- We would like to thank the reviewers for spending time on this paper and for providing an interesting set of comments. The comments have helped the authors and have improved the paper. Please find a point-by-point discussion and answer of the issues raised by the reviewers. To facilitate the work of reviewers and the editor, the reviewer's comments and suggestions are preceding each reply in italic face.
- 6 The corrections are highlighted in yellow in the article (after the author comments).

# 7 <u>Reviewer 1</u>

- 8 All the corrections given by the reviewer have been done in the text.
- 9 1) Please use "rawinsoundings" and rawinsondes" where appropriate.
- 10 Yes, the correction has been done.
- 11 2) Preferably referred as "S", as most commonly used in the lidar literature.
- 12 Yes, but for us, S is also the lidar signal. So, we prefer to use LR for lidar ratio.
- 13 3) not appropriate citations. Please use more recent and well established ones (e.g see in the
   14 ICCP 2014)
- 15 The references have been changed and the IPCC has been cited.
- 17 *4) Please use more recent and well established ones (e.g see in the ICCP 2014)*
- 18 The reference to ICCP (2014) has been added. Nevertheless, even if new references explain
- the role of aerosol, this is not new in atmospheric science. Thus, the earlier references have tobe cited.
- 5) This is not true. It has to be very clearly presented (e.g., put in () which instruments are
  available with what range and time resolution).
- 23 We agree that this sentence is awkward. It was withdrawn.
- 6) This part is too "bavardeux". It has to be completely revised. Please cite more updated
  literature.
- We do not agree, it is not "bavardeux". It is a fast overview of the lidar use for atmospheric moisture survey. As asked by the reviewer in its next remark, we have added a reference to the DWD Ramses lidar.
- 7) Of course this kind of instrumentation exists since the 90s'. and many relevant instruments
  have been developed so far, some of them on an operational level (cite here the existing
  instruments such as the DWD Ramses lidar, the ARM lidar etc.).
- The ARM lidar has been cited in the previous section. This is the reason of this introductive part. Yes, the DWD has to be cited and we have added this quote
- part. Yes, the DWD has to be cited and we have added this quote.

34 35	8) cite here according to which standards				
36 37	The standard has been added (EN 60825-1).				
38	9) not clearly written. pease rephrase				
<ol> <li>39</li> <li>40</li> <li>41</li> <li>42</li> <li>43</li> <li>44</li> <li>45</li> <li>46</li> </ol>	We have added complementary information as also asked by the third reviewer: "The receiver is composed of 3 distinct detection boards using small collector diameters of 15 cm. The total number of detection channels is four. Note that the reason to have separate paths for the two Raman channels is to be able to set-up independently each channel to keep as much flexibility as possible. Hence, we can easily replace a detection board to change the lidar measurements."				
47	10) please correct inside all manuscript				
48 49 50	We have replaced "interferential filter" by 'interference filter" in all manuscript.				
51 52	11) insert "(FWHM)"				
53 54	It has been added.				
55	12) Please provide citations here!				
56	The reference to Bock et al. (2009) has been added.				
57	13) Start a new paragraph here!				
50 59 60	The correction has been made.				
61	14) not clear! please rephrase				
62	We have replace "lower" by "smaller".				
63	15) insert "measurement"				
64	The correction has been done.				
65	16) have already been				
66	The correction has been done.				
67	17) insert "presenting"				
68 69	The correction has been done: "After presenting the lidar calibration and the assessment of the different error sources, we will present a study on a typical meteorological situation using a				

- 70
   synergy between the WVMR and aerosols lidar measurements.".
- 71 18) This statement is false and has to be replaced, as there are a lot of data in the literature.
  72 There are several papers for lidar ratio at 355 nm (Mona et al., 2012; SAMUM papers;

*doi:10.1029/2007JD009028; doi:10.1029/2005JD006190; doi:10.1029/2004GL019881; and many others*)

We agree, the end of the sentence has been removed. The reference to Mona et al. (2012) andBalis et al. (2004) have been added.

19) I do not like this text at all. Please rephrase it completely, referring to the current state of
the art of the UV Raman lidar technique. No need to go back to the early lidar papers (of
the year 1962)

80

This section has been entirely rephrased to highlight the specificity of our system compared to the current state of the art.

