difference between the two options for probing H_2O . Shifting the online wavelength off the absorption peak (option 2) results in a noticeable reduction in the temperature error. However, this comes at the expense of increased pressure error and lower signal-to-noise ratio and thus increased random error for unchanged laser energy, integration time, and bandwidth. Considering the mentioned systematic error contributions, option 2 for H_2O proves to be the preferred wavelength choice with the intention of reducing the systematic error, especially if the temperature profile along the line of sight is not known with accuracy better than ± 0.5 K.

For the case of instrument-related errors, we assume perfect estimated systematic errors arising from the laser wavelength locking control and spectral quality of the laser source and beam. To estimate only the systematic error arising from the accuracy of the transmitter on and offline wavelengths. For our estimate we use the sensitivity to laser line stability a laser frequency deviation δf ranging from 2.5 MHz to 10 MHz corresponding to wavelength stabilities reliably achievable over several minutes with our envisioned OPO/OPA approach coupled to a commercial wavemeter, which can suffer thermal drifts of a few MHz over several tens of minutes. The relative wavelength error was calculated according to Eq. (1416) by introducing a wavelength detuning δf to the on- and offline wavelengths. Due to the narrower absorption line of H₂O at 1982.93 nm, we find that such wavelength detuning results in a much largergreater error compared to the spectrally larger HDO line. Option 2 for H₂O measurement drastically reduces the wavelength error. The systematic error due to the finite laser linewidth was estimated numerically by substituting the absorption cross sections of Eq. (5) by the effective absorption cross sections defined as:

Considering the three mentioned systematic error contributions, option 2 proves to be the preferred wavelength choice with the intention of reducing the systematic error, especially if the temperature profile along the line of sight is not known with accuracy better than ±0.5 K.

$$\sigma_{eff} = \frac{\int L(\nu, r) \cdot \sigma(\nu, r) \cdot d\nu}{\int L(\nu, r) \cdot d\nu}$$
(17)

where L represents spectral intensity distribution of the laser transmitter and v denotes the wavenumber. The laser spectral distribution L is assumed to be an altitude-independent Gaussian function with a width of 50 MHz (FWHM), which correlates roughly to the 10 ns pulse duration assuming transform-limited pulses. For comparison, the air broadened Laurentzian widths

of the absorption lines under standard atmospheric conditions are on the order of a few GHz. The calculated relative error is on the order of 0.1% for the narrowest (thus most critical) H₂O line at 1982.93 nm.

Another systematic error arises for thesounding HDO retrievalat 1982.47 nm (option 2) from the insufficient knowledge of the optical depth due to athe non-negligible H₂O absorption feature at the HDO line at 1982.47 nm. Assuming a relative uncertainty of 1.52% in the VMR profile of H₂O, which is a conservative estimate for the combined systematic error of the H₂O measurement due to temperature, pressure, and wavelength uncertainty, calculations reveal relative errors in the VMR retrieval varying between 0.2328% for the arctic model and 0.5583% for the tropical model. Considering this additional bias and the highly temperature sensitive H₂O interference line for HDO option 1, option 2 should be the preferred wavelength option for HDO in any case. It should be noted that even for the measurement of H₂O an interference contribution due to higher HDO absorption at the off-line wavelength leads to a bias. However, this error is relatively small compared to other systematic errors and the achievable random error—which justifies the proposed method of calculating the H₂O VMR with no a priori knowledge of HDO and then using the obtained profile to correct the differential optical depth of the HDO retrieval according to Eq. (6).

Finally, systematic errors in the VMR retrieval due to uncertainties related to spectroscopic parameters were analyzed by introducing deviations between 1% to 5% to the HITRAN 2016 parameters of line intensity and air-broadened width and deviations of 1% to 10% to the temperature-dependence width coefficient and the pressure shift parameter. The resulting systematic errors are shown in Fig. 8 for each parameter. Uncertainties in parameters of line intensity and air-broadened width largely contribute to the error budget highlighting the importance of the precise knowledge of these quantities. It should be noted that the assumed uncertainties have a rather demonstrative character as their precise quantification is still the subject of ongoing spectroscopic studies. A summary of the presented systematic errors in the form of an error budget for each of the three atmospheric models is given in Table 23.

3.4 Precision estimate applied to field campaign data

400

410

415

420

425

In order to complete our previous numerical studies and relate to more realistic atmospheric conditions, we present here the results of performance calculations initialized with observations obtained during the L-WAIVE (Lacustrine-Water vApor Isotope inVentory Experiment) field campaign at the Annecy lake in the French alpine region (Chazette et al., 2020). This experiment was specifically carried out in order to obtain reference profiles that can be used to simulate the WaVIL lidar vertical profiles. Hence, the data include vertical profiles of pressure and temperature as well as vertical profiles of H₂O and HDO isotopologue concentrations which were obtained by an ultra-light aircraft equipped with an in-situ cavity-ring-down-spectrometer (CRDS) isotope analyzer. As aerosols were present above the planetary boundary layer on 14 June 2019, we chose data acquired from that day, ranging up to an elevation of 2.3 km. To simulate atmospheric conditions during the measurement campaign as realistically as possible, we used aerosol extinction data from the lidar WALI (Weather and Aerosol Lidar) (Chazette et al., 2014) operated during the L-WAIVE campaign on the same day (see Fig. 8a9a). The backscatter coefficient was estimated with a lidar ratio of 50 sr and extrapolated to a wavelength of 2 μm using the Angstrom exponent

derived from sun-photometer measurements. For the purpose of our simulation study, we do not take into account any measurement uncertainties in the described profiles. Figures 8b9b and 8e9c show the expected in-situ measured δD profile from the field campaign and the hypothetical precision in the DIAL retrieved isotopic ratio in terms of δD as shaded area depending on detector characteristics and laser energy (calculationif that same profile was measured with the here presented DIAL system (precision estimate based on wavelength option 1 for H₂O), and option 2 for HDO). For the commercial InGaAs PIN photodiode the simulations show for the limiting optimum case of 20 mJ laser energy that the uncertainty related to noise is sufficiently low so that the characteristic variations in the experimentally obtained δD profile could be fully resolved with the proposed DIAL system. The expectedIn terms of absolute precision for this configuration, which is well belowvisualized as the width of the shaded error band around the in-situ profile, δD could be determined with a precision better than 5% within the first 1.5 km and reaches better than 10% at a range height of 2.3 km. A setup with 10 mJ would deliver an absolute precision of close to 20% at that height. The expected precisions are on the order of or better than the columnar measurements obtained with other remote sensing techniques deployed from the ground (between 5 and 35% for Fourier Transform Infrared Spectrometer and Total Carbon Column Observing Network) or from space (~40% for the Tropospheric Emission Spectrometer and the Infrared Atmospheric Sounding Interferometer, see Table 1 of Risi et al., 2012) but with a much greater resolution on the vertical. On the other hand, the expected precision is roughly 2 to 4 times lower than for in situ airborne CRDS measurements with a similar vertical resolution (see Table 3 of Sodemann et al., 2017). Simulations performed with the HgCdTe APD indicate extremely promising precision levels over the entire range of under 3\% and 5\% (in absolute terms) for 20 mJ and 10 mJ, respectively. It should be noted that the presented profiles represent a rather favourable case since the aerosol backscatter coefficient increases with altitude (due to the presence of an elevated dust layer) which is the contrary to the baseline atmospheric models described in the previous numerical analysis. These simulations incorporating observed H₂O and HDO profiles clearly show the potential of a ground-based DIAL instrument to measure isotopic mixing ratios with high spatio-temporal resolution in the lower troposphere.

