



1 Mitigation of bias sources for atmospheric temperature and

- 2 humidity in the mobile Weather & Aerosol Raman Lidar
- 3 (WALI)

4 Julien Totems, Patrick Chazette and Alexandre Baron

5 [1]{Laboratoire des Sciences du Climat et de l'Environnement, CEA, Gif-sur-Yvette, France}

6 Correspondence to: J. Totems (julien.totems@cea.fr)

7 Abstract

8 Lidars using vibrational and rotational Raman scattering to continuously monitor both the water 9 vapor and temperature profiles in the low and middle troposphere offer enticing perspectives 10 for applications in weather prediction and studies of aerosol/cloud/water vapor interactions by 11 deriving simultaneously relative humidity and atmospheric optical properties. Several heavy 12 systems exist in European laboratories but only recently have they been downsized and 13 ruggedized for deployment in the field. In this paper, we describe in detail the technical choices 14 made during the design and calibration of the new Raman channels for the mobile Weather and 15 Aerosol Lidar (WALI), going over the important sources of bias and uncertainty on the water 16 vapor & temperature profiles stemming from the different optical elements of the instrument. 17 For the first time, the impacts of interference filters and non-common-path differences between 18 Raman channels, and their mitigation, are particularly investigated, using horizontal shots in a 19 homogenous atmosphere. For temperature, the magnitude of the highlighted biases can be much 20 larger than the targeted absolute accuracy of 1°C defined by the WMO. Measurement errors are 21 quantified using simulations and a number of radiosoundings launched close to the laboratory.

22 1 Introduction

Atmospheric temperature and humidity in the low atmosphere are together essential to comprehend weather phenomena and their evolution in a changing climate. Through the effect of relative humidity on aerosol hygroscopicity and cloud formation, they also influence the radiative balance of the Earth, generating the largest uncertainties in climate projections (IPCC, 2013). For both weather and climate prediction, observation means have evolved tremendously, notably with satellite retrievals of moisture and temperature routinely assimilated in numerical models. Yet remote-sensing techniques from spaceborne missions have difficulties probing the





30 lower troposphere below 2-3 km in altitude, and have vertical resolutions that are too low, 31 greater than 1 km in the lower troposphere (e.g. Prunet et al., 1998; Crevoisier et al., 2014). 32 They are thus unable to resolve temperature inversions and thin dry/humid air masses (e.g. 33 Chazette et al., 2014; Hammann et al., 2015; Totems et al., 2019). Providing complementary 34 profiles of the important thermodynamic variables in the first kilometres of the atmosphere, 35 where most of the water vapour and temperature vertical variability is confined, is of paramount 36 importance for both weather forecast and reducing aerosol-induced uncertainty on climate 37 models (Wulfmeyer et al., 2015).

38 Given their capacity for continuous, well-resolved and precise temperature measurements in 39 the lower troposphere, Vibrational Raman (VR) and Rotational Raman (RR) lidars have 40 emerged as adequate tools in this endeavour. Water vapor profilers are now well-established 41 (from Whiteman et al., 1992 to e.g. Dinoev et al., 2013), whereas temperature profilers have 42 recently become more widespread and powerful (from Cooney, 1972 and Vaughan et al., 1993 to e.g. Weng et al., 2018 or Martucci et al., 2021). Without tackling turbulence-scale resolution 43 44 which is the prerogative of heavier systems like the Raman lidars of the University of 45 Hohenheim (Behrendt et al., 2015), the University of Basilicata (Di Girolamo et al., 2017) or 46 ARTHUS (Atmospheric Raman Temperature and Humidity Sounder, Lange et al., 2019), there 47 is a need for field-deployable instruments capable of fulfilling the breakthrough requirements 48 set by the World Meteorological Organization in terms of accuracy on atmospheric temperature 49 and humidity in the low troposphere (WMO, 2017). Lidar profiles have proven beneficial for 50 both numerical weather prediction (NWP) models (e.g. Adam et al., 2016; Fourrié et al., 2019), 51 the study of dynamic processes in the planetary boundary layer (PBL) (e.g. Behrendt et al., 52 2015) or interactions between water vapor and aerosols (e.g. Navas-Guzmán et al., 2019). But 53 to obtain the absolute accuracies demanded here, especially that of 1°C or less on temperature, 54 the required accuracy on the lidar channel ratios and their calibration is extremely stringent, 55 and the sources of bias seldom discussed in the literature (Behrendt and Reichardt, 2000; 56 Simeonov et al., 1999; Whiteman et al., 2012).

Within the European lidar landscape, WALI (Weather and Aeorosol Lidar) is a seasoned mobile Rayleigh-Mie-Raman system, eyesafe at 355 nm, first deployed during the HyMeX international field campaign and subsequently ChArMEx and PARCS, for aerosol and water vapor profiling (resp. Hydrological cycle in the Mediterranean eXperiment, Chemistry and Aerosol in the Mediterranean Experiment, Pollution in the Arctic System; Chazette et al., 2014b, 2018; Totems et al., 2019; Totems and Chazette, 2016). In its latest evolution, the VR channels have been replaced by a Newton reflector and a polychromator also including RR





channels for temperature profiling. On this occasion, we have established that biases due to various sources, in particular from the dependency of spectral filtering on the angle of incidence, detector non-uniformities and other non-common-path differences between Raman channels, may be several times greater than the requirements if left unchecked. Correctible as they are by measuring the ratios of overlap factors on the individual channels, these effects are not reported in the literature of lidar temperature measurements. However, they were bound to appear given the physical characteristics of the systems mentioned hereabove.

The aims of this paper are: i) to compile for the first time the sources of bias that must be considered and mitigated when using a Raman lidar to profile atmospheric temperature and humidity, ii) to validate WALI as a dependable profiler deployable for field campaigns, satisfying the requirements set by the WMO.

The theory of the Raman lidar retrieval of the atmospheric temperature and WVMR, the error budget on these parameters, and the known sources of bias are recalled in section 2, as well as the principle and limitations of the overlap measurement method. In section 3, after summarizing the characteristics of WALI, we propose a sequential review of the components of the lidar chain, characterizing and mitigating the error sources. The results of a calibration and qualification experiment using radiosondes follow in section 4. A conclusion and outlooks are presented in section 5.

82 2 Theoretical considerations

83 2.1 Raman lidar retrieval of humidity and temperature

We will introduce notations by briefly recalling the theory of the retrieval of water vapor content
and temperature by the Raman lidar technique; the complete theory has been extensively
derived before, by Whiteman et al. (1992) and Behrendt (2005) respectively, among others.

The vertical profiles of water vapor mixing ratio (WVMR) r_{H2O} and temperature *T* are calculated from the ratios of the H₂O / N₂-vibrational Raman (VR) channels and the RR2 (high-J number) / RR1 (low-J number) rotational Raman (RR) channels, respectively:

$$R(z) = \frac{S_{H_2O}(z)}{S_{N_2}(z)}$$
(1)

$$Q(z) = \frac{S_{RR2}(z)}{S_{RR1}(z)}$$
(2)





Signals $S_j(z)$ of Raman channels *j* have all been previously averaged over the required altitude and time to improve the signal to noise ratio (SNR), and corrected for i) electronic baseline variations by subtracting a baseline recorded every few profiles with detector (photomultiplier tube, PMT) gain set to zero, ii) the sky background mean value assessed on pre-trigger or postsignal samples, iii) PMT gain variations (allowed on the VR channels to optimize daytime dynamic range, eg. Chazette et al. (2014b)), iv) known leakage of the elastic return in the RR filters (Behrendt and Reichardt, 2000). $S_j(z)$ are thus expressed as:

$$S_{j}(z) = \frac{1}{G_{j}(U_{j})} \left(S_{j,raw}(z) - \hat{L}_{j}(z) - \hat{B}_{j} \right) - \hat{\varepsilon}_{j} S_{elas}(z)$$
(3)

97 where G_j is the channel gain controlled by PMT voltage U_j , $S_{j,raw}$ is the raw lidar signal, \hat{L}_j is 98 the estimated baseline, \hat{B}_j is the estimated sky background parasitic signal, $\hat{\varepsilon}_j$ is the estimated 99 residual transmittance of the emitted laser wavelength through the interference filter (IF) of 100 Raman channel *j* compared to the elastic channel, and S_{elas} is the elastic signal.

Both R and Q must then also be corrected from the difference of atmospheric transmission between the two Raman channels and the ratio of overlap factors:

$$R'(z) = \frac{\exp(\Delta \tau(z))}{\overline{OR_R}(z)} R(z)$$
(4)

$$Q'(z) = \frac{1}{\widehat{OR_Q}(z)}Q(z)$$
(5)

103 where $\Delta \tau(z)$ is the difference of optical thickness from the lidar until range z observed between 104 the wavelengths of the two VR channels, and where $\widehat{OR_R}(z)$ and $\widehat{OR_Q}(z)$ are the estimated 105 ratios of the overlap factors of the two VR / RR channels respectively (expressed in section 106 2.4). With an emitted wavelength at 355 nm, $\Delta \tau(z)$ between 387 and 407 nm seldom produces 107 deviations above 5%, and can be efficiently estimated using an average atmospheric density 108 profile for molecular optical thickness and the N₂-Raman channel itself for aerosol optical 109 thickness (e.g. Whiteman, 2003).