- 83 20) Corrections on Fig. 1
- 84

86

91

- 85 The correction has been made (P-pol => p-pol, S-pol => s-pol).
- 87 <u>Reviewer 3</u>
- 1) L43: I think that the references of Melfi et al.(1989) and Kulmala et al. (1993) are not
  suitable because they did not directly study the influence of water vapor to the energy
  balance of the atmosphere. Please cite more suitable papers.
- 92 The references have been changed and the IPCC has been cited.
- 93
  94 2) L79: I don't understand what the "natural" evolution of the lidar is. Please explain it.
- 95
  96 "Natural" is not to consider alone, it is a natural evolution in the frame of scientific programs
  97 HyMeX and ChArMEx, as explained in the text. We have removed "natural".
- 98
- 99 3) L102: Is the word "than" grammatically correct?
- We have corrected the sentence by; "The WALI instrument has been developed at LSCE
  based on the same technology as its precursor instruments LESAA".
- 103
  104 4) L115: Do you use 3 receiving telescope? Please make clear the explanation (also
  105 Fig. 2).
- 106
- Yes, we use 3 receiving telescopes. In fact, the explanation is given later. We have move the sentence and complete it: "The receiver is composed of 3 distinct detection boards using small collector diameters of 15 cm. The total number of detection channels is four. Note that the reason to have separate paths for the two Raman channels is to be able to set-up independently each channel to keep as much flexibility as possible. Hence, we can easily replace a detection board to change the lidar measurements."
- 113

- 114 5) L119: compactedness ! compactness
- 116 The correction was made.

117						
118	6) L127: Do you mean that you use the separate HV supply unit for the Raman nitrogen					
119	and water vapor channels? Please correct the sentence.					
120						
121	The sentence has been corrected: "As separate HV supply units for the Raman nitrogen and					
122	water vapor channels are used, a careful calibration of the relative channel gain versus HV has					
123	to be performed."					
124						
125	7) L145: The word "other' might be necessary before "than" (please check grammar).					
126						
127	The correction has been done.					
128						
129	8) L163: Add the explanation of the lidar ratio (i.e. particle extinction-to-backscatter					
130	ratio).					
131						
132	The explanation has been added.					
133						
134	9) L170: What value of A you used in the study?					
135						
136	We have considered a value of 1 for the error budget. The value has been given in the section					
137	4.					
138						
139	10)L175: "total" should be "unity".					
140						
141	"total" has been replaced by "complete" as asked by the first reviewer.					
142						
143	11) L195: Which altitude is correct of full overlap, 500 m, 700 m (L175) or 200-300 m					
144	( <i>L120 and Table 1</i> )?					
145						
146	As explained, the correct value is 200-300 m, but for the water vapor channel the field					
147	diaphragm did not collect the entire image field in the optical configuration used. This leads to a					
148	degraded overlap factor.					
149						
150	12) L198: How do you correct the spectral dependency of the aerosol extinction be-					
151	tween the two Raman wavelengths?					
152						
153	It is not a major error as explained in the text and the correction is done with an Angstrom					
154	exponent of 1 (see section 4). The aerosol channels of the lidar give the vertical profile of the					
155	aerosol extinction coefficient.					
156						
157	21) L204: What is the reference altitude you used in this study?					
158						
159	The altitude range has been given in the text: "For this study, $z_0$ has been chosen above the					
160	aerosol layers, between 4 and 6 km amsl."					
161						
162	22) L208: What value is used for $beta_E(z0)$ in Eq. (6).					
163						