450 4 Conclusion

430

435

440

445

Probing the troposphere for water isotopologues with high spatio-temporal resolution is of great interest to study processes related to weather and climate, atmospheric radiation, and the hydrological cycle. In this context, the Water Vapor Isotope Lidar (WaVIL), which will measure H₂O and HDO based on the differential absorption technique, is under development. The spectral window between 1982–1984 nm has been identified to perform such measurements. Indeed, HDO displays sufficiently high ab sorption lines in this range. Interference with the main isotopologue H₂O is manageable, especially since both species would be measured simultaneously. The selected absorption lines of H₂O and HDO have a sufficiently high line strength to probe the lower 1.5 km of atmosphere with better than 1% relative error in the tropics and mid-latitude regions with high water vapor concentrations. The selected absorption lines are temperature sensitive, requiring an accurate knowledge of the

temperature profile along the line of sight for the concentration retrieval. Such a profile would have to be provided by auxiliary measurements, for example by using a Raman lidar.

460

465

470

475

480

485

490

We performed a sensitivity analysis and an error budget for this system taking instrument-specific and environmental parameters into account. The numerical analysis included models of mid-latitude, polar, and tropical environments with realistic aerosol loads derived from the AERONET database extrapolated to the 2 μ m spectral region. We showed that the retrieval of H₂O and HDO mixing ratios is possible with relative precisions and merrors better than 1% within the atmospheric boundary layer (<2 km) in mid-latitude and tropical conditions, the latter giving rise to the highest precision due to favourable differential absorption. Based on these precisions of the mixing ratio measurements, the isotopic abundance expressed in δD notation can be derived with a precision necessary to resolve vertical variations in δD of a few tens of permil. Performance simulations also revealed differences in precision of almost one order of magnitude between the tropical and arctic model. Reduced precision under arctic conditions is due to low water vapour content and reduced aerosol load. These findings have been obtained for laser pulse energies of 20 mJ, a measurement bandwidth of 1 MHz (150 m range resolution), an integration time of 10 min, and a commercial InGaAs PIN photodiode. As an interesting perspective option, we also investigated the theoretical performance of a state-of-the-art HgCdTe avalanche photodiode featuring a NEP reduced roughly by one order of magnitude. The use of such a detector would relax the requirement on laser energy and integration time and enable high-precision, range-resolved measurement of the isotopic ratio.

An error budget has been performed to outline systematic errors due to uncertainties in atmospheric, spectroscopic, and instrument-related parameters. The H₂O on-line wavelength at 1982.93 nm shows a pronounced temperature sensitivity imposing strict requirements on accurate temperature profiles for the VMR retrieval. This can be mitigated by tuning the online wavelength to 1982.97 nm which, however, comes at the cost of slightly increased pressure sensitivity and reduced differential absorption. slightly reduced differential absorption. For the HDO isotopologue, two wavelength options have been studied. Option 2 with the on-line wavelength at 1983.93 nm was found to be more suitable since it has no H₂O interference (as is the case for HDO option 1 at 1982.47 nm). The slightly smaller differential absorption for option 2 is a price worth paying and the resulting increase in random error can be offset by longer signal averaging. Including systematic errors due to inexact spectroscopic parameters in our analysis, we highlighted the importance of their accurate knowledge for DIAL measurements and the necessity for ongoing spectroscopic studies of water vapor isotopologues in the two-micrometer region. Finally, using a measured H₂O/HDO profile obtained during the recent L-WAIVE field campaign, our calculations have shown that sufficient precision in the mixing ratios of H₂O and HDO can be achieved with the presented system parameters so that characteristic, vertical variations of the isotopic content δD eanobserved during the field campaign could be resolved with the proposed DIAL system, showing the potential to complement existing methods. Although an effort has been made to conduct the sensitivity analysis and error budget as thoroughly as possible, it should be nevertheless noted that the predicted performance of the presented DIAL system can be understood as a best-case scenario. Assumptions made for the transmitter model, such as no laser beam pointing or perfect spectral purity, are very challenging aspects in the actual laser development. Future work will consist of improving our knowledge in the spectroscopy of HDO in the 1982–1984 nm spectral region and testing the DIAL system in the framework of a forthcoming field campaign.

Appendix A

Table A1 lists databases and locations used to derive the three atmospheric models discussed in this paper. Available data from the year of 2019 was used for all locations.

Table A1. Overview of atmospheric sounding and AERONET sites used to derive an atmosphere model for the sensitivity analysis. For all sites data from 2019 was used. Note that for the arctic station, AERONET photometer products are from February until September.

	Sounding profiles (pressure, temperature, humidity)	AERONET (Level 2 aerosol optical depth)				
Mid-latitude station:	Trappes	Palaiseau				
Paris region, France	48.77°N, 2.01°E	48.71°N, 2.22°E				
	Météo France data					
	(https://donneespubliques.meteofrance.fr/)					
Arctic station:	Ittoqqortoormiit	Ittoqqortoormiit				
Ittoqqortoormiit, Denmark	70.49°N, 21.95°W	70.49°N, 21.95°W				
	University of Wyoming data	February – September 2019				
	(http://weather.uwyo.edu/upperair/sounding.html)	, ,				
Tropical station:	La Réunion (Gillot)	La Réunion (St. Denis)				
La Réunion Island, France	20.89°S, 55.51°E	20.90°S, 55.49°E				
	Météo France data (https://donneespubliques.meteofrance.fr/)					

500 Author contribution

Conceptualization of measurement concept, J.H., J.-B.D., M.R., R.S., A.G., J.-M.M., J.T., P.C. and C.F.; lidar performance simulator, J.H.; L-WAIVE campaign data curation, H.S., A.S., J.T. and P.C.; writing—original draft preparation, L.R., C.C., M.R. and J.H.; writing—review and editing, all authors; project administration, C.F. and C.E.; All authors have read and agreed to the published version of the manuscript.

505 Competing interests

The authors declare that they have no conflict of interest.

Acknowledgements

The authors would like to thank the AERONET network and responsible PIs for sun photometer products (https://aeronet.gsfc.nasa.gov/, last access: 14 November 2020). The authors acknowledge the provision of meteorological

sounding data by the University of Wyoming (http://weather.uwyo.edu/upperair/sounding.html, last access: 16 November 2020) and the French national meteorological service Météo-France (https://donneespubliques.meteofrance.fr, last ac-cess: 26 November 2020).

This work was partially funded through WAVIL project: ANR grant Nr. ANR-16-CE01-0009 and has received funding from the European Union's Horizon 2020 research and innovation program under grand agreement Nr. 821868. H.S. and A.S. acknowledge funding obtained through the ERC Consolidator Grant Nr. 773245 (ISLAS).

References

515

- Agence Nationale de la Recherche: Differential absorption lidar for monitoring water vapour isotope HDO in the lower troposphere WaVIL. Available online: https://anr.fr/Project-ANR-16-CE01-0009 (accessed on 12 March 2021).
- Ångström, A.: On the Atmospheric Transmission of Sun Radiation and on Dust in the Air. Geogr. Ann., 11 (2), 156–166.