110 The WVMR is simply proportional to the VR scattering ratio between H₂O and N₂, since the 111 latter can be considered with a constant mixing ratio in the troposphere and stratosphere. The 112 temperature is retrieved from the more complex dependency of the RR scattering cross sections 113 between the two channels RR1 and RR2. The respective estimates \hat{r}_{H_2O} and \hat{T} are obtained, 114 after calibration, by:





$$\hat{r}_{H_20}(z) = \hat{K}R'(z)$$
 (6)

$$\hat{T}(z) = \hat{f}^{-1}(Q'(z))$$
(7)

where \hat{K} is the estimate of the calibration coefficient for WVMR combining all instrumental constants. Calibration function \hat{f} is the estimate of the temperature dependency of the ratio of RR cross-sections. It takes into account the instrumental constants of the two RR channels. We take the model previously selected for operational purposes by Behrendt (2005):

$$Q' = f(T) = \exp\left(a + \frac{b}{T} + \frac{c}{T^2}\right)$$
(8)

119 with *a*, *b*, *c* the coefficients of a polynomial regression of $\ln(Q')$ as a function of 1/T. \hat{K} and \hat{f} 120 are obtained by confronting lidar profiles of *R*' and *Q*' with collocated in-situ measurements of 121 r_{H2O} and *T* (e.g. from a radiosounding), aiming for a wide range of values for a better constraint 122 on the calibration.

123 2.2 Simple error budget

124 In this section, we will make a first assessment of the acceptable error on R and Q starting from 125 the accuracy requirements for WVMR and temperature profiles, which ensue from each 126 scientific need, as compiled by Wulfmeyer et al. (2015) for key applications. Monitoring, 127 verification (e.g. model qualification or calibration/validation of satellites) and data 128 assimilation purposes can be adequately addressed by a profiler capable of i) <5% noise error and <2-5% bias for water vapor, ii) <1°C noise error and <0.2-0.5°C bias for temperature. In a 129 simple error budget, we can use requirements of $\left(\frac{\Delta r_{H_2O}}{r_{H_2O}}\right)_{max} = 5\%$ for WVMR, and 130 $\Delta T_{max} = 1^{\circ}$ C for temperature, to give a first idea of the different expectations for the 131 132 performance of a VR/RR lidar.

Eqs. (4-8) allow to derive constraints on the acceptable relative error on the corrected lidar observables R' and Q', for either random noise or bias, as:

$$\left(\frac{\Delta R'}{R'}\right)_{max} = \left(\frac{\Delta r_{H_2O}}{r_{H_2O}}\right)_{max} \tag{9}$$

$$\left(\frac{\Delta Q'}{Q'}\right)_{max} = \frac{\frac{\mathrm{d}Q'}{\mathrm{d}T}}{Q'} \Delta T_{max}$$
(10)





- The relative error on *R* is equal to the constraint on WVMR, i.e. 5%. An assessment of the relative error on *Q* is performed considering the RR filter parameters given in Table 2 (section 3) to yield the following numerical application: around $T_0 = 0^{\circ}$ C, $Q'(T_0) = 0.44$ and $dQ'/dT(T_0)$ $= +0.35/100^{\circ}$ C, so that: $\left(\frac{\Delta Q'}{Q'}\right)_{max} = 0.79\% \Delta T_{max}(^{\circ}$ C).
- 139 Table 1. Summary of accuracy requirements from Wulfmeyer et al. (2015) and corresponding
- 140 constraints on ratios R' and Q'. Resulting errors on relative humidity RH at 0° C and 50° RH.

Parameter	Random error	Systematic error (bias)
r _{H2O}	<5% relative	<2-5% relative
Т	<1°C	<0.2-0.5°C
R'	<5% i.e. <i>SNR</i> > 20	<2-5%
Q'	<0.8% at 0°C i.e. <i>SNR</i> > 125	<0.12-0.4% at 0°C
RH	4.3%RH	1.2-2.9%RH
	at $T = 0^{\circ}$ C, $RH = 50\%$	at $T = 0^{\circ}$ C, $RH = 50\%$

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The results, summarized in Table 1, have very important implications. Typically, Q' must be 6 to 10 times more accurate than R' to deliver meaningful results in terms of temperature. Raman cross-sections being larger for the RR channels than for the H₂O VR channel, the main difficulties shift from constraints linked to signal-to-noise ratio (SNR) to also encompass strong constraints linked to instrumental biases. SNR as used in Table 1 is defined on R and Q at the final resolution, and is calculated from the individual signal variances (including laser & skybackground photon noise, detection noise), as:

$$SNR_{R} = \left(\frac{\operatorname{var}(S_{N_{2}})}{\langle S_{N_{2}} \rangle^{2}} + \frac{\operatorname{var}(S_{H_{2}O})}{\langle S_{H_{2}O} \rangle^{2}}\right)^{-\frac{1}{2}}$$
(11)

$$SNR_{Q} = \left(\frac{\operatorname{var}(S_{RR1})}{(S_{RR1})^{2}} + \frac{\operatorname{var}(S_{RR2})}{(S_{RR2})^{2}}\right)^{-\frac{1}{2}}$$
(12)

SNR_R, typically limited by the H₂O channel, must be above ~20 and SNR_Q must be above ~125 to satisfy the requirements given above. Such high values can be reached by increasing the laser power and pulse repetition frequency (PRF), or enlarging the integration over altitude and time, as SNR is usually magnified by the square roots of the energy and number of averaged samples. However, limits on the latter are also set by Wulfmeyer et al. (2015) for the same applications;





154 integration range Δz should be below 100 m in the PBL and 300 m in the lower free troposphere, 155 whereas an integration time Δt between 15 (assimilation and verification) and 60 min 156 (monitoring) is required.

157 We derive the errors expected on RH given those on temperature and WVMR at the bottom of 158 Table 1. Here and in the following, %RH denote absolute percentage units on RH, whereas % 159 denote relative errors. Relative humidity is derived as a function of atmospheric pressure, 160 temperature and WVMR, using standard empirical relationships for the water vapor saturation 161 pressure. Here, we use the Buck equation (Buck, 1981), which is accurate within 0.2% between 162 -40° C and $+100^{\circ}$ C:

$$P_{wv,sat} = 6.1121 \frac{T}{T + 257.14^{\circ}\text{C}} \exp\left(18.678 - \frac{T}{T + 234.5^{\circ}\text{C}}\right)$$
(13)

$$RH = \frac{P}{P_{wv,sat}} \frac{r_{H_2O}}{r_{H_2O} + 621.991 \,\mathrm{g \, kg^{-1}}}$$
(14)

163 with P pressure and $P_{wv,sat}$ the water vapor saturation pressure in hPa, T temperature in °C.

164 **2.3 Sources of bias**

Biases arising from inaccurate measurement of any of the estimated factors of Eqs. (3-7), or from a variation after that measurement due to instabilities in the instrument, must also be smaller than the aforementioned values of 2-5% for WVMR and 0.12-0.4%, an especially difficult goal to reach for temperature. Their impact must be mitigated either by careful design or by precise estimation.

170 The expected (i.e. noiseless) values of *R* and *Q* can be detailed as:

$$\overline{R(z)} = \frac{O_{H_2O}(z)}{O_{N_2}(z)} \frac{K_{H_2O}}{K_{N_2}} \frac{\sigma_{H_2O}}{\sigma_{N_2}} r_{H_2O}(z)$$
(15)

$$\overline{Q(z)} = \frac{O_{RR2}(z)}{O_{RR1}(z)} \frac{K_2 \sigma_{RR2}(T(z))}{K_1 \sigma_{RR1}(T(z))}$$
(16)

with \bar{x} denoting the expected value of variable *x*, K_j and $O_j(z)$ the instrumental constant and overlap factor of channel *j*, respectively. To simplify our discussion, we choose to incorporate any deviation that affects the ratios without a range-dependence into the instrumental constant ratio, and any deviation with a range-dependence into the overlap ratio.





- As previously explained, the impact of deviations on variables in Eq. (15) remains tolerable below a few percent, but for the distinctly more constrained temperature retrieval, the variables in Eq. (16) are affected by the following effects that directly induce significant bias:
- Laser wavelength drift or filter central wavelength (CWL) drift with temperature both 178 179 affect the ratios indiscriminately with range. By simulating the variation of Q with the 180 WALI filter parameters (section 3), we find a large impact of a wavelength drift $\Delta\lambda$ 181 (measured between the laser on one side and both interference filters on the other side): $dO/O/d\lambda \approx -0.26 \text{ pm}^{-1}$ and $\Delta T \approx -0.34^{\circ}\text{C pm}^{-1} \Delta \lambda$, meaning just 3 pm drift in 182 183 either filter or laser wavelengths can lead to biases above 1°C. That is one of the reasons 184 why the laser must be frequency-stabilized. Also, IFs subjected to fluctuations of local temperature are known to experience CWL drifts; for WALI's filters manufactured by 185 Materion, this amounts to 1.28 pm °C⁻¹ (value given by the manufacturer after their 186 187 material dilation simulation). The temperature of the polychromator must thus be kept 188 stable within 1°C for this bias to become negligible.