164 165 166	As explained in section 2.2, the molecular extinction and backscatter coefficients are determined with the polynomial approximation proposed by Nicolet (1984). The value is a function of the altitude.
107	22) In the word "upper independing" well accorded in the community of the atmospheric
168	25) is the word radwindsounding well accepted in the community of the atmospheric
169	science? Please check it.
1/0	
1/1	we have made the correction by "rawinsounding".
1/2	
1/3	24) L259: Please explain the method for determining the overlap factor ratio.
1/4	
175	A complementary explanation has been given in section 3: "K is first assessed using the upper
176	part of the rawinsounding profile and $\xi$ is then retrieved from the lower part (below 0.8 km
177	amsl)."
178	
179	17) L262: What height range or point did you compare the water vapor mixing ratio
180	between the lidar and radiosonde to obtain the calibration constant and how did you
181	compare the ratios (e.g. least square method)? Please explain.
182	
183	We have added the method: "The calibration adjustments have been computed using the
184	minimum of the mean square deviation between the lidar and the rawinsounding profiles."
185	
186	18) L344: Please give a comment on the uncertainty of the overlap function and its
187	influence on the derived water vapor mixing ratio.
188	
189	The discussion about the effect of the overlap function on the WVMR relative error is given
190	in section 4 (Calibration). It leads to a relative uncertainty ~ 4% for altitudes between 0.3 and
191	0.80 km amsl.
192	
193	19) L439:"southwest" ! "southwesterly"?
194	
195	The correction has been made.
196	
197	20) L462: "stronger" should be "higher". Please correct the same word in L476 and
198	L480.
199	
200	The corrections have been made.
201	
202	21) L470. Please explain how the dust plumes destabilized the air masses in more
203	detail.
204	
205	Dust aerosol presence in the atmospheric column impacts the radiative balance and by this
206	way modifies the vertical equilibrium by increasing the convection. This point has been added
207	in the text.
208	
209	22) L518: The paper of Smullin and Fiocco (1962) is not cited in the reference list.
210	
211	We do not cite this paper anymore in our revised manuscript.
212	

- 213 23) Fig. 3: The photograph of the lidar-van is not clear. It would be better if it is replaced
  214 with the close-up of the van.
- 215
- 216 The picture has been resized for better clarity.
- 218 25) Fig. 8 The "S355" in the legend should be "PR355".
- 219

220 The corrections have been done on Figures 4 and 7, and a sentence has been modified in the

text: "The presence of clouds, <u>highlighted on the elastic range-corrected lidar signal S355</u>,
prevents us from verifying the agreement between the instruments over 1.6 km amsl."

223

224 25) Caption of Fig. 12: Add the explanation of a)-f). Are they corresponds to the time 225 periods of Table 2?

- The dates and time are given in the caption of Fig. 12 for each case. We have added the
- reference to Table 2: "The time periods from a) to f) correspond to the ones of Table 2".

228	The mobile Water vapor Aerosol Raman LIdar and its implication in the frame of the
229	HyMeX and ChArMEx programs: application to a dust transport process
230	

231 Patrick Chazette, Fabien Marnas, and Julien Totems

232

Laboratoire des Sciences du Climat et de l'Environnement (LSCE), UMR8212, Laboratoire
mixte CEA-CNRS-UVSQ, CEA Saclay, 91191 Gif-sur-Yvette, France.

235

Abstract.

237

238 The increasing importance of the coupling of water and aerosol cycles in environmental 239 applications requires observation tools which allow simultaneous measurements of these two fundamental processes for climatological and meteorological studies. In this purpose, a new 240 241 mobile Raman lidar, WALI (Water vapor and Aerosol LIDAR), has been developed and implemented within the framework of the international HyMeX/IODA-MED and ChArMEx 242 programs. This paper presents the key properties of this new device and its first applications 243 to scientific studies. The lidar uses an eye-safe emission in the ultra-violet range at 354.7 nm 244 and a set of compact refractive receptors. Cross-comparisons between rawinsoundings 245 246 performed from balloon or aircraft and lidar measurements have shown a good agreement in the derived water vapor mixing ratio (WVMR). The discrepancies are generally less than 247 0.5 g/kg and therefore within the error bars of the instruments. A detailed study of the 248 uncertainties was conducted and shows a 7 to 11% accuracy of the WVMR retrieval, which is 249 largely constrained by the quality of the calibration. It also proves that the lidar is able to 250 measure the WVMR during daytime over a range of about 1 km. The WALI system otherwise 251 provides measurements of aerosol optical properties such as the lidar ratio (LR) or the 252 Page **7** sur **56** 

particulate depolarization ratio (PDR). An important example of scientific application 253 addressing the main objectives of the HyMeX and ChArMEx programs is then presented, 254 following an event of desert dust aerosols over the Balearic Islands. This dust intrusion may 255 256 have had a significant impact on the intense precipitations that occurred over southwestern France and the Spanish Mediterranean coasts. During this event, the LR and PDR values 257 obtained are in the ranges of ~45-63±6 sr and 0.1-0.19±0.01, respectively, which is 258 representative of dust aerosols. The dust layers are also shown to be associated with 259 significant WVMR, i.e between 4 and 6.7 g/kg. 260