 doi:10.1080/20014422.1929.11880498, 1929.
 - Ansmann, A., Bösenberg, J., Chaikovsky, A., Comerón, A., Eckhardt, S., Eixmann, R., Freudenthaler, V., Ginoux, P., Komguem, L., Linné, H., Márquez, M. Á. L., Matthias, V., Mattis, I., Mitev, V., Müller, D., Music, S., Nickovic, S., Pelon, J., Sauvage, L., Sobolewsky, P., Srivastava, M. K., Stohl, A., Torres, O., Vaughan, G., Wandinger, U., Wiegner, M.: Long-Range Transport of Saharan Dust to Northern Europe: The 11–16 October 2001 Outbreak Observed with EARLINET. J. Geophys. Res. Atmospheres, 108 (D24). doi:10.1029/2003JD003757, 2003.
 - Barrientos Barria, J., Mammez, D., Cadiou, E., Dherbecourt, J. B., Raybaut, M., Schmid, T., Bresson, A., Melkonian, J. M., Godard, A., Pelon, J., Lefebvre, M.: Multispecies High-Energy Emitter for CO2, CH4, and H2O Monitoring in the 2 μm Range. Opt. Lett., 39 (23), 6719–6722. doi:10.1364/OL.39.006719, 2014.
- Bösenberg, J.: Differential-Absorption Lidar for Water Vapor and Temperature Profiling. In Lidar: Range-Resolved Optical Remote Sensing of the Atmosphere; Weitkamp, C., Ed.; Springer Series in Optical Sciences; Springer: New York, NY, pp 213–239. doi:10.1007/0-387-25101-4 8, 2005.
 - Browell, E., Ismail, S., Grossmann, B.: Temperature sensitivity of differential absorption lidar measurements of water vapor in the 720-nm region, Appl. Opt. 30, 1517-1524, 1991.
- Bruneau, D., Quaglia, P., Flamant, C., Meissonnier, M., Pelon, J.: Airborne Lidar LEANDRE II for Water-Vapor Profiling in the Troposphere. I. System Description. Appl. Opt., 40 (21), 3450–3461. doi:10.1364/AO.40.003450, 2001.
 - Cadiou, E., Dherbecourt, J.-B., Gorju, G., Raybaut, M., Melkonian, J.-M., Godard, A., Pelon, J., Lefebvre, M.: 2-µm Direct Detection Differential Absorption LIDAR For Multi-Species Atmospheric Sensing. In Conference on Lasers and Electro-Optics (2016), paper STh1H.2; Optical Society of America; p STh1H.2. doi:10.1364/CLEO_SI.2016.STh1H.2, 2016.
- Chazette, P., Marnas, F., Totems, J.: The Mobile Water Vapor Aerosol Raman LIdar and Its Implication in the Framework of the HyMeX and ChArMEx Programs: Application to a Dust Transport Process. Atmospheric Meas. Tech., 7 (6), 1629–1647. doi:10.5194/amt-7-1629-2014, 2014.

- Chazette, P. and Royer, P.: Springtime Major Pollution Events by Aerosol over Paris Area: From a Case Study to a Multiannual Analysis. J. Geophys. Res. Atmospheres, 122 (15), 8101–8119. doi:10.1002/2017JD026713, 2017.
- Chazette, P., Totems, J., Shang, X.: Atmospheric Aerosol Variability above the Paris Area during the 2015 Heat Wave Comparison with the 2003 and 2006 Heat Waves. Atmos. Environ., 170, 216–233. doi:10.1016/j.atmosenv.2017.09.055,
- 2017.
 - Chazette, P., Raut, J.-C., Totems, J.: Springtime Aerosol Load as Observed from Ground-Based and Airborne Lidars over Northern Norway. Atmospheric Chem. Phys., 18 (17), 13075–13095. doi:10.5194/acp-18-13075-2018, 2018.
 - Chazette, P., Flamant, C., Sodemann, H., Totems, J., Monod, A., Dieudonné, E., Baron, A., Seidl, A., Steen-Larsen, H. C.,
- Doira, P., Durand, A., Ravier, S.: The Lacustrine-Water Vapor Isotope Inventory Experiment L-WAIVE. Atmospheric Chem. Phys. Discuss. Prepr., 1–43. doi:10.5194/acp-2020-1194, 2020.
 - Collis, R. T. H. and Russell, P. B.: Lidar Measurement of Particles and Gases by Elastic Backscattering and Differential Absorption. In Laser Monitoring of the Atmosphere; Hinkley, E. D., Ed.; Topics in Applied Physics; Springer: Berlin, Heidelberg, pp 71–151. doi:10.1007/3-540-07743-X_18, 1976.
- 555 Craig, H.: Standard for Reporting Concentrations of Deuterium and Oxygen-18 in Natural Waters. Science, 133 (3467), 1833–1834. doi:10.1126/science.133.3467.1833, 1961.
 - Di Pierro, M., Jaeglé, L., Eloranta, E. W., Sharma, S.: Spatial and Seasonal Distribution of Arctic Aerosols Observed by the CALIOP Satellite Instrument (2006–2012). Atmospheric Chem. Phys., 13 (14), 7075–7095. doi:10.5194/acp-13-7075-2013, 2013.
- Ehret, G., Kiemle, C., Wirth, M., Amediek, A., Fix, A., Houweling, S.: Space-Borne Remote Sensing of CO2, CH4, and N2O by Integrated Path Differential Absorption Lidar: A Sensitivity Analysis. Appl. Phys. B, 90 (3), 593–608. doi:10.1007/s00340-007-2892-3, 2008.
 - European Space Agency. ARMA Reference Model of the Atmosphere, In Technical Report APP-FP/99-11239/AC/ac, 1999
 - Galewsky, J., Steen-Larsen, H. C., Field, R. D., Worden, J., Risi, C., Schneider, M.: Stable Isotopes in Atmospheric Water
- Vapor and Applications to the Hydrologic Cycle. Rev. Geophys., 54 (4), 809–865. doi:10.1002/2015RG000512, 2016.
 - Geng, J. and Jiang, S.: Fiber Lasers: The 2 μm Market Heats Up. Opt. Photonics News, 25 (7), 34–41. doi:10.1364/OPN.25.7.000034, 2014.
 - Gibert, F., Dumas, A., Rothman, J., Edouart, D., Cénac, C., Pellegrino, J.: Performances of a HgCdTe APD Based Direct Detection Lidar at 2 Mm. Application to Dial Measurements. EPJ Web Conf., 176, 01001. doi:10.1051/epjconf/201817601001, 2018.
- 570 doi:10.1051/epjconf/201817601001, 2018.
 - Godard, A.: Infrared (2–12 μm) Solid-State Laser Sources: A Review. Comptes Rendus Phys., 8 (10), 1100–1128. doi:10.1016/j.crhy.2007.09.010, 2007.
 - Gordon, I. E., Rothman, L. S., Hill, C., Kochanov, R. V., Tan, Y., Bernath, P. F., Birk, M., Boudon, V., Campargue, A., Chance, K. V., Drouin, B. J., Flaud, J.-M., Gamache, R. R., Hodges, J. T., Jacquemart, D., Perevalov, V. I.; Perrin, A., Shine,
 - K. P., Smith, M.-A. H., Tennyson, J., Toon, G. C., Tran, H., Tyuterev, V. G., Barbe, A., Császár, A. G., Devi, V. M.,

Furtenbacher, T., Harrison, J. J., Hartmann, J.-M., Jolly, A., Johnson, T. J., Karman, T., Kleiner, I., Kyuberis, A. A., Loos, J., Lyulin, O. M., Massie, S. T., Mikhailenko, S. N., Moazzen-Ahmadi, N., Müller, H. S. P., Naumenko, O. V., Nikitin, A. V., Polyansky, O. L., Rey, M., Rotger, M., Sharpe, S. W., Sung, K., Starikova, E., Tashkun, S. A., Auwera, J. V., Wagner, G., Wilzewski, J., Wcisło, P., Yu, S., Zak, E. J.: The HITRAN2016 Molecular Spectroscopic Database. J. Quant. Spectrosc. Radiat. Transf., 203, 3–69. doi:10.1016/j.jqsrt.2017.06.038, 2017.

580

595

605

pp.D05304, 2012.