$$\Delta CWL(\theta') \approx CWL \frac{{\theta'}^2}{2n_{eff}^2}$$
(17)

where *CWL* is the filter central wavelength, θ' is the angle of incidence on the filter (assumed small), and n_{eff} is the effective index of the filter. For the RR1 filter (n_{eff} = 1.62), we obtain as much as $\Delta CWL(\theta') \approx 43 \text{ pm } \theta'(^{\circ})^2$. The problem stems from the fact that because the filter is in the pupil plane, after collimation of the received beam, each angle of incidence corresponds to a different point in the focal plane of the receiver, which in turns corresponds to a field angle θ of the lidar, as seen on Figure 1 a). Aperture number conservation across the receiving optical system imposes

$$\theta' = \frac{f}{f'}\theta > \frac{D_{rec}}{D_{IF}}\theta \tag{18}$$

199 where f and f' are the receiver/recollimation focal lengths, D_{rec} and D_{IF} are the receiver 200 and IF diameters. For a 150-mm diameter receiver using a 1-inch diameter (22 mm clear 201 aperture) IF, we obtain at least $\theta' = 0.39^{\circ}$ for a $\theta = 1$ mrad field angle, producing 202 $\Delta CWL(\theta') \approx 6.6 \, pm$ and already $\Delta T \approx 2,2^{\circ}$ C. Note that the impact gets





203 proportionately larger with the diameter of the receiver. Because the optical path of each 204 channel is independently aligned, this always induces different overlap factors even 205 when sharing the same telescope. This large effect must be calibrated and corrected, yet 206 its impact was never discussed before in the RR lidar literature, despite being three times 207 as large in other systems with 450 mm receivers. This impact can be mitigated by 208 attacking the filters at normal incidence, where the derivative of CWL as a function of 209 AOI (see Eq. (17)) is minimal.



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211

212	Figure 1. a) Definition of useful parameters for field angle θ and filter angle of incidence
213	θ ' calculations. f: receiver focal length, D_{rec} : receiver diameter, ϕ : full lidar field-of-
214	view, f' : collimation focal length, D_{IF} : IF diameter. b) Definition of metrics for overlap
215	calculations. e : emitter-receiver separation, H : hyperfocal distance, z_e : entry distance of
216	laser into field-of-view.

Detector response non-uniformity up to ±12%, both as a function of impact point on the active surface and of angle of incidence, is now specified on the cathodes of PMTs used at 400 nm wavelength (Hamamatsu (2007), Section 4.3.3). The amplitude was found to be much larger by Simeonov et al. (1999), with significant impact. This effect has been bluntly limited in all our lidars by putting the cathode plane as far as possible before the focal plane, while still avoiding vignetting. It can still be responsible for differences of overlap factors between channels.







• Slight variations of overlap or channel transmittance after calibration will be directly responsible for bias. In the next sub-section, we discuss how they can appear.

228 2.4 Overlap measurement with horizontal shots and limitations

Range-dependent biases influence the lower part of the profiles just like overlap factors, at 229 230 varying distances from the emitter depending on both the quality of the alignments and 231 characteristics of the receiving optics. Two methods are used in the literature to estimate the 232 overlap factors of a Raman lidar: i) an iterative Klett inversion of elastic and Raman channels 233 sharing the same telescope is easy to achieve (Wandinger and Ansmann, 2002) but inefficient 234 when non-common path errors are involved, whereas ii) the method of aiming the lidar 235 horizontally (e.g. Sicard et al., 2002; Chazette and Totems, 2017) is sometimes impractical but 236 more direct and yields more accurate results in an horizontally homogeneous atmosphere over 237 a range of 1 to 2 km. In the context of RR measurements, it is necessary to implement the latter, 238 and also to measure the ratios of overlap factors, rather than the overlap factors themselves, 239 thus avoiding errors due to an imprecise estimation of atmospheric extinction.

240 Considering a horizontal line of sight in a supposedly homogeneous atmosphere, the expected 241 values of ratios *R* and *Q* can be expressed as:

$$\overline{R(z)} = R(z_{\infty}) \frac{O_{H_2O}(z)}{O_{N_2}(z)} \exp(-\Delta \alpha \cdot z)$$
(19)

$$\overline{Q(z)} = Q(z_{\infty}) \frac{O_{RR2}(z)}{O_{RR1}(z)}$$
(20)

where $R(z_{\infty})$ and $Q(z_{\infty})$ are the values observed when all overlap factors have become constant at a sufficiently large range from the lidar, noted z_{∞} , after which variations of the optical path inside the reception channels become negligible. $\Delta \alpha = \alpha(407\text{nm}) - \alpha(387\text{nm})$ is the difference of atmospheric extinction between the two VR wavelengths.

To evaluate z_{∞} , we introduce in Figure 1 b) parameters that characterize the overlap of a paraxial 246 247 or coaxial lidar (e.g. Kuze et al., 1998): i) $z_e = 2e/\phi$ at which the emitted laser beam located at distance e from the receiver axis enters the field of view, whose full size is ϕ ; z_e is null for a 248 249 coaxial system ii) $H = D_{rec}/\phi$, the so-called hyperfocal distance, minimum range from which the beam originating from a point still fully enters the field stop; iii) $H_{IF} = 2D_{rec}f/f'\theta'_{max}$, that 250 251 we might call the filter hyperfocal distance, similarly to the former, the minimum range from 252 which the image of a point does not exceed θ'_{max} , the AOI on the IF that significantly changes 253 its transmittance. z_{∞} is above the maximum of those three, which is usually H_{IF} . If we use for





- 254 θ'_{max} the AOI value causing 1°C bias on temperature per Eq. (10) and Eq. (15), we find: $z_{\infty} > H_{IF}$ 255 = 780 m. Note that z_{∞} can reach several km with misaligned filters.
- 256 If for instance the lidar can be mounted on a rotating platform capable of aiming horizontally,
- the overlap ratios can be estimated with suitable precision by averaging over time and range
- 258 (and correcting for differential of extinction on the VR ratio):

$$\widehat{OR_R}(z) = \frac{R(z)}{R(z_{\infty})} \exp(\Delta \alpha \cdot z)$$
(21)

$$\widehat{OR_Q}(z) = \frac{Q(z)}{Q(z_{\infty})}$$
(22)

- 259 However, assumptions are made for this estimation, namely:
- As explained above, the atmosphere is assumed to be homogeneous in WVMR and temperature (down to <0.5°C) up until z_∞, whereas the overlap ratios must be constant (down to <0.4%) after z_∞. Also, the maximum range (with sufficient SNR) of the lidar must exceed z_∞, implying nighttime measurements for the Raman channels. Therefore, the effects generating overlap variation after a few hundred meters must be prevented.
- The lidar is assumed to retain the exact same overlap functions when aiming horizontally and vertically. Considering a field of view around 1 mrad, the stability of the emission and reception optical paths must be better than ~10 µrad between these two positions. This is feasible for a small refractor but difficult for a Raman system such as WALI, with a heavy laser and large reflector.
- These difficulties make it extremely challenging to estimate the overlap ratios with an accuracy better than a few percent. This is enough for the WVMR, but we find that a correction must be applied by comparing with in-situ sounding for temperature measurements by Raman lidar.

273 3 Implementation and bias mitigation on the WALI system

In this section, we describe the WALI instrument from the emitter to the reception channels, characterizing the critical elements in the framework of WVMR and temperature measurements. The system has evolved from its previous implementation described in Totems et al. (2019), by adding RR channels and a fibered telescope receiver. A global diagram presenting the main lidar sub-systems is shown in Figure 2, and a summary of its characteristics is given in Table 2.









Figure 2. Global diagram of the lidar system. The main sub-systems are: the emitter (center), the elastic receiver using a refractor (top), the Raman receiver using a fibered parabolic reflector (bottom), and a separate, thermally stabilized polychromator (upper right). See Figure 5 for the detail of the polychromator design.

285 Its main features are a single rotatable platform (lightweight carbon fiber breadboard by 286 CarbonVision GmbH) carrying both its emission and reception paths, a 150-mm refractor for 287 the elastic channels (for aerosol studies), and a 150-mm diameter parabolic fibered reflector for 288 all the Raman channels. The separation of the four Raman channels takes place in a deported 289 polychromator set in a thermally controlled enclosure, fed by the optical fiber. Fiber optics are 290 also known to partly scramble the input illumination, minimizing the range-dependance of filter 291 transmittance for the different Raman channels. The output signals from the photomultiplier tubes (PMTs) in the polychromator are digitized by a NITM PXI system (not shown). 292

Table 2. WALI instrument characteristics summary (PRF: pulse repetition frequency, FOV:field of view, CWL: central wavelength in vacuum, FWHM: full width at half-maximum, OOB:

295 out-of-band blocking specification, OD: optical density)

Emitter	Laser	Lumibird TM Q-Smart 450 SLM, tripled Nd:YAG, frequency stabilized $\lambda_{\text{laser}} = 354.725 \text{ nm in vacuum}, E_p = 100 \text{ mJ}, PRF = 20 \text{ Hz}.$						
	Optics	High-power polarizing beamsplitter and 10x beam expander Output beam diameter: 65 mm, Em/Rec separation: 200 mm						
Elastic	Optics	Ø150 mm F/2 UV fused-silica refractor						
receiver	Spatial filter	0.67 x 2 mrad FOV						
	Spectral filter	CWL = 354.71 nm, FWHM = 0.22 nm, OOB: OD >4.0						