261 1 Introduction

By the end of the 21<sup>st</sup> century, climate models forecast a significant increase in the loss of 262 fresh water in densely populated areas. For instance, the decrease of fresh water reserves 263 around the Mediterranean Sea has been assessed to be 40 % higher for 2070-2090 than for 264 1950-1999 (Sanchez-Gomez et al., 2009). These results should be evaluated in the context of 265 rising anthropogenic pressure in the Mediterranean region, with a population growth expected 266 in the range of 300% around the Mediterranean basin within the next 25 years (with more 267 than 500 million inhabitants). The Mediterranean area has thus been identified as a hot-spot in 268 the projections of future climate change (Giorgi and Lionello, 2008) where the water-vapor 269 270 mixing ratio is a key meteorological parameter for the energy balance of the atmosphere (e.g. Held and Soden, 2000; IPCC, 2013). 271

Moreover, it is now known that the cycles of aerosols, clouds and water-vapor are closely coupled within the climate change scenarios. Indeed, water-vapor is involved in the aerosol and cloud formation when aerosols contain hygroscopic components (e.g. *Larson and Taylor*, 1983; *Rood et al.*, 1987; *Radriamiarisoa et al.*, 2006) and thus influences the Earth-Atmosphere radiative balance. Aerosol hydration remains one of the largest sources of uncertainty in the climate models (*Covert et al.*, 1979; *Boucher and Anderson*, 1995; *Haywood et al.*, 1997; IPCC, 2014). Aerosols also lead to a visibility reduction in the atmosphere, which impacts the socio-economical activities. As the densely populated areas of the planet are especially characterized by their vulnerability to changes in the coupled cycles of water and aerosol, precise measurements are now necessary to assess the model uncertainties in both the water-vapor mixing ratio and the aerosol amounts in the lower and middle troposphere.

As written by Whiteman et al. (1992), lidar is a well-established technique for measuring the 284 water-vapor mixing ratio in the atmosphere. Cooney (1970) and Melfi et al. (1969) showed as 285 early as the late 1960's that the Raman lidar is a powerful tool for this measurement and 286 Vaughan et al. (1988) used for the first time Raman lidar to perform water-vapor mixing ratio 287 288 measurements up to the tropopause. Following these pioneer works, Ansmann et al. (1992) performed simultaneously measurement of the water-vapor mixing ratio and aerosol optical 289 290 properties, Turner et al. (1999) used Raman lidar in continuous measurements in the framework of the atmospheric radiation measurement program (ARM), and Veselovskii et al. 291 (2000) also yielded profiles of the water-vapor mixing ratio in the troposphere. More recently, 292 the German Meteorological Service has been equipped with a Raman lidar (*Reichardt et al.*, 293 2012). The differential absorption lidar technique (e.g. Noah et al., 1994; Bruneau et al., 294 2001) could also be used but requires greater instrumental constraints and makes it difficult to 295 comply with eye-safety conditions. Lidar is also an often-used instrument for aerosol survey 296 (Fiocco and Grams, 1964) and particularly Raman lidar (e.g. Melfi et al., 1989; Ansmann et 297 al., 1992; Turner et al., 1999). More recently, an eye-safe, compact and light Nitrogen-Raman 298 lidar has been developed at the Laboratoire des Sciences du Climat et de l'Environnement 299 (LSCE) to track the aerosol pollution around Paris as well as the ash emitted in the 300 atmosphere by the Eyjafjallajökull volcano (Royer et al., 2010; Chazette et al., 2011). The 301 Page **9** sur **56**  evolution of such a lidar, in the frame of the scientific programs Hydrological cycle in the
Mediterranean eXperiment (HyMeX, http://www.hymex.org/) and Chemistry-Aerosol
Mediterranean Experiment (ChArMEx, http://www.mistrals-home.org), was the addition of a
water-vapor Raman channel.