- Hamperl, J., Capitaine, C., Santagata, R., Dherbecourt, J.-B., Melkonian, J.-M., Godard, A., Raybaut, M., Régalia, L., Grouiez, B., Blouzon, F., Geyskens, N., Evesque, C., Chazette, P., Totems, J., Flamant, C.: WaVIL: A Differential Absorption LIDAR for Water Vapor and Isotope HDO Observation in the Lower Troposphere Instrument Design. In Optical Sensors and Sensing Congress, paper LM4A.4; Optical Society of America, 2020; p LM4A.4, 2020.
- Matthias, V., Balis, D., Bösenberg, J., Eixmann, R., Iarlori, M., Komguem, L., Mattis, I., Papayannis, A., Pappalardo, G., Perrone, M. R., Wang, X.: Vertical Aerosol Distribution over Europe: Statistical Analysis of Raman Lidar Data from 10 European Aerosol Research Lidar Network (EARLINET) Stations. J. Geophys. Res. Atmospheres, 109 (D18). doi:10.1029/2004JD004638, 2004.
- Refaat, T. F., Singh, U. N., Yu, J., Petros, M., Ismail, S., Kavaya, M. J.; Davis, K. J.: Evaluation of an Airborne Triple-Pulsed
 2 μm IPDA Lidar for Simultaneous and Independent Atmospheric Water Vapor and Carbon Dioxide Measurements. Appl. Opt., 54 (6), 1387–1398. doi:10.1364/AO.54.001387, 2015.
 - Risi, C., Noone, D., Worden, J., Frankenberg, C., Stiller, G., Kiefer, M., Funke, B., Walker, K., Bernath, P., Schneider, M., Bony, S., Lee, J., Brown, D., Sturm, C.: Process-evaluation of tropospheric humidity simulated by general circulation models using water vapor isotopic observations: 2. Using isotopic diagnostics to understand the mid and upper tropospheric moist bias in the tropics and subtropics. Journal of Geophysical Research: Atmospheres, American Geophysical Union, 117 (5),
 - Singh, U. N., Refaat, T. F., Ismail, S., Davis, K. J., Kawa, S. R., Menzies, R. T., Petros, M.: Feasibility Study of a Space-Based High Pulse Energy 2 µm CO2 IPDA Lidar. Appl. Opt., 56 (23), 6531–6547. doi:10.1364/AO.56.006531, 2017.
- Spuler, S. M., Repasky, K. S., Morley, B., Moen, D., Hayman, M., Nehrir, A. R.: Field-Deployable Diode-Laser-Based
 Differential Absorption Lidar (DIAL) for Profiling Water Vapor. Atmospheric Meas. Tech., 8 (3), 1073–1087.
 doi:10.5194/amt-8-1073-2015, 2015.
 - Sodemann, H., Aemisegger, F., Pfahl, S., Bitter, M., Corsmeier, U., Feuerle, T., Graf, P., Hankers, R., Hsiao, G., Schulz, H., Wieser, A., and Wernli, H.: The stable isotopic composition of water vapour above Corsica during the HyMeX SOP1 campaign: insight into vertical mixing processes from lower-tropospheric survey flights, Atmos. Chem. Phys., 17, 6125–6151, doi: 10.5194/acp-17-6125-2017, 2017.
 - Stevens, B. and Bony, S.: Water in the Atmosphere. Phys. Today 2013, 66 (6), 29–34. doi:10.1063/PT.3, 2009.
 - Tomasi, C., Kokhanovsky, A. A., Lupi, A., Ritter, C., Smirnov, A., O'Neill, N. T., Stone, R. S., Holben, B. N., Nyeki, S., Wehrli, C., Stohl, A., Mazzola, M., Lanconelli, C., Vitale, V., Stebel, K., Aaltonen, V., de Leeuw, G., Rodriguez, E., Herber, A. B., Radionov, V. F., Zielinski, T., Petelski, T., Sakerin, S. M., Kabanov, D. M., Xue, Y., Mei, L., Istomina, L., Wagener,

- R., McArthur, B., Sobolewski, P. S., Kivi, R., Courcoux, Y., Larouche, P., Broccardo, S., Piketh, S. J.: Aerosol Remote Sensing in Polar Regions. Earth-Sci. Rev., 140, 108–157. doi:10.1016/j.earscirev.2014.11.001, 2015.
 - Wagner, G. A. and Plusquellic, D. F.: Multi-Frequency Differential Absorption LIDAR System for Remote Sensing of CO2 and H2O near 1.6 µm. Opt. Express, 26 (15), 19420–19434. doi:10.1364/OE.26.019420, 2018.
- Wandinger, U.: Raman Lidar. In Lidar: Range-Resolved Optical Remote Sensing of the Atmosphere; Weitkamp, C., Ed.;

 Springer Series in Optical Sciences; Springer: New York, NY; pp 241–271. doi:10.1007/0-387-25101-4_9, 2005.
 - Whiteman, D. N., Melfi, S. H., Ferrare, R. A.: Raman Lidar System for the Measurement of Water Vapor and Aerosols in the Earth's Atmosphere. Appl. Opt., 31 (16), 3068–3082. doi:10.1364/AO.31.003068, 1992.
 - Winker, D. M., Tackett, J. L., Getzewich, B. J., Liu, Z., Vaughan, M. A., Rogers, R. R.: The Global 3-D Distribution of Tropospheric Aerosols as Characterized by CALIOP. Atmospheric Chem. Phys., 13 (6), 3345–3361. doi:10.5194/acp-13-3345-2013, 2013.

625

Wirth, M., Fix, A., Mahnke, P., Schwarzer, H., Schrandt, F., Ehret, G.: The Airborne Multi-Wavelength Water Vapor Differential Absorption Lidar WALES: System Design and Performance. Appl. Phys. B, 96 (1), 201. doi:10.1007/s00340-009-3365-7, 2009.

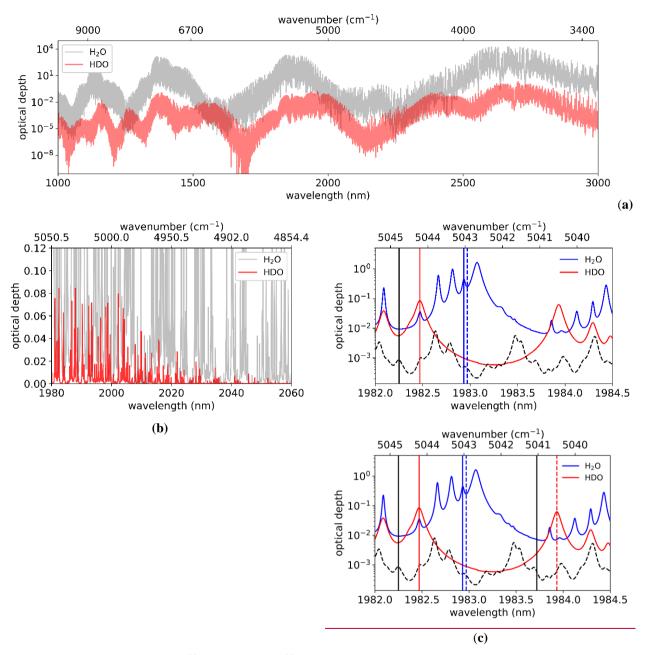


Figure 1: Optical depth over 1 km for $H_2^{16}O$ (H_2O) and $HD^{16}O$ (HDO) with uniform volume mixing ratios of 8400 ppmv and 2.6 ppmv, respectively, (relative humidity of 50% at 15°C). (a) Spectral overview between 1 μ m and 3 μ m; (b) Close-up window for wavelengths around 2 μ m with decreasing HDO absorption towards 2.05 μ m; (c) Spectral range of interest for simultaneous H_2O and HDO sounding. The dashed black line represents the total optical depth of other species (CO_2 , CH_4 , N_2O) with their typical atmospheric concentrations. The vertical vertical black line indicates lines indicate the position of the possible off-line

wavelengthwavelengths. On-line wavelengths are indicated for H₂O (vertical blue line for option 1 at 1982.93 nm, dashed <u>blue</u> line for option 2 at 1982.97 nm) and HDO (vertical red line <u>for option 1 at 1982.47 nm, dashed red line for option 2 at 1983.93 nm</u>). Spectra calculations are based on the HITRAN 2016 database assuming a temperature of 15°C and an atmospheric pressure of 1013.25 hPa.

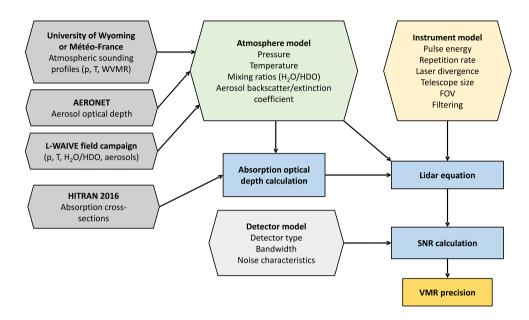


Figure 2: Block diagram of the DIAL simulator. Input models and databases in hexagons, principal calculations indicated by rectangles. p: pressure, T: temperature, WVMR: water vapor mixing ratio, FOV: telescope field of view. The signal-to-noise ratio (SNR) is used to calculate the statistical random error (precision) of the volume mixing ratio (VMR) of H₂O/HDO.