Raman	Optics	Ø150 mm F/4 Newton reflector
receiver	Spatial filter	Ø1.67 mrad FOV
	Fiber optics	Ø1 mm, 2-m long, OH-rich multimode fiber
	VR spectral filters	365 nm longpass (OD >2) + 395 nm (OD >2) beamsplitter + Materion TM interference filters:
		N ₂ : CWL = 386.76 nm, FWHM = 0.27 nm, OOB: OD >4.0
		$H_2O: CWL = 407.59 \text{ nm}, FWHM = 0.34 \text{ nm}, OOB: OD > 4.0$
	RR spectral filters	365 nm shortpass (OD >2) + CWL = 355 nm, FWHM = 10 nm, flat-top, OOB: OD >6.0 + 50:50 non-polarizing beamsplitter + Materion TM interference filters: RR1: CWL = 354.09 nm, FWHM = 0.24 nm, OD >6.0 at 354.7 nm RR2: CWL = 353.22 nm, FWHM = 0.54 nm
Detection	Photodetectors	Hamamatsu H10721-210 photomultiplier tubes (PMT) with >0.13 A/W cathode sensitivity
	Amplification	Up to 2 10 ⁶ . Elastic & RR: fixed, VR: sky-background piloted
	Acquisition	3x NI TM PXI-5124 two-channel digitizers
		Sampling frequency: 200 MHz, 12-bit, Q-switch-trigggered
	Recording	$1000 \text{ shots } (\Delta t_0 = 1 \text{ min}), 200 \text{ MHz } (\Delta z_0 = 0.75 \text{ m})$ Analog + photon-counting

296 **3.1 Emitter**

The emitter is a commercial Lumibird/Quantel "Q-Smart 450" Nd:YAG pulsed laser, stabilized by injecting the output of a single longitudinal mode fiber laser emitting at 1064.175 nm into the main cavity ("SLM" option), and frequency-tripled to emit at wavelength $\lambda_{\text{laser}} = 354.725$ nm (in vacuum). The nominal pulse energy for the Q-Smart 450 with SLM is 100 mJ at 355 nm, with a Pulse Repetition Frequency (PRF) of 20 Hz. These values set WALI near the eye safety limit for pulsed energy, making the system eyesafe at the output of a 2-meter funnel due to built-in leaks at 532 nm.

A critical issue to be cleared before using the Q-Smart 450 SLM in WALI was the spectral purity and stability of the laser, in terms of linewidth and wavelength drift. The laser seeder at 1064.175 nm is specified with a 50 MHz (0.062 pm at 355nm) stability at fixed temperature, and 37 MHz $^{\circ}C^{-1}$ (0.046 pm $^{\circ}C^{-1}$ at 355 nm) temperature drift.





Nevertheless, the stability of the Q-Smart emission at 354.725 nm has been verified with a dedicated optical setup, sending the output of a Michelson interferometer with optical path differences (OPD) between 0 and 100 mm on a UV-sensitive CCD camera. By extracting the contrast and phase variations of the fringes at large OPDs from the videos, we were able to ascertain:

- the laser linewidth, without seeder, to be 24±2 pm (versus 26.5 pm datasheet value), and
 with seeder, to be small compared to 1 pm (versus 0.2 pm datasheet value),
- the wavelength drift, without seeder, to be below 8 pm over 10 minutes, and with seeder,
 to be below 0.2 pm RMS (root mean square fluctuations) over 5 minutes. We consider
 the remaining fluctuations to be due to the ~0.05 pm °C⁻¹ temperature-linked drift of
 the seeder, which is not temperature-controlled.
- 319 Given the requirements derived in Section 2.3, this makes the seeded Q-Smart laser 320 theoretically suitable for RR measurements of temperature.

321 3.2 Raman receiver

322 In this sub-section we discuss the possible impact on the VR/RR ratios of the fibered reflector 323 (beam scrambling and fiber optics fluorescence), of Raman filters characteristics, and of the 324 polychromator design and alignment. As far as we know, this type of comprehensive study does 325 not exist in the literature for Raman lidars.

326 3.2.1 Fibered reflector telescope and scrambling of the lidar field-of-view

The elastic and Raman receivers are both 150 mm in diameter. The focal length of the refractor (elastic channels) is ~300 mm, which with a 200x600 μ m field stop achieves full overlap at ~150-200m. However, the focal length of the reflector (Raman channels) is 600 mm (parabolic mirror with aperture F/4); this implies using a multimode fiber optics about 1 mm in diameter as the field stop to allow similar results in terms of field-of-view and overlap. The chosen fiber optics is an OH-rich UV fused silica fiber, 2 m in length and 1000 μ m in core diameter, with numerical aperture 0.22 (Avantes FC-UV1000-2).

Coupling the reflector output into a multimode fiber (e.g. Chourdakis et al., 2002) allows: i) to minimize occultation of the primary mirror (here only 12 mm in diameter), ii) to deport the Raman channel separation away from the telescope, making it a separately tunable optical system, minimizing the overall lidar size and making light or temperature confinement easier, iii) in theory, to scramble the fiber output illumination versus the lidar field angle, therefore





minimizing the range-dependence of AOIs on the IFs discussed in Section 2.3, and flatteningoverlap ratios after the geometrical full-overlap distance.

341 The scrambling of the lidar field-of-view, via the multiple internal reflections in the fiber, has

342 been experimentally tested by imaging the output of the fiber, with a varying point-like input.

343 The results are shown in Figure 3. Note that the radial coordinate of the output point relative to

344 the center of the fiber corresponds to a given AOI on a well-aligned IF in the following

345 polychromator, after a f' = 50 mm doublet lens.



346

Figure 3. Images of the output facet of the 1 mm diameter multimode fiber optics for a) centered and b) decentered (at $x_{in} = 0.38$ mm horizontal offset from the center of the core) input point of a 20-mm beam focused on the input facet of the fiber, and energy density profiles along the x and y axes.

351 It appears on Figure 3 b) that the input energy is mostly redistributed tangentially (i.e. along the 352 angular polar coordinate, as opposed to radially) by its passage through the fiber. The radial 353 dispersion remains small, and the mean output radius is approximately equal to the input radial 354 coordinate. Manually applying curvature to the fiber, as suggested by so-called "mode 355 scrambling" devices, did not make the energy distribution more uniform so much as creating 356 unwanted losses (effect not shown). Even for a centered input, the energy radial distribution – 357 i.e. the percentage of the total output in a given radial bin, that will therefore impact a well-358 aligned filter at the same AOI - is uniform. We conclude that while minimizing effects of filter 359 misalignment, the use of fiber optics does not substantially make the angle of incidence on the 360 interference filters independent from the image position in the focal plane of the telescope, in contrast to what could be expected. Range-dependent biases will not be strongly mitigated. 361





362 3.2.2 Fiber optics fluorescence

It has been shown by Sherlock et al. (1999) and discussed by Whiteman et al. (2012) that fiber optics fluorescence could be an obstacle to water vapor measurements, because elastic scattering at 532 nm was inducing fluorescence in an OH-poor fiber at a non-negligible level compared to the atmospheric Raman scattering. It was solved by using an OH-rich fiber, but it was predicted in the latter work that the effect could be larger at 355 nm.

We have characterized this effect in the WALI fiber optics, using a narrowband CW laser excitation centered at 355 nm. The output of the fiber was analyzed by a Fourier transform spectrometer (Thorlabs OSA201C spectrum analyzer), behind a longpass dichroic plate cutting the direct LED emission, and the same collimating achromat as in the polychromator. The resulting spectrum is shown in Figure 4.





Figure 4. 1000-µm diameter, 2-meter long fiber fluorescence measurement with 355 nm laserillumination.

We plot both the raw spectrum and the Fourier transform spectrometer noise floor after 1000 profile integrations, to highlight the very weak features observed at 780 to 910 nm, and the high associated uncertainty. Due to the noise level, and given the dichroic plate residual transmittance of the laser wavelength, we can only ascertain that the fluorescence power spectral density (PSD) around 400 nm is lower than 10⁻⁶ times the peak laser PSD, although no feature can be detected in this spectral domain. Note that fluorescence between 400 and 500 nm





- 382 was indeed observed using a broadband excitation from a fibered LED at 340 nm (not shown).
- 383 Nevertheless, the amount of rejection observed for a 355 nm excitation is sufficient to exclude
- an adverse impact of the OH-rich fiber optics for Raman lidar measurements.

385 3.3 Raman channels

386 3.3.1 Polychromator configuration

387 The RR+VR polychromator configuration used in WALI is presented in Figure 5. Dichroic and non-polarizing beamsplitters are used to separate the channels. In contrast to the design of 388 389 Hammann et al. (2015) which optimizes throughput and laser-line rejection on the RR channels, 390 we chose to implement a splitter-based configuration, favouring a compact system (25x25 cm, 391 easier to confine) and normal incidence on the filter, at the expense of SNR. Indeed, designing the filters for a correct CWL at 5° incidence (as in the cited work) instead of 0° dramatically 392 393 narrows the filter angular acceptance, as can be deduced by deriving Eq. (15) as a function of incidence θ' . In the WALI polychromator, the output from the fiber is collimated by a near-UV 394 395 achromat with 50 mm focal length, resulting in a 22 mm diameter beam. Dichroic beamsplitters 396 with adequate cut-on wavelengths are used to separate channels. On each separated channel, an 397 aspheric lense condenses light on the PMT surface, located 4 mm before the focal plane. A cage system assembly holds all parts with great stability, however beamsplitters are not always 398 399 perfectly aligned at 45° in the stock cage cubes. That is why all filter, lens and PMT sub-400 assemblies are mounted on tiltable mounts to allow precise alignment at normal incidence.