We present in this paper the new transportable eye-safe and mobile Water-vapor and Aerosol 306 Raman LIdar (WALI) that is able to measure simultaneously the water-vapor mixing ratio 307 (WVMR) and the aerosol optical properties with a sufficient reliability for meteorological and 308 climatological studies in the lower and middle troposphere. The first results obtained on the 309 retrieval of the WVMR and aerosol optical properties will be presented and discussed 310 hereafter following the fall campaign of the HyMeX/IODA-MED (Innovative Observing and 311 312 Data Assimilation systems for the MEDiterranean Weather) program. The datasets gathered 313 on aerosol properties also represents the first measurements provided to the ChArMEx program. 314

In a first section, the Raman lidar will be presented along with the experimental set-up. The 315 classical theoretical approaches for the retrieval of the WVMR and aerosol optical properties 316 will be also reminded. For the lidar calibration, a comparison to WVMR vertical soundings 317 performed by rawinsoundings and aircraft measurements will be presented in a second 318 319 section. The main uncertainties will be assessed and discussed in a third section. In a fourth section we will analyze an example of dust event observed in the frame of the 320 HyMeX/IODA-MED and ChArMEx programs. Finally, the conclusions will recall the main 321 characteristics of the instrument and the first results obtained. 322

323 2 Experimental and theoretical tools

The WALI instrument is here described as well as the signal processing used for the retrieval of both the WVMR and the aerosol optical properties. The experimental sites where lidar measurements have been performed are also presented.

327 2.1 Technical characteristics of WALI

The WALI instrument has been developed at LSCE based on the same technology as its precursor instruments LESAA (Lidar pour l'Etude et le Suivi de l'Aérosol Atmosphérique, (*Chazette et al.*, 2005) and LAUVA (Lidar Aérosol UltraViolet Aéroporté (*Chazette et al.*, 2007; *Raut and Chazette*, 2009). It is a home-made instrument mainly dedicated to atmospheric research activities.

The lidar operates with an emitted wavelength of 354.7 nm and is designed to fulfill eye-333 safety standards (EN 60825-1). Its emitter is a pulsed Nd:YAG laser (BRILLANT) 334 manufactured by the QUANTEL company (www.quantel.com). The acquisition system is 335 based on a PXI (PCI eXtensions for Instrumentation) technology with 12 bits digitizers at 200 336 MS/s (Mega sampling by second) manufactured by the National Instruments company. Its 337 main characteristics are summarized in Table 1. During all the experiment the acquisition was 338 339 performed for mean profiles of 1000 laser shots leading to a temporal sampling close to 1 minute. The UV pulse energy is ~60 mJ and the pulse repetition frequency is 20 Hz. 340

A schematic representation of WALI is given in Figure 1. The receiver is composed of 3 distinct detection boards using small collector diameters of 15 cm. The total number of detection channels is four. Note that the reason to have separate paths for the two Raman channels is to be able to set-up independently each channel to keep as much flexibility as possible. Hence, we can easily replace a detection board to change the lidar measurements. Using short focal length refractive telescopes instead of a reflector ensures a low altitude

347 overlap for the lidar and increases its stability, transmittance and compactness. The wide

field-of-view (FOV) ~2.3 mrad allows a full-overlap of the transmission and reception paths 348 beyond ~ 200-300 m. On each channel, optical detection is performed by a photomultiplier 349 tube placed behind an interference filter and a focusing lens. The amplification gain of the 350 351 tube between its anode and cathode is directly linked to the input high voltage (HV) chosen by the lidar acquisition software. HV variation allows optimizing the detection dynamic for 352 both nighttime and daytime measurements (with strong sky background light). As separate 353 HV supply units for the Raman nitrogen and water vapor channels are used, a careful 354 calibration of the relative channel gain versus HV has to be performed. 355