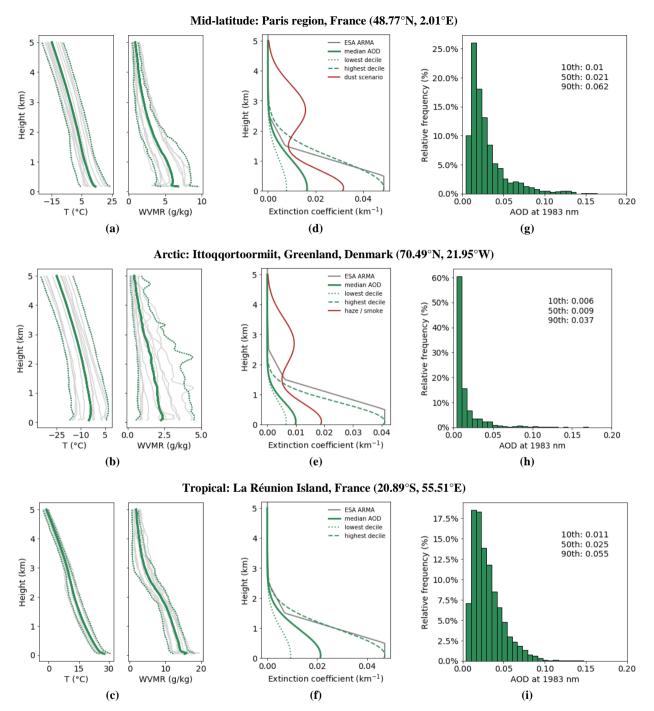
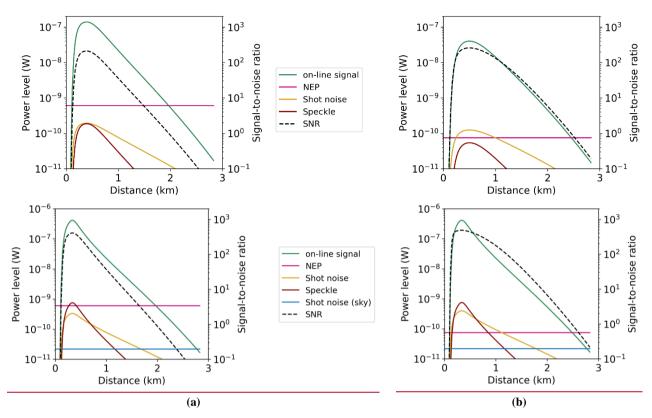


Figure 3: Atmosphere models: (a–c) Vertical sounding profiles of pressure, temperature, and water vapor mixing ratio (WVMR).

Grey lines indicate monthly averages, solid green line is the yearly average of 2019 (baseline profile). Dotted lines indicate profiles of lowest and highest monthly temperatures and WVMR; (e–f) Model profiles of aerosol extinction coefficient; (g–h) Distribution of the aerosol optical depth at 1983 nm for AERONET level 2.0 data of 2019.



650 Figure 4: Received power according to Equation (2) (solid green line) and power-equivalent levels of major noise contributions related to the H₂O on-line signal for a single 20 mJ pulse and resulting signal-to-noise ratio (SNR, dashed black line, right vertical axis) as function of lidar range: (a) InGaAs PIN detector; (b) low-noise HgCdTe APD.

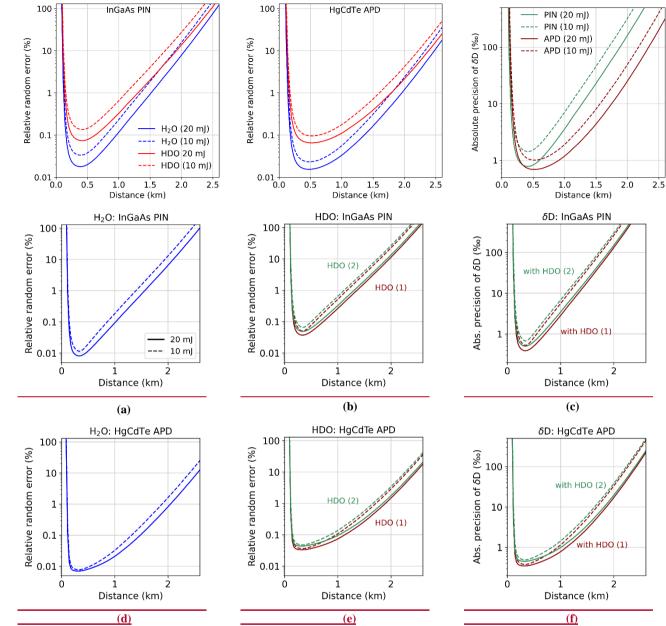
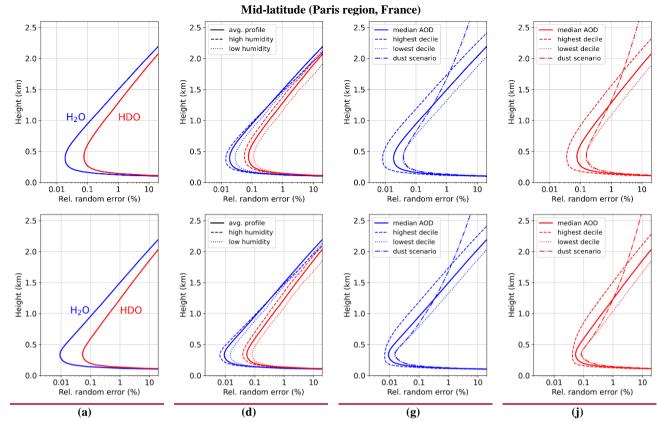
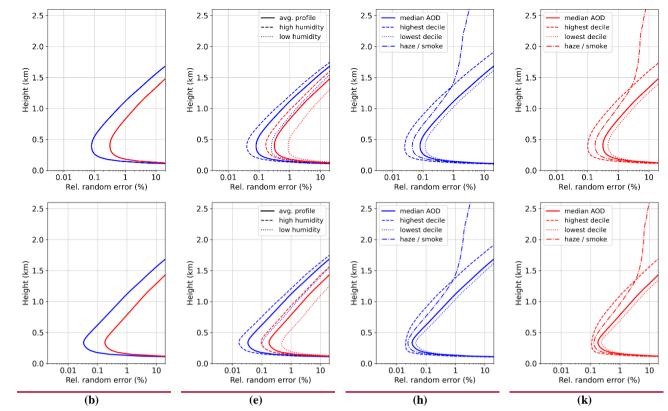


Figure 5: Expected relative random error on the volume mixing ratio of H₂O and HDO for different pulse energies (solid lines: 20 mJ, dashed lines: 10 mJ) and detectors: (a-c) InGaAs PIN detector; (bd-e) HgCdTe APD; (c/f) Corresponding absolute uncertainty (standard deviation) on δD as a function of distance from the lidar instrument. A detection bandwidth of 1 MHz is assumed and signal averaging time is 10 min.