401

Figure 5. Compact rotational & vibrational Raman separation configuration used in WALI. IF:
interference filter (with CWL - FWHM given in nm), OD: optical density, BS: beam splitter,
LPD: long-pass dichroic beamsplitter (with cut-off wavelength given in nm), PMT: photomultiplier tube. This polychromator is thermally regulated in a dedicated light-tight enclosure.





406 3.3.2 Filters qualification

407 All interference filters were custom-made by Materion, including the RR filters on 408 specifications graciously shared by the team of A. Behrendt (following Hamann et al., 2015). 409 They were characterized on the Fourier transform spectrometer (described in section 3.2.2) 410 prior to mounting, using fibered LEDs peaking at 340, 385 and 405 nm as the light source; the 411 beam was collimated by the same near-UV achromat with 50 mm focal length. We give the 412 measurements results for the RR filters in Figure 6 and Table 3.

413 The effective index and angular acceptance of the filters (arbitrarily chosen for a 10% loss at 414 the CWL) were assessed by tilting the filters of a known angle. A critical parameter, the 415 transmittance of both filters at the laser line λ_{laser} in operational conditions was assessed on the 416 lidar itself, by measuring the energy of an echo on a hard target located at 200 m, and switching 417 between an elastic IF of known transmittance with a known strong optical density and the RR 418 IF in question. The excellent extinction in the RR1 filter guarantees a minimal effect of elastic 419 signal leak in temperature retrievals, but it was nevertheless subtracted as in Eq. (3). Note that 420 no significant echo was detected on the H2O-Raman channel, indicating extinction better than a few 10^{-9} , thanks to the two dichroic plates. 421



422

Figure 6. RR filters spectral transmittance measured on optical spectrum analyzer with illumination by a 340 nm LED: RR1 (low-J) and RR2 (high-J) filter at 0° incidence.

425 Table 3. Measured RR IF characteristics. All CWL values are given in vacuum.





	RR1 filter	RR2 filter	Uncertainty
CWL	354.09 nm	353.22 nm	0.01 nm
FWHM	0.24 nm	0.54 nm	0.01 nm
n _{eff}	1.62	2.03	0.05
Max transmittance	69%	51%	5%
Laser line transmittance	2.7 10-8	2.9 10-7	10% relative
Angular acceptance (AOI for 10% loss at CWL)	1.5°	2.5°	0.2°
CWL shift at max field angle (i.e. edge of fiber, AOI = 0.59°)	-9.8 pm	-2.5 pm	0.3 pm

426 3.3.3 Polychromator alignment and qualification

427 Due to the filter CWL shift evolving as the square of the AOI in Eq. (15), it is essential to 428 minimize range-dependent biases by aligning the filters at a precisely normal incidence from 429 the input beam. However off-the-shelf beam splitter plate holders are found to be misaligned 430 by up to 1° from an ideal 45° incidence. All PMTs are mounted jointly with their own IF and 431 lens into a tiltable mount to correct for this (represented on Figure 7).

The alignment of these mounts is performed in the lab by conjugating an input multimode fiber of $600 \,\mu\text{m}$ diameter replacing the lidar input, into a target fiber $200 \,\mu\text{m}$ in diameter at the focus of the PMT lens, through the polychromator. Fibered LEDs are used for illumination like in section 3.3.2. All the channels are sequentially addressed in this manner. By obtaining a maximal energy and a radially uniform profile at the output of the target fiber, one can ensure alignment with a precision of 0.1 to 0.3° .



438





Figure 7. Method for polychromator alignment validation. Light from LEDs is input in the
WALI fiber optics, passes through the polychromator, and into a multi-mode fiber (MM fiber,
Ø600 µm) analysed by a Thorlabs OSA201C Fourier transform spectrometer. Channel central
wavelengths are expected not deviate from those of the filter measured independently at normal
incidence, to validate alignment.

To verify the result, the spectral transmittance of the polychromator channels themselves are characterized by the Fourier transform spectrometer, as shown on Figure 7. By illuminating the channel with a LED coupled in the actual lidar fiber, we ensure that the polychromator is studied in operational conditions. The CWL of each channel is expected not deviate by more than 20 pm (twice the empirical accuracy) from the CWL measured on the individual filter at normal incidence, to validate the alignment. The polychromator aligned using the procedure proposed above passes this test.

451 **3.4 Detectors**

Hamamatsu 10721P-210 PMTs, with >0.13 A W⁻¹ cathode sensitivity at 400 nm, and up to 452 453 $\sim 2 \ 10^6$ controllable internal gain, are used to transform the optical flux into an electric current, 454 directly digitized at 200 MHz (0.75 m sampling along the line of sight) by three NI PXI-5124 455 two-channel digitizers with 50 Ω load. The acquisition software, custom-made with Labview, 456 conducts analog and photon-counting (thresholding at ~3 standard deviations of the noise) accumulations in parallel during 1000 shots (50 seconds), every minute, which are then pre-457 458 processed and recorded (~10 seconds down time). Every ~8 minutes, baselines are recorded 459 with PMT gains set at zero. The next sub-sections describe critical points of the detectors 460 affecting the RR and VR channel ratios.

461 3.4.1 PMT response variability

As explained in Section 2.3, the non-uniformity of the PMT response can affect the ratios of Raman channels as a function of range. We tested the sensitivity profiles of WALI's H₂O-Raman PMT to continuous laser illumination at 405 nm wavelength, first using a 1-mm diameter collimated beam, as a function of both point and angle of incidence. A cumulated ~6.0 optical density was used to avoid saturation of the PMT.

467 As shown on Figure 8 a), a strong variation of sensitivity by a factor of almost 2 is found on the 468 PMT surface, much larger than specified. The relative sensitivity is lowest near the center of 469 the PMT and highest on the sides, on a diameter of 4 mm approximately equal to the spot size 470 in the lidar. Indeed the PMT surface is 4 mm before the focal plane of the 0.5 NA condensing





- 471 aspheric lens. This is consistent with the results of Simeonov et al. (1999) on an older generation
 472 of detectors, excluding a suspected hole-burning phenomenon over the lifetime of our PMT.
 473 On the vertical axis, we also note the effect of the gridded cathode. Note that sensitivity does
- 474 not vary by than a few percent as a function of angle of incidence (not shown).



475

Figure 8. Study of the non-uniformity of the PMT response: a) as a function of point of impact on the active area along the horizontal (blue) and vertical (red), with a 2 mm collimated beam from a 405 nm laser, b) as a function of angle of incidence on the lens and PMT assembly similar to the ones used in the WALI polychromator, with a 21 mm collimated beam from the same laser. Dashed lines represent uncertainty calculated over multiple measurements.

We then put the condensing lens used in the polychromator in front of the PMT, and studied its response as a function of AOI on the lens+PMT assembly, which is shown in Figure 8 b). The input beam was the nominal size in the polychromator ie. ~22 mm in diameter. We find that the curve corresponds well to the measured sensitivity profile, smoothed by its convolution by the spot on the PMT. The problem is that at normal incidence, the derivative of sensitivity with incidence is 2-5% per degree. In the future, the condensing lenses will be replaced with afocal beam reducers to reduce this dependency.

488 3.4.2 Baseline and EM parasites correction

The baseline induced by the detection chain is found to vary between channels and in time. It is also subject to electro-magnetic (EM) interference causing parasitic signals of both high frequency, mostly due to the flashlamp high peak current radiating over the system, and low frequency, probably due to other neighboring electronics. For this reason, the channel baselines are evaluated regularly (by averaging 1000 shots with PMT gain set to zero, every 8 minutes), smoothed and corrected (L_j in Eq. (2)). However, for the Raman channels (H₂O and RR2 specifically), the weakness of the signals requires a specific care of EM compatibility, as





repeating parasitic spikes were found to jam the channels (especially photon counting) startingat altitude 6-7 km.

Figure 9 a) shows an example of perturbed baseline. Trial and error established that common methods to avoid ground loops were not all efficient: star grounding of the various cables worsened the problem, whereas physically separating coaxial signal cables from direct current power supply and control voltage cables, and grounding all connectors and opto-mechanics again on the breadboard side, mitigated it, reaching the baseline plotted in Figure 9 b).



Figure 9. Analog detection baseline measurements (red) over 1000 laser shots with PMT gains set to zero, expressed in photon counts equivalent on the RR2 channel: a) in an unfavorable case (no mitigation), showing both baseline fluctuations over time (20 km ~ 133 µs) and strong electro-magnetic parasites at large distance; b) on the WALI system, after mitigation. The final estimated baseline ($\hat{L}_j(z)$ in Eq. (3)) obtained after smoothing, which is subtracted to all recorded profiles, is in black.

510 3.4.3 PMT gain adaptation

511 On each channel, PMT internal amplification gain G (using photoelectron multiplication) is a 512 definite function of its control voltage U. The variation of G by ~ 2 orders of magnitude allows 513 for the optimization of the dynamic range. This helps deal with the different Raman cross-514 sections in each filter, with variations of atmospheric transmittance, and especially with sky 515 background levels during daytime. The gain is pushed at its maximum possible value still satisfying two conditions: i) the signal voltage maximum does not exceed the range of the 516 517 digitizer, ii) the sky background signal does not exceed the maximum output current of the PMT that guarantees linearity (100 μ A, ie. $\langle S_{raw} \rangle < 5$ mV). This is indispensable for day-round 518





- 519 measurements of WVMR, otherwise the channels would be saturated during daytime (Chazette
- 520 et al., 2014b), or suboptimal in SNR during nighttime.