The first board is dedicated to the detection of the elastic molecular, aerosols and cloud 356 backscatter from the atmosphere. Two different channels are implemented on that board to 357 358 detect i) the total (co-polarized and cross polarized with respect to the laser emission) and ii) 359 the cross-polarized backscatter coefficients of the atmosphere. The separation between the two beams is carried out using a beam-splitter and a Brewster plate. The interference filters 360 361 (IF1), with spectral bandwidths of 0.2 nm (FWHM), are manufactured by Barr Associates. This reception channel design is similar to the one used for previous studies on tropospheric 362 aerosols (e.g. Rover et al., 2011; Chazette et al., 2012). The second and third boards are 363 dedicated to the measurements of the inelastic nitrogen (N<sub>2</sub>-channel) and water vapor (H<sub>2</sub>O-364 channel) Raman backscattered signals. They measure the backscattered Stokes component of 365 the inelastic vibrational Raman scattering because this process is much more likely at the 366 typical tropospheric temperatures (compared to the anti-Stokes component of Raman 367 scattering). Such scattering happens at a larger wavelength than that emitted, i.e. ~386.6 nm 368 and ~407.5 nm for N<sub>2</sub>- and H<sub>2</sub>O-channel, respectively. The measured water-vapor Raman 369 signal is ~4 orders of magnitude (~3 orders for the nitrogen Raman signal) less than the elastic 370 backscattered signal. Therefore, the H<sub>2</sub>O-channel was found to require an extremely high 371 rejection of all radiation apart from the Raman Stokes central peak, with a transmission ratio 372 Page **12** sur **56** 

approaching 10 orders of magnitude, assuming a complete rejection of the elastic Rayleigh-373 Mie return (Whiteman et al., 1992; Whiteman et al., 2007). This is done by using a dichroic 374 plate, as drawn in Figure 1, associated to a specific interference filter. The spectral bandwidth 375 of this interference filter (IF3), also built by Barr Associates, is 0.3 nm to optimize the 376 contribution of the rotational lines considering the signal to noise ratio. The N<sub>2</sub>-channel is 377 equipped with both a Brewster plate to decrease the background sky contribution and a Barr 378 Associate interference filter (IF2) with a 0.2 nm spectral bandwidth. Note that, considering 379 the spectral bandwidths of the interference filters used here, the Raman backscatter cross 380 sections do not depend on the atmospheric temperature (Bribes et al., 1976; Penney and Lapp, 381 382 1976; Whiteman et al., 1992).

383 2.2 Lidar signal parameterization

The range corrected lidar signals  $S_{\lambda}$  at wavelength  $\lambda$  of a ground-based lidar situated at the altitude  $z_G$  above the mean sea level (amsl) is given as a function of backscatter coefficient  $\beta_{j,}$ and aerosol (molecular) extinction coefficient  $\alpha_{a(m)}$  against altitude *z* by (e.g. *Measures*, 1984)

$$S_{\lambda}(z) = C_{\lambda} \cdot F_{\lambda}(z) \cdot \beta_{\lambda}(z) \cdot$$

$$exp\left(-\int_{z_{G}}^{z}\left[(1+\eta_{m\lambda})\cdot\alpha_{m}(z')+\left(1+\eta_{a\lambda}(z)\right)\cdot\alpha_{a}(z')\right]\cdot dz'\right)$$
(1)

Where: i) for the elastic channel at  $\lambda = 354.67$  nm (subscript *E* thereafter),  $\beta_E(z) = k_f \frac{3\alpha_m(z)}{8\pi} + \frac{\alpha_a(z)}{LR(z)}$ , is the sum of the molecular (m,  $\beta_m(z) = k_f \frac{3\alpha_m(z)}{8\pi}$ ) and the aerosol (a,  $\beta_a(z) = \frac{\alpha_a(z)}{LR(z)}$ ), with  $k_f$  the King factor of air (King, 1923) and *LR* the lidar ratio (particle extinction-to-backscatter ratio), ii) for the nitrogen Raman channel at  $\lambda = 386.63$  nm (subscript *N* thereafter),  $\beta_N(z) = N_N(z) \cdot \sigma_N^{\pi}$ , with the nitrogen density profile  $N_N(z)$ , and iii) for the water-vapor Raman channel at  $\lambda = 407.5$  nm (subscript *H* thereafter),  $\beta_H(z) = N_H(z) \cdot \sigma_H^{\pi}$ , with the water vapor density profile  $N_H(z)$ .  $\sigma_X^{\pi}$  stands for the Raman differential Page 13 sur 56