Arctic (Ittoqqortoormiit, Greenland, Denmark)



Tropical (Réunion Island, France)

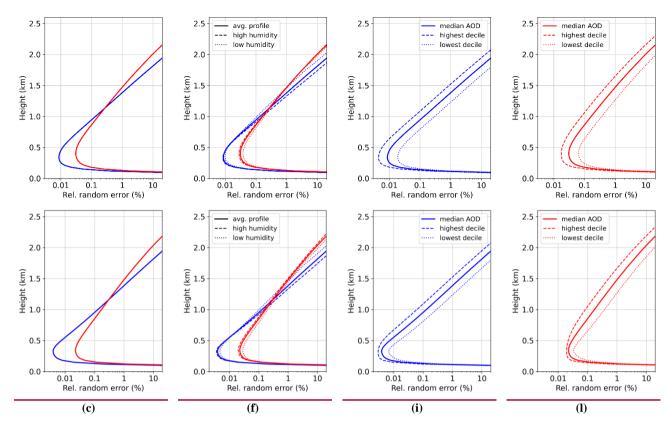
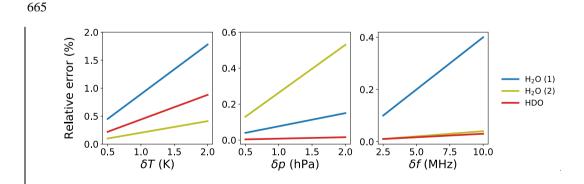


Figure 6: Sensitivity with respect to variability of atmospheric parameters: resulting statistical uncertainty for range-resolved DIAL measurement of H₂O (blue, only wavelength option 1 at 1982.93 nm) and HDO (red, option 2 at 1983.93 nm). Simulation parameters: 20 mJ pulse energy, 1 MHz bandwidth, 10 min integration time, InGaAs PIN detector. (a-c) Reference model based on average columns of pressure, temperature, and humidity. Aerosol baseline profile using median AOD assumed; (d-f) Sensitivity to water vapor variability; (g-i) Sensitivity to different aerosol profiles (H₂O); (j-l) Sensitivity to different aerosol profiles (HDO).



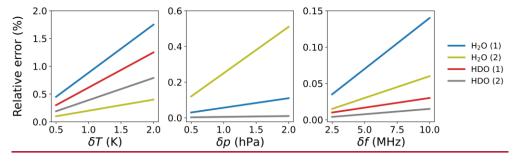


Figure 7: Maximal relative error in the VMR retrieval (over 3 km range, mid-latitude baseline) due to uncertainties in the profiles of temperature (δT) and atmospheric pressure (δp) as well as transmitter on- and off-line wavelength (δf). (1) and (2) stand for the two possible on-line wavelength options for measuring H₂O.

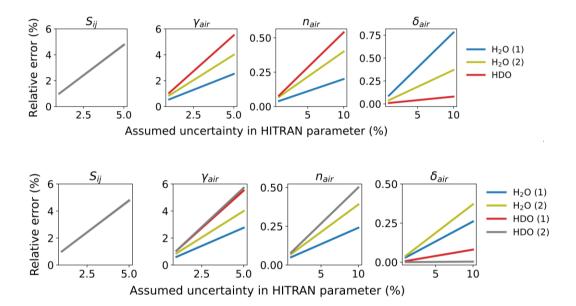


Figure 8: Maximal relative error in the VMR retrieval (over 3 km range, mid-latitude baseline) due to uncertainties in HITRAN parameters. S_{ij} : line intensity; γ_{air} : air-broadened half width; n_{air} : coefficient of the temperature dependence of γ_{air} ; δ_{air} : pressure shift.

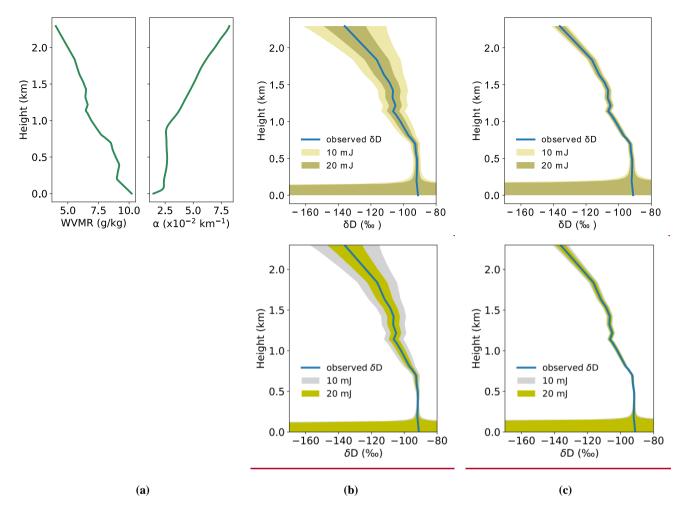


Figure 9: (a) Experimental profiles of water vapor mixing ratio (WVMR) and aerosol extinction coefficient (α) obtained from the L-WAIVE field campaign. Expected precision in the isotopic ratio in terms of δD for the InGaAs PIN photodetector (b) and the low-noise HgCdTe avalanche photodiode detector (c). Shaded areas indicate the absolute uncertainty based on random noise in terms of standard deviation for laser energies of 10 mJ and 20 mJ. High uncertainty in the first 200 m is due to the low signal caused by overlap function increasing from zero to unity (see Eq. (2)). Calculations based on a measurement bandwidth of 1 MHz (150 m spatial resolution) and an integration time of 10 min.

Table 1: Spectroscopic parameters for selected absorption lines

	<u>v</u>	<u>λ</u>	<u>S</u>	<u>E''</u>	<u> Yair</u>	<u>n_{air}</u>
<u>H</u> ₂ ¹⁶ O (1)	5043.0476	1982.928	2.17×10^{-24}	920.21	0.0367	0.49
<u>HD¹⁶O (1)</u>	5044.2277	<u>1982.464</u>	$\underline{1.17\times10^{\text{-}24}}$	91.33	0.1036	<u>0.71</u>
HD ¹⁶ O (2)	5040.4937	<u>1983.933</u>	9.38×10^{-25}	<u>116.46</u>	0.1003	<u>0.71</u>
H ₂ ¹⁶ O at HD ¹⁶ O (1)	5044.2300	<u>1982.463</u>	$\underline{2.29\times10^{\text{-}25}}$	<u>2756.42</u>	0.0456	0.37

 ν (cm⁻¹): wavenumber; λ (nm): vacuum wavelength; S (cm⁻¹/molecule cm⁻²): line intensity at 296 K; E" (cm⁻¹): lower-state energy; γ_{air} (cm⁻¹ atm⁻¹): air-broadened Lorentzian half-width at half-maximum (HWHM) at 1 atm and 296 K; γ_{air} temperature exponent for γ_{air}

Table 2: DIAL instrument parameters

Transm	iitter	Receiver					
Energy	10–20 mJ		i)	ii)			
Pulse duration	10 ns	Telescope aperture	40 cm	40 cm			
Repetition rate	150 Hz	Detector type	InGaAs PIN	HgCdTe APD			
$\lambda_{\text{on}} (H_2^{16}O) (1)$	1982.93 nm	Detector diameter	300 μm	180 µm			
$\lambda_{on} (H_2^{16}O)$ (2)	1982.97 nm	Field of view (FOV)	630 µrad	380 <u>630</u> μrad			
$\lambda_{\text{on}} \left(HD^{16}O_{\underline{(1)}} \right)$	1982.47 nm	NEP	600 fW Hz ^{-1/2}	75 fW Hz ^{-1/2}			
$\lambda_{\text{off}} \underline{\lambda_{\text{on}} \text{ HD}^{16} \text{O } (2)}$	1982.25 <u>1983.93</u> nm	Bandwidth	1 MHz	1 MHz			
Divergence	270 µrad		Responsivity: 1.2 AW ⁻¹	Quantum efficiency: 0.8			
				Excess noise factor: 1.2			

Table 23: Systematic errors for mid-latitude, arctic, and tropical atmospheric models (maximal error over 3 km range). (1) and (2) denote the two wavelength options for H₂O measurement and HDO measurements.