521

Figure 10. Calibration of PMT gain *G* versus control voltage *U*: a) log-gain measurements and
second-degree polynomial model for all Raman channels, and b) relative gain ratio error
between model and measurements for vibrational and rotational Raman channel ratios.

525 However, PMT gain adaptation leas to biases on the Raman channel ratios if the gain versus 526 control voltage characteristics are not known with a better precision than the requirements 527 stated in Table 1 (2% on VR channels, 0.4% on RR channels). In Figure 10 a), we show the 528 experimental calibration of G versus U as well as second-degree polynomial fits for each 529 channel. The relative error on the VR and RR channel gain ratios approximated by these models 530 is plotted on Figure 10 b), with the measurement uncertainty. This uncertainty is mostly due to 531 variations of atmospheric parameters and laser energy during calibration. Since all relative 532 errors are well centered, we compute that the possible error for the gain ratio with these models 533 is ~1.3 %. This is compatible with WVMR measurements but not with temperature 534 measurements. Therefore, the PMT gain should only be adapted on the VR channels, and the 535 RR channels should be kept at a fix value of gain.

536 3.4.4 Merging analog and photon-counting signals

Both analog and photon-counting raw signals are recorded. The analog signal has lesser SNR
at high altitude during nighttime, whereas the photon-counting signal is saturated at low altitude
and by daylight; by merging them correctly, an optimal SNR can be obtained (Newsom et al.,





540 2009). For signal processing, the photon-counting raw signals are first desaturated (details in 541 Chazette et al., 2014b). Merging is performed during nighttime on the pre-processed signals 542 defined in Eq. (3). After calculating a photon to Volts conversion constant at an altitude where 543 photon-counting is not saturated, the converted photon-counting profile replaces the analog 544 profile after a predefined altitude depending on signal strength (from 1 km for the H₂O VR 545 channel, up to 4 km for the elastic channel).

546 We wish to emphasize here that baselines L_j and background signals B_j in Eq. (3) must be 547 estimated separately for the analog and photon-counting recorded profiles (which have no 548 baseline, and a smaller but non-zero background value due to the suppression of electronic 549 noise). Otherwise, the merged signal will show discontinuities at the cut-off altitude, and biases 550 at high altitude at dusk and dawn. Their impacts are typically much larger than the requirements 551 of Section 2.2.

552 4 Qualification on the atmosphere

In this section, we qualify the WALI system starting with the measurement of its overlap factor ratios, followed by its calibration and comparisons with radiosoundings. Remaining biases are highlighted and corrected, and experimental measurement errors are evaluated.

556 4.1 Experimental set-up and strategy

We put the lidar into operation in our laboratory near Saclay (48°42'42"N 2°08'54"E) over a period of two weeks in May 2020. It was placed on a rotating platform below a trapdoor equipped with silica windows for zenith shots, and in front of a window at a height of about 9 m above the ground level (agl) for horizontal shots. During the latter, the lidar aimed North <5° above the horizon (beam elevation <80 m per km of range). In that direction, land use is fields up to 800 m range, buildings and trees between 800 and 2 km range, and fields again up to 5.5 km range.

To calibrate and qualify the lidar measurements, we use radiosoundings launched two to three times daily from the operational Météo-France station located in Trappes (48°46'27"N 2°00'35"E, 12.3 km WNW from the lidar near Saclay, approximately upstream in the prevailing winds).





568 **4.2** Measurement of overlap ratios with horizontal shots

569 The overlap factors and their ratios were estimated on signals averaged over 3 hours after sunset 570 on December 19th, 2019, with a rather lukewarm, unturbulent but hazy atmosphere (aerosol extinction coefficient 0.32 km⁻¹ at 355 nm with Angström exponent ~1.5, 11°C ground 571 temperature, and WVMR at ground level around 6.5 g kg⁻¹). With a planetary boundary layer 572 (PBL) height of ~900 to 1000 m, and slow gradients of temperature (-1 to -4°C km⁻¹) and 573 574 WVMR (-0.8 to -1.2 g kg⁻¹ km⁻¹) in that PBL (as measured by radiosoundings launched from Trappes at ~12:00UTC and 0:00UTC, presented in the next subsection), conditions were 575 576 excellent for a homogeneous atmosphere within the first 5 km at least. 577 The estimated overlap factors of the different channel, with atmospheric extinction fitted

577 The estimated overlap factors of the unreferred channel, with autospheric extinction fitted 578 between 800 and 2000 m, are shown in Figure 11 a). Full geometrical overlap is obtained as 579 expected between 150 and 200 m, but the curves differ by several percent between the Raman 580 channels. Atmospheric extinction drifts from the estimated value after 2 km.

- The estimated ratios of overlap factors OR_R and OR_Q are plotted in Figure 11 b) and c), at 7.5 m resolution (thin line) and after smoothing (thick line, final correction used hereafter). Peak
- divergence is 5 to 7 %, at ~150 m. Convergence within 1% happens at ~400 m, but oscillations
- of lower amplitude persist until ~3 km. We note that for OR_Q , deviations do not exceed the
- $\pm 0.7\%$ required to maintain bias below 1°C. They are nevertheless corrected.



586









Figure 11. a) Overlap factors measured over 3 hours of nighttime measurements with a
horizontal line-of-sight on Dec. 19, 2019. Estimated overlap ratios between VR (b) and RR (c)
channels: native resolution (thin blue line), final estimate after smoothing (thick red line).

591 4.3 Comparison to radiosoundings and calibration, estimation of residual error

12 nighttime and 24 daytime radiosoundings were launched from Trappes between May 20th and June 2nd, 2020. Lidar profiles are averaged from 0 to 40 minutes after the radiosounding launch time. The range averaging is progressive and defined to keep the night time temperature error below 1.5°C: range bins are 15 m long below 100 m agl, growing to 360 m above 8 km agl.

597 In order to debias WVMR and temperature measurements from residual errors on OR_R and 598 OR_Q , we perform a three-step calibration:





First step: we exclude the first 1500 m agl of the profiles when fitting *r_{H2O} in-situ* vs *R*'
 and *Q*' vs *T in-situ* to estimate *K* and *f* respectively. This initial calibration is shown in
 Figure 12 a) & d).

- 602 Second step: using these first estimates, we then plot the ratios between the lidar observables R' & Q' and the expected observables deduced from the in-situ 603 604 measurements and these initial calibration parameters. This provides an estimate of the 605 remaining biases on OR_R and OR_Q , which we find to be up to ~4% and ~1.8% respectively. This represents a small correction to the overlap ratios estimated while 606 607 shooting horizontally, but remains larger than the requirements of precision specified in Table 1. The modeled corrections of OR_R and OR_Q are plotted in red in Figure 12 b) & 608 609 e).
- Third step: we apply the previous estimates of OR_R and OR_Q and we perform a new calibration using all the data (down to 200 m agl), yielding more precise estimates of calibration constants, as shown in Figure 12 c) & f).
- 613 In the three steps, data with SNR lower than 10 for *R*' and 30 for *Q*' are rejected so as to 614 limit the impact of noise present at higher altitudes.



615

Figure 12. Results of calibration on 12 nighttime and 24 daytime radiosoundings launched from
Trappes between May 20th and June 2nd, 2020 for WVMR (upper row) and temperature (lower
row), in three steps: calibration on measurements above 1500 m (a/d) with samples as crosses
(one color per radiosonde) and calibration curve in black; residual overlap ratio estimation (b/e)





620 with samples as crosses, mean ratio in blue and model in red; calibration on all results (c/f). 621 Daytime samples are limited to SNRs above 10 for R' (WVMR) and 30 for Q' (temperature). 622 The reliability of this calibration along time has been tested by comparing to the same exercise performed two months later at the end of July 2020. After calibration in the same conditions 623 624 than in May, we found K decreased by \sim 7.3%, and the temperature associated to a given value 625 of Q' to be ~2.1°C higher. However, OR_R and OR_Q were still accurate within the reachable precision, ie. ~0.2%. It was later proven that a malfunction of the laser seeder was responsible 626 627 for a slow drift of the emitted wavelength. Thus, although a regular verification of the 628 calibration is necessary, the measurement of the overlap ratios is reliable.



629

Figure 13. Residual deviations between lidar and Trappes radiosoundings in terms of WVMR,
temperature and relative humidity, for night time (a/b/c) and daytime (d/e/f), with mean
deviation (thick lines), and RMS error (colored rectangles). The error corresponding to noise





633 levels on the lidar signal is shown as darker rectangles. Cloudy profiles have been discarded. 634 Daytime measurements are limited to SNRs above 5 for R (WVMR) and 20 for Q (temperature). 635 In Figure 13, we examine the residual deviations between the lidar and the same series of 636 radiosoundings used for the calibration. RH has been derived using Eq. (14) from lidar-637 estimated WVMR and temperature, and the pressure profile given by radiosoundings. For each 638 parameter r_{H2O} , T and RH, we plot for daytime and night time profiles the mean and RMS deviations averaged over large range bins as colored bars, as well as the propagated signal error 639 640 as darker shaded areas. This allows to compare the observed random error to what could be 641 expected from the level of noise on the lidar measurements. Note that only profiles with good 642 SNR unperturbed by clouds have been selected for this comparison.

643 On WVMR, the results show little bias, and RMS deviation is dominated by atmospheric 644 variability by night at low altitude and by lidar noise otherwise. On temperature, most of the 645 RMS deviation is explained by noise; a ~1°C significant bias is still seen below 800 m. This 646 seems to be due to an underestimated correction of OR_Q . On Figure 13 c) and f) are plotted the 647 consequences of this bias on relative humidity *RH* to be around 2 to 4%RH, but also the 648 resulting error to be expected. We see that with the defined averaging, random error is around 649 2% RH up to 5 km agl during nighttime and 1 km agl during daytime, growing fast above.

Table 4. Statistics of observed differences for r_{H2O} , *T*, and *RH*: experimental Mean Differences (MD), Root-Mean Square Differences (RMSD), averaged over two different range bins, in the low troposphere (1-2 km) and the free troposphere (5-6 km). Comparison to the "natural" atmospheric variability between the lidar and RS sites as modelled by the ECMWF/IFS ERA5 reanalyses (difference over considered period between grid points nearest to each of the two sites), and to the theoretical root-mean-square error (RMSE) derived from the variance of the RR signals.

	Range	Range resolution Δz	Model atmos. MD	Experimental MD (night/day)	Model atmos. RMSD	Theo. RMSE (night/day)	Experimental RMSD (night/day)
WVMR	1-2 km	84 m	-0.03	+0.06/-0.05	0.41	0.03/0.4	0.54/0.65
(g/kg)	5-6 km	168 m	<10 ⁻²	-0.07	0.11	0.04	0.15
Temperature (°C)	1-2 km	84 m	+0.15	+0.25/+0.3	0.33	0.4/0.7	0.6/0.7
	5-6 km	168 m	+0.05	+0.4	0.28	0.75	0.95





Relative humidity	1-2 km	84 m	-0.23	+0.8/-0.5	6.37	1.7/5.5	6.5/10	
(%RH)	5-6 km	168 m	-0.70	-3.3	7.52	2.2	7	

657

To support the above interpretation, in Table 4 we compare the experimental mean difference 658 659 and RMS difference plotted on Figure 13, averaged over two altitude ranges (low troposphere, LT, 1 to 2 km, and free troposphere, FT, 5 to 6 km), to i) the natural variability of the atmosphere 660 between the radiosondes at Trappes and the lidar at LSCE, as modelled by ERA5 reanalyses of 661 the ECMWF/IFS weather model, ii) the expected random error given the noise level on the RR 662 663 signals. Nighttime and daytime values are indicated in the LT, only nighttime values in the FT. We see that the experimentally observed values of RMSD are rather consistent with the 664 quadratic sum of the RMS variability of the atmospheric variables between Trappes and LSCE, 665 and of the noise-induced RMS error. The excess random difference is thus well explained by 666 667 the distance. There is still a discrepancy with the mean difference of temperature however; it is 668 not explained by the modelled differences of temperature at the locations of the two soundings.

669 **5 Conclusion**

670 During the qualification of the rotational Raman channels for the WALI lidar of LSCE, with 671 the aim of providing profiles of relative humidity, we encountered important sources of bias that are seldom described in the now abundant literature involving such systems. We 672 673 highlighted the predominant effects of the dependency of filter transmittance and detector 674 sensitivity upon angle of incidence and point of impact, respectively. Because the latter 675 parameters are directly proportional to field angle, they cause range-dependent biases on the 676 RR/VR signal ratios that are several times greater than the required accuracy of lidars for temperature measurements (only 0.79% for 1°C here), less so for water vapor measurements. 677 678 We established that this effect cannot be suppressed by using fiber optics between the receiver 679 and polychromator, because scrambling of the lidar field of view does not happen radially in the fiber. Mitigation efforts impose the careful alignment of each filter at normal incidence to 680 681 the input beam, and the verification of the spectral transmittance of each channel on a 682 spectrometer. The thermal stability of the polychromator is also of prime importance. Other significant bias sources include electro-magnetic perturbations of signal baselines and PMT 683 gain variation, which must be mitigated. The impact of fiber optics fluorescence, and of the 684





measured laser linewidth or short-term wavelength drift were shown to be negligible in theWALI system.

687 After a measurement of RR/VR channel ratios during horizontal shots, which showed the 688 significant impact of the above phenomena (up to 5% bias on ratios below 300m, ~1% higher), we calibrated and de-biased the WALI measurements using radiosondes launched from the 689 690 nearby Trappes station of Météo-France. Between the de-clouded lidar measurements and the 691 radiosonde profiles, the remaining mean differences are small (below 0.1 g/kg on water vapor, 692 1°C on temperature) and RMS differences are consistent with the expected error from lidar 693 noise, calibration uncertainty, and horizontal inhomogeneities of the fields between the lidar 694 and radiosondes. On relative humidity we thus reach a goal of ~10%RH random error and 695 5% RH systematic error up to 9 km by night and 1.5 km by day, with 40 min time integration 696 and progressive vertical integration of 15 to 360 m at 10 km. The systematic error on RH is 697 dominated by bias on temperature, whereas the random error is dominated by noise on water 698 vapor measurements.

Thus exhaustively qualified, the WALI system may be applied in the near future to exercises assimilating thermodynamic profiles in weather models, as is expected within the WaLiNeAs (Water vapor Lidar Network Assimilation experiment) project (Flamant et al., 2021).

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 discussion. Radiosoundings from the Tessereinc de Bort station (Trappes) were obtained at
- 705 https://donneespubliques.meteofrance.fr/?fond=produit&id_produit=97&id_rubrique=33,
- 706 courtesy of Météo France. ERA5 reanalyses of the ECMWF/IFS model were obtained at
- 707 <u>https://cds.climate.copernicus.eu/cdsapp#!/home</u> courtesy of the Copernicus Climate Change
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710 References

Adam, S., Behrendt, A., Schwitalla, T., Hammann, E. and Wulfmeyer, V.: First assimilation of
temperature lidar data into an NWP model: impact on the simulation of the temperature field,
inversion strength and PBL depth, Q. J. R. Meteorol. Soc., 142(700), 2882–2896,
doi:10.1002/qj.2875, 2016.

715 Behrendt, A.: Temperature Measurements with Lidar, in Lidar: Range-Resolved Optical





- Remote Sensing of the Atmosphere, vol. 102, edited by C. Weitkamp, pp. 273–306, Springer-
- 717 Verlag, New York., 2005.
- 718 Behrendt, A. and Reichardt, J.: Atmospheric temperature profiling in the presence of clouds
- with a pure rotational Raman lidar by use of an interference-filter-based polychromator, Appl.
 Opt., 39(9), 1372, doi:10.1364/AO.39.001372, 2000.
- 721 Behrendt, A., Wulfmeyer, V., Hammann, E., Muppa, S. K. and Pal, S.: Profiles of second- to
- 722 fourth-order moments of turbulent temperature fluctuations in the convective boundary layer:
- 723 first measurements with rotational Raman lidar, Atmos. Chem. Phys., 15(10), 5485-5500,
- 724 doi:10.5194/acp-15-5485-2015, 2015.
- Buck, A. L.: New Equations for Computing Vapor Pressure and Enhancement Factor, J. Appl.
 Meteorol., 20(12), 1527–1532, doi:10.1175/1520-0450, 1981.
- Chazette, P. and Totems, J.: Mini N2-Raman Lidar onboard ultra-light aircraft for aerosol
 measurements: Demonstration and extrapolation, Remote Sens., 9(12), doi:10.3390/rs9121226,
 2017.
- Chazette, P., Marnas, F., Totems, J. and Shang, X.: Comparison of IASI water vapor retrieval
 with H2O-Raman lidar in the framework of the Mediterranean HyMeX and ChArMEx
 programs, Atmos. Chem. Phys., 14(18), 9583–9596, doi:10.5194/acp-14-9583-2014, 2014a.
- Chazette, P., Marnas, F. and 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, Atmos. Meas. Tech., 7(6), 1629–1647, doi:10.5194/amt-7-1629-2014,
 2014b.
- Chazette, P., Raut, J.-C. and Totems, J.: Springtime aerosol load as observed from groundbased and airborne lidars over northern Norway, Atmos. Chem. Phys., 18(17), 13075–13095,
 doi:10.5194/acp-18-13075-2018, 2018.
- Chourdakis, G., Papayannis, A. and Porteneuve, J.: Analysis of the receiver response for a
 noncoaxial lidar system with fiber-optic output, Appl. Opt., 41(15), 2715,
 doi:10.1364/AO.41.002715, 2002.
- Cooney, J.: Measurement of Atmospheric Temperature Profiles by Raman Backscatter, J. Appl.
 Meteorol., 11(1), 108–112, doi:10.1175/1520-0450(1972)011<0108:MOATPB>2.0.CO;2,
 1972.
- 746 Crevoisier, C., Clerbaux, C., Guidard, V., Phulpin, T., Armante, R., Barret, B., Camy-Peyret,





C., Chaboureau, J.-P., Coheur, P.-F., Crépeau, L., Dufour, G., Labonnote, L., Lavanant, L.,
Hadji-Lazaro, J., Herbin, H., Jacquinet-Husson, N., Payan, S., Péquignot, E., Pierangelo, C.,
Sellitto, P. and Stubenrauch, C.: Towards IASI-New Generation (IASI-NG): impact of
improved spectral resolution and radiometric noise on the retrieval of thermodynamic,
chemistry and climate variables, Atmos. Meas. Tech., 7(12), 4367–4385, doi:10.5194/amt-74367-2014, 2014.