		Maximal relative error ε_s (%)											
Parameter	Assumed	<u>Mid-latitude</u>			<u>Arctic</u>			<u>Tropic</u>					
1 di diffetti	uncertainty 1	$\underline{\mathbf{H}_{2}\mathbf{O}}$	$\underline{\mathbf{H}_{2}\mathbf{O}}$	HDO	HDO	$\underline{\mathbf{H_{2}O}}$	H_2O	HDO	HDO	$\underline{\mathbf{H_2O}}$	$\frac{\mathbf{H_2O}}{2}$	HDO	HDO
		<u>(1)</u>	<u>(2)</u>	<u>(1)</u>	<u>(2)</u>	<u>(1)</u>	(2)	<u>(1)</u>	<u>(2)</u>	<u>(1)</u>	<u>(2)</u>	<u>(1)</u>	(2)
Temperature	<u>±1 K</u>	0.85	0.21	0.61	0.39	0.95	0.27	0.19	0.40	0.79	0.15	1.12	0.37
Pressure	<u>±1 hPa</u>	0.08	0.27	< <u>0.01</u>	<u><0.01</u>	0.06	0.26	<0.01	< <u>0.01</u>	0.05	0.25	< 0.01	<u><0.01</u>
On/off wavelength	<u>5 MHz</u>	0.07	0.03	0.02	< 0.01	0.07	0.04	0.02	<u><0.01</u>	0.07	0.03	0.03	<u><0.01</u>
VMR of H ₂ O bias	<u>2% ²</u>	Ξ	Ξ	<u>0.52</u>	Ξ	Ξ	Ξ	<u>0.28</u>	Ξ	Ξ	Ξ	0.83	Ξ
HITRAN 2016 parameters													
Line intensity	<u>1%</u>	<u>1.00</u>	1.00	1.01	1.00	1.00	1.00	1.01	0.99	1.00	1.00	1.01	0.99
Air-broadened width γ_{air}	1%	0.60	0.86	1.05	1.06	0.59	0.87	1.07	1.06	0.57	0.83	1.05	1.05
<u>T-exponent of γ_{air}</u>	<u>5%</u>	0.14	0.20	0.33	0.33	0.21	0.31	0.49	0.49	0.07	0.1	0.16	<u>0.16</u>
Pressure shift	<u>5%</u>	<u>0.15</u>	<u>0.19</u>	0.03	< 0.01	<u>0.15</u>	<u>0.16</u>	0.04	<u><0.01</u>	0.13	<u>0.21</u>	0.03	<0.01
Combined (geometric sum)		1.48	1.39	<u>1.69</u>	1.54	1.53	1.42	<u>1.59</u>	<u>1.58</u>	1.41	<u>1.35</u>	2.02	<u>1.50</u>

¹ relative uncertainty if stated in %

² conservative estimate of combined systematic error for H₂O measurement

715

725

730

Author's response to reviewer comments

We thank both reviewers for their helpful comments and suggestions.

Reviewer 1:

This is an interesting paper on the predicted performance of a theoretical DIAL system to remotely measure water vapor isotopes. The authors have identified a region in the infrared around 1983 nm that shows potential for measurement of both H(16)OH and HD(16)O. Overall this is a very thorough analysis and is well done.

710 General comments:

1) To have a scientific impact, is the desired/required goal to achieve a <1% relative error? Is a specific water vapor isotope abundance precision in permil required?

The better than 1% relative precision for H_2O/HDO mixing ratios can be understood as required limit in order to still reach precisions of <10 permil in the isotopic abundance. Since the isotopic abundance can vary by a few tens of permil, precisions greater than that value would be of not much use for observations in a scientific context.

Modifications made: Page 1, line 28 in abstract: Specifies why it is important to have relative precision in H₂O/HDO of better than 1%

720 2) The laser development needed to bring the proposed instrument to reality will be far from trivial. The modeling effort assumes that the transmitter will have perfect qualities required for DIAL (such as spectral purity) which is a significant assumption. To be fair, the conclusions could be more clear to say that the predicted performance is a best-case scenario because of this assumption.

Indeed, the calculations represent rather optimistic best-case scenarios.

Modifications made: Page 16, line 487 et seq: Emphasis that calculations are best-case scenario and do not take into account important aspects which are challenging in practice (such as perfect spectral purity).

3) While reading through, the biggest question I had was if the temperature sensitivity of the line strength was included in the model. I think the answer is yes, but it wasn't very clear in the text.

Yes, temperature dependence of the line strengths is included in the model. We added supplementary equations, tables as suggested in the points below.

3.1) Line selection criteria for water vapor DIAL requires choosing lines that (1) avoid interferences, (2) have the appropriate line strength (will provide the optimal optical depth), and (3) are insensitive to temperature as highlighted in Browell et al. 1991 (Browell, E. V., S. Ismail, and B. E. Grossman, 1991: Temperature sensitivity of differential absorption lidar measurements of water vapor in the 720 nm region. Appl. Opt., 30, 1517–1524). Could this line selection consideration be more clearly articulated in section 2.1?

More emphasis was put on the fact that the chosen absorption lines are not temperature independent as ideally preferred for DIAL. This requires the accurate knowledge of the temperature profile, which has to be provided by auxiliary measurements (for example a Raman lidar next to the DIAL).

Modifications made: page 4, lines 125-137

735

740

750

755

760

3.2) In the sections around Equation 5 would it help to include, or at least mention the temperature dependence in the absorption cross-sections?

Equations for the Voigt line shape as well as the temperature dependence of the line strength have been added.

Modifications made: page 6, lines 174 et seq: Equations 6 and 7 added

- 3.3) The HD(16)O line at 1982.47 nm has a reasonably temperature-insensitive ground state energy of 91 cm^-1 (something closer to 250 cm^-1 would be ideal), but the interference H(16)OH line, directly underneath it, has a ground state energy of 2756 cm^-1. Is not some discussion warranted about how this creates a high sensitivity to temperature? Using the equations in Browell et al. (1991) it appears that the uncertainty in the number density would be up to 5% per K on line center. Unfortunately, the optical depth when tuned to the line center of that HD(16)O line is already less than optimal for DIAL. But should there be an online second option evaluated, moving to the weaker line at 1983.93nm to avoid this even if constraining the measurements to regions with high water vapor concentrations? It is true that the initially proposed HDO line at 1982.47 nm violates a number of basic DIAL line selection rules. As shown in our error budget, measuring HDO at this line is only possible if 1) the temperature profile is well known (applies to all selected lines, practical implementation via Raman lidar for example) and 2) the distribution of the main isotope is known to correct for the interference bias. In theory this is all possible, but in practice this procedure adds further uncertainties, indeed. As suggested, we included the HDO line at 1983.93 nm into our analysis. Despite slightly reduced optical depth (thus higher random error, but could be offset by longer averaging), this should
- 765 *Modifications made:*

page 4, line 124: Introduction of the second HDO option at 1983.93 nm;

definitely be the preferred option for HDO.

page 11, line 310: HDO line at 1983.93 nm leads only to slight increase in expected random error Figure 5 (page 26). Figure has been updated/modified substantially to include second option for HDO. Figure has

been split into upper and lower panel for the two different detectors.

770

775

780

785

790

795

800

3.4) The online for H(16)OH at 1982.93 nm has a ground state of 920 cm^-1 which is still rather large compared to a typical water vapor DIAL system. Would it be helpful if the ground state energies were listed somewhere to help the reader understand that the model is taking these fundamental factors into account?

Table 1 was added in section 2.1 at page 4 listing important spectroscopic parameters for the investigated absorption lines.

4) The larger than typical DIAL errors/sensitivity to atmospheric temperature seems to be sidestepped somewhat by simply constraining the model temperature uncertainty to +/- 0.5°K. Although there is a sentence in the conclusions about this, it is a significant issue and warrants more discussion. Could the authors suggest how this will be done in practice? For example, if reanalysis data will be needed to reach these levels of certainty, the DIAL measurement would not be useful in real time. Are they expecting another instrument like a Raman lidar to provide this information?

As already mentioned, temperature data has to be provided by auxiliary measurements (for example by radio sounding or a Raman lidar). For a future field campaign, it is intended to employ a Raman lidar next to the here presented DIAL. So indeed, real time measurements would not be possible. Please note that we changed the assumed temperature uncertainty for the error budget in Table 3 from +/-0.5 to +/-1K which is an accuracy more realistically achievable in the tempeture measurement by Raman lidar.