753 Dinoev, T., Simeonov, V., Arshinov, Y., Bobrovnikov, S., Ristori, P., Calpini, B., Parlange, M.

and van den Bergh, H.: Raman Lidar for Meteorological Observations, RALMO – Part 1:

- Instrument description, Atmos. Meas. Tech., 6(5), 1329–1346, doi:10.5194/amt-6-1329-2013,
 2013.
- 757 Flamant, C., Chazette, P., Caumont, O., Di Girolamo, P., Behrendt, A., Totems, J., Lange, D.,

758 Fourrié, N., Brousseau, P., Augros, C., Baron, A., Cacciani, M., Comeron, A., De Rosa, B.,

759 Ducrocq, V., Genau, P., Labatut, L., Munoz-Porcar, C., Rodriguez-Gomez, A., Summa, D.,

760 Thundathil, R. and Wulfmeyer, V.: A network of water vapor Raman lidars for improving heavy

761 precipitation forecasting in southern France - Introducing the WaLiNeAs initiative, Bull.

762 Atmos. Sci. Technol., (submitted), 2021.

763 Fourrié, N., Nuret, M., Brousseau, P., Caumont, O., Doerenbecher, A., Wattrelot, E., Moll, P.,

764 Bénichou, H., Puech, D., Bock, O., Bosser, P., Chazette, P., Flamant, C., Di Girolamo, P.,

765 Richard, E. and Saïd, F.: The AROME-WMED reanalyses of the first special observation period

of the Hydrological cycle in the Mediterranean experiment (HyMeX), Geosci. Model Dev.,

767 12(7), 2657–2678, doi:10.5194/gmd-12-2657-2019, 2019.

Di Girolamo, P., Cacciani, M., Summa, D., Scoccione, A., De Rosa, B., Behrendt, A. and
Wulfmeyer, V.: Characterisation of boundary layer turbulent processes by the Raman lidar
BASIL in the frame of HD(CP)<sup&gt;2&lt;/sup&gt; Observational
Prototype Experiment, Atmos. Chem. Phys., 17(1), 745–767, doi:10.5194/acp-17-745-2017,
2017.

- Hamamatsu: Characteristics of photomultiplier tubes, in Photomultiplier tubes: basics and
 applications, third edition, Hamamatsu Photonics K.K. Electron Tube Division. [online]
 Available from: https://www.hamamatsu.com/resources/pdf/etd/PMT_handbook_v3aEChapter4.pdf (Accessed 29 April 2021), 2007.
- Hammann, E., Behrendt, A., Le Mounier, F. and Wulfmeyer, V.: Temperature profiling of the
 atmospheric boundary layer with rotational Raman lidar during the
 HD(CP)<sup&gt;2&lt;/sup&gt; Observational Prototype Experiment,





- 780 Atmos. Chem. Phys., 15(5), 2867–2881, doi:10.5194/acp-15-2867-2015, 2015.
- 781 Hayden Smith, W. and Smith, K. M.: A polarimetric spectral imager using acousto-optic
- 782 tunable filters, Exp. Astron., 1(5), 329–343, doi:10.1007/BF00454329, 1990.
- 783 IPCC: Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to
- the Fifth Assessment Report of the Intergovernmental Panel on Climate Change, edited by T.
- 785 F. Stocker, D. Qin, G.-K. Plattner, M. Tigno, S. K. Allen, J. Boschung, A. Nauels, Y. Xia, V.
- 786 Bex, and P. M. Midgley, Cambridge University Press, Cambridge., 2013.
- Kuze, H., Kinjo, H., Sakurada, Y. and Takeuchi, N.: Field-of-view dependence of lidar signals
 by use of Newtonian and Cassegrainian telescopes, Appl. Opt., 37(15), 3128,
 doi:10.1364/AO.37.003128, 1998.
- Lange, D., Behrendt, A. and Wulfmeyer, V.: Compact Operational Tropospheric Water Vapor
 and Temperature Raman Lidar with Turbulence Resolution, Geophys. Res. Lett., 46(24),
 14844–14853, doi:10.1029/2019GL085774, 2019.
- Martucci, G., Navas-Guzmán, F., Renaud, L., Romanens, G., Gamage, S. M., Hervo, M.,
 Jeannet, P. and Haefele, A.: Validation of pure rotational Raman temperature data from the
 Raman Lidar for Meteorological Observations (RALMO) at Payerne, Atmos. Meas. Tech.,
 14(2), 1333–1353, doi:10.5194/amt-14-1333-2021, 2021.
- Navas-Guzmán, F., Martucci, G., Collaud Coen, M., Granados-Muñoz, M. J., Hervo, M.,
 Sicard, M. and Haefele, A.: Characterization of aerosol hygroscopicity using Raman lidar
 measurements at the EARLINET station of Payerne, Atmos. Chem. Phys., 19(18), 11651–
 11668, doi:10.5194/acp-19-11651-2019, 2019.
- Newsom, R. K., Turner, D. D., Mielke, B., Clayton, M., Ferrare, R. and Sivaraman, C.:
 Simultaneous analog and photon counting detection for Raman lidar, Appl. Opt., 48(20), 3903,
 doi:10.1364/AO.48.003903, 2009.
- Prunet, P., Thépaut, J.-N. and Cassé, V.: The information content of clear sky IASI radiances
 and their potential for numerical weather prediction, Q. J. R. Meteorol. Soc., 124(545), 211–
 241, doi:10.1002/qj.49712454510, 1998.
- 807 Sherlock, V., Garnier, A., Hauchecorne, A. and Keckhut, P.: Implementation and Validation of
- a Raman Lidar Measurement of Middle and Upper Tropospheric Water Vapor, Appl. Opt.,
 38(27), 5838, doi:10.1364/AO.38.005838, 1999.
- 810 Sicard, M., Chazette, P., Pelon, J., Won, J. G. and Yoon, S.-C.: Variational method for the





- 811 retrieval of the optical thickness and the backscatter coefficient from multiangle lidar profiles,
- 812 Appl. Opt., 41(3), 493, doi:10.1364/AO.41.000493, 2002.
- 813 Simeonov, V., Larcheveque, G., Quaglia, P., van den Bergh, H. and Calpini, B.: Influence of
- the photomultiplier tube spatial uniformity on lidar signals, Appl. Opt., 38(24), 5186,
 doi:10.1364/AO.38.005186, 1999.
- Totems, J. and Chazette, P.: Calibration of a water vapour Raman lidar with a kite-based humidity sensor, Atmos. Meas. Tech., 9(3), 1083–1094, doi:10.5194/amt-9-1083-2016, 2016.
- 818 Totems, J., Chazette, P. and Raut, J.-C.: Accuracy of current Arctic springtime water vapour
- 819 estimates, assessed by Raman lidar, Q. J. R. Meteorol. Soc., 145(720), doi:10.1002/qj.3492,
 820 2019.
- Vaughan, G., Wareing, D. P., Pepler, S. J., Thomas, L. and Mitev, V.: Atmospheric temperature
 measurements made by rotational Raman scattering, Appl. Opt., 32(15), 2758,
 doi:10.1364/AO.32.002758, 1993.
- Wandinger, U. and Ansmann, A.: Experimental determination of the lidar overlap profile with
 Raman lidar, Appl. Opt., 41(3), 511, doi:10.1364/AO.41.000511, 2002.
- 826 Weng, M., Yi, F., Liu, F., Zhang, Y. and Pan, X.: Single-line-extracted pure rotational Raman
- 827 lidar to measure atmospheric temperature and aerosol profiles, Opt. Express, 26(21), 27555,
- 828 doi:10.1364/OE.26.027555, 2018.
- Whiteman, D. N.: Examination of the traditional Raman lidar technique I Evaluating the
 temperature-dependent lidar equations, Appl. Opt., 42(15), 2571, doi:10.1364/AO.42.002571,
 2003.
- Whiteman, D. N., Melfi, S. and Ferrare, R.: Raman lidar system for the measurement of water
 vapor and aerosols in the Earth's atmosphere, Appl. Opt., 31(16), 3068–82,
 doi:10.1364/AO.31.003068, 1992.
- Whiteman, D. N., Cadirola, M., Venable, D., Calhoun, M., Miloshevich, L., Vermeesch, K.,
 Twigg, L., Dirisu, A., Hurst, D., Hall, E., Jordan, A. and Vömel, H.: Correction technique for
 Raman water vapor lidar signal-dependent bias and suitability for water vapor trend monitoring
 in the upper troposphere, Atmos. Meas. Tech., 5(11), 2893–2916, doi:10.5194/amt-5-28932012, 2012.
- 840 WMO: WMO Oscar : List of all requirements, [online] Available from: https://www.wmo-841 sat.info/oscar/requirements (Accessed 28 April 2021), 2017.





- 842 Wulfmeyer, V., Hardesty, M. R., Turner, D. D., Behrendt, A., Cadeddu, M. P., Di Girolamo,
- 843 P., Schlüssel, P., Baelen, J. Van and Zus, F.: A review of the remote sensing of lower
- 844 tropospheric thermodynamic profiles and its indispensable role for the understanding and the
- simulation of water and energy cycles, Rev. Geophys., 819–895, doi:10.1002/2014RG000476,
- 846 2015.
- 847