Modifications made:

the aspect of having to have an auxiliary temperature measurement was added at multiple locations: p. 4 lines 129 et seq, p. 13 lines 362-363, in the conclusion, p. 16 lines 475 et seq.

5) Was the model using a yearly or seasonal mid-latitude average (high and low)? It was unclear if the proposed instrument is expected to perform well in mid-latitude winter conditions.

The model uses yearly averages as a baseline. Since in mid-latitude winter conditions less water vapor is present in the atmosphere, the instrument is expected to perform less well than in summer conditions. The main effect here is clearly the water vapor content and thus the difference in differential absorption optical depth. When conducting the calculations with fixed water vapor profile but lowest and highest monthly temperature profiles (green dashed lines in Fig. 3 a-c), no significant difference in the achievable precision was found.

6) Was the performance model limited to nighttime only? Was there any solar background modeled?

We used a constant solar background irradiance of 1 $W/m^2/\mu m/sr$ and an optical filter bandwidth of 50 nm for our calculations. This information was missing before and was added to the manuscript. Shot noise due to solar background added to the plots in Fig. 4.

805 *Modifications made:*

page 8, line 229: background irradiance and filter bandwidth specified

Figure 4: Shot noise due to solar background has been added to the graphs

- 7) What is the expected lowest useful range of the proposed instrument? The plots in Figure 6 indicate measurement would be limited to approximately >150m above ground level (if 1% error is the goal). However, based on the curve shapes, the full overlap is not achieved until above 500m which could push the minimum range upward. As I'm sure the authors know, very slight differences in online and offline overlap or pointing can result in large systematic errors when pushing too far into the incomplete overlap region. Could these limitations be discussed a bit more clearly so the science community has realistic expectations of the proposed DIAL instrument?
- Indeed, the plots in Figs. 5 and 6 suggest that high measurement performance can be already achieved onwards from 150 m or so. We added the aspect of incomplete overlap leading to biases due to laser beam pointing. Please note that all curve shapes in Figs. 4, 5, 6 and 9 have slightly changed due to an error we identified in the submitted version concerning the overlap. Now with the correct parameters used, the lidar instrument reaches full overlap between 450-500 m. In the submitted version it was closer to 800 m due to an error. As suggested by the reviewer, we included a sentence stating the lowest useful range to be around 500 m.

Modifications made:

Figs. 4, 5, 6 and 9 have been updated due to an error in the overlap function of the submitted version page 11, lines 318 et seq: specification of the lowest useful range due to incomplete overlap at ranges <0.5 km

Specific comments:

825

830

1) line 46. Does it have to be a high-power laser transmitter?

High-power laser transmitter was simply the wrong word. It is the laser energy which has to be sufficient (mJ level) for range-resolved DIAL measurements.

Modifications made: now page 2, line 47: word "high-power" has been deleted

2) line 101. "... with and out of a gas absorption feature.." is unclear

Modifications made: page 4, line 101...formulated differently

- 3) line 155 "two wavelength in (lambda_on) and out of (lambda_off) coincidence" is a bit awkward in English *Modifications made:* page 6, line 158...formulated differently
 - 4) line 157, the lasers also need to be sufficiently close in wavelength (not just time)

Modifications made: page 6, line 163..."close in wavelength and in time"

840

850

5) line 166, Why use a mixing ratio? A DIAL measures the number density of the absorbing molecule. When converting to a mixing ratio one has to assume a pressure and thereby increase the uncertainty.

For the scientific context, mixing ratios are required.

- 845 6) In section 3.4 the precision estimate is referring to figure 9, yet the text says figure 8 (in multiple locations) Wrong figure references have been corrected.
 - 7) The x-axis labels in figure 9 seem to be incorrect? The text at line 397 says "The expected absolute precision for this configuration is well below 5% within the first 1.5 km and reaches 10% at a range height of 2.3 km" but the labels don't match so it makes it hard to evaluate what is going on.

The idea of Figure 9 b/c is to visualize the hypothetical precision in the form of an error band (shaded areas) around the dD profile obtained in situ during the field campaign. It is true that it is hardly possible to read the exact precision values from the figure. That's why they have been given in a sentence afterwards. We tried to be more clear in the corrected version (p.15 lines 428 et seq)

855 8) line 415 "Indeed, HDO displays sufficiently high ab-sorption [sic] lines in this range" Suggest adding a qualifier here such as "for measuring the lower 1km of the atmosphere with better than 1% relative error in the mid-latitudes and regions with higher water vapor concentrations."

The suggested qualifier was added to be more concise (p. 15, line 455)

Reviewer 2:

Hamperl et al. presents a theoretical analysis and performance evaluation of a DIAL system to measure vertical profiles of water vapor H2(16)O and its isotopologue HD(16)O. The paper is well written and detailed. I recommend it to be published after the following comments are addressed:

865 General comments:

1) I understand the authors decided to exclude the laser linewidth from the analysis, nevertheless I think it would be good if they at least provide a first order estimate of its impact.

An error estimation was provided by introducing the concept of effective absorption cross section for taking into account the finite bandwidth of the laser transmitter. The additional error is found to be in the order of 0.1% for the narrowest H2O line.

Modifications made: page 13, line 385 et seq: introduction of effective cross sections to calculate numerically the bias due to a laser line width of 50 MHz

- 2) Is the 'efficiency' of the receiver optics (Tr in Eq. 2) assumed to be 1? If so, is that a reasonable assumption?
- The efficiency is assumed to be 0.5. We added this specification (p. 6, line 159).
 - 3) The authors include the effect of solar background in Eq. 10, but there is no further discussion regarding its impact on the instrument performance (and the optical filter bandwidth is not included in Table 1).

We used a constant solar background irradiance of $1 \text{ W/m}^2/\mu\text{ m/sr}$ and an optical filter bandwidth of 50 nm for our calculations.

This information was missing before and was added to the manuscript.

Modifications made: page 8, line 229: background irradiance and filter bandwidth specified

4) As the previous reviewer pointed out, it would be nice to have a more detailed analysis of temperature sensitivity of the line strengths and its impact of the overall retrieval uncertainty.

As suggested, more details on the temperature dependence of the line strengths have been added to show that this aspect is taken into account by the calculations.

please see Reviewer 1, point 3)

5) Have you considered exploiting an absorption line with strong temperature dependence to try to retrieve temperature simultaneously? I'm unsure if a reasonable uncertainty can be achieved, but it might be worth exploring it.

We have so far not considered this idea. In the case of a true simultaneous H_2O + temperature measurement, a multiwavelength switching scheme would have to be realized, which is possible but technically more challenging. And since in that case the repetition rate for each wavelength decreases, the SNR will suffer for unchanged integration time. As shown in the paper, even with only a two-wavelength switching there are conditions under which the instrument performs rather poorly. It is an interesting idea to study by further calculations, but in practice we are not there yet...

895

885

Additional remarks

905

915

- 1) As already mentioned in the response to reviewer 1, the calculations of the originally submitted version contained an error in the overlap function. This has been corrected now and affects Figs. 4, 5, 6 and 9. Updated figures are underlined in red.
- 2) Please note that the considered field of view for the detector option 2 was changed (see Table 2) to the same value as for detector 1. (p. 7, lines 205 et seq). The FOV angle stated in the original manuscript would be too small in practice and heavily degrade the overlap function.
- 3) Table 3 has been updated. The assumed temperature uncertainty was changed from 0.5 to 1K. The assumed H2O bias for HDO (1) was changed to relative uncertainty of 2% for a more conservative estimate. The wavelength errors for H2O (1) were erroneous and have been corrected.
 - 4) Figure 1c was updated to include second option for HDO at 1983.93 nm.
 - 5) Figures 7 and 8 were updated to include second option for HDO at 1983.93 nm.
 - 6) Line 214: The number of laser shots per wavelength has to be 45.000, not 30.000.