backscatter cross section of the nitrogen (x = N) or water-vapor (x = H) channels. Coefficients 395  $\eta_{m\lambda} = \left(\frac{\lambda}{354.67}\right)^{-4.09}$  and  $\eta_{a\lambda}(z) = \left(\frac{\lambda}{354.67}\right)^{-A(z)}$  are used to take into account the spectral 396 dependency effects due to the molecules and aerosols (via the Angstrom exponent A), 397 respectively. Note that only zenithal lidar measurements have been performed during this 398 work.  $C_{\lambda}$  are the instrumental constants.  $F_{\lambda}$  are the overlap functions, which have been 399 400 experimentally measured during the campaign for each channel and shown on Figure 2. The overlap function of the H<sub>2</sub>O-channel has been deduced from both that of the N<sub>2</sub>-channel and 401 the calibration in terms of WVMR hereafter presented. It is not complete under ~0.7 km 402 403 because the field diaphragm did not collect the entire image field in the optical configuration used. Hence, a correction, which is included in the calibration process, has to be applied. 404

The molecular extinction and backscatter coefficients are determined with the polynomial approximation proposed by *Nicolet* (1984) using a reference atmospheric density calculated from ancillary measurements (e.g. *Chazette et al.*, 2012). The uncertainty on the a priori knowledge of the molecular contribution has been previously assessed to be lower than 2% (*Chazette et al.*, 2010). Considering  $k_f = 1$  leads to an overestimation on the molecular volume backscatter coefficient of only 1.5% at 355 nm (*Collis and Russel*, 1976).

### 411 2.2.1 Water-vapor mixing ratio

412 The WVMR ( $r_H$ ) is defined as the mass of water vapor divided by the mass of dry air in the 413 same volume:

414 
$$r_H(z) = \frac{N_H(z)}{N_N(z)} \cdot \frac{M_H}{M_N} \cdot r_N$$
(2)

where  $r_N$  is the nitrogen mixing ratio that can be considered as a constant in the troposphere.  $M_H$  and  $M_N$  are the molar masses of water-vapor and nitrogen, respectively. The WVMR can be directly derived from the ratio of the H<sub>2</sub>O-channel and N<sub>2</sub>-channel as

$$r_H(z) = K \cdot \xi(z) \cdot \frac{S_H(z)}{S_N(z)}.$$

418 
$$exp\left(-[\eta_{mN} - \eta_{mH}] \cdot \int_{z_G}^{z} \alpha_m(z') \cdot dz' - \int_{z_G}^{z} [\eta_{aN}(z') - \eta_{aH}(z')] \cdot \alpha_a(z') \cdot dz'\right)$$
(3)

419 where *K* is the instrumental constant, expressed as

420 
$$K = \frac{C_N}{C_H} \cdot \frac{\sigma_N^{\pi}}{\sigma_H^{\pi}} \cdot \frac{M_H}{M_N} \cdot r_N$$
(4)

421  $\xi$  is the ratio between the overlap factors of the N<sub>2</sub>-Raman and H<sub>2</sub>O-Raman channels 422 gradually approaching unity with altitude and reaching it around 700 m. Both *K* and  $\xi$  have to 423 be assessed during a calibration procedure. In the second part of the equation, the second term 424 represents the atmospheric corrections associated to the spectral dependencies of the 425 extinction properties of both molecules and aerosols.

## 426 2.2.2 Aerosol optical properties

The retrieval of the aerosol optical properties coupled to backtrajectory analyses can contribute to the identification of the air mass origin and to the radiative balance studies above the Mediterranean basin. Those properties are obtained using the following procedure. Firstly, after the correction of the molecular contribution, the aerosol optical thickness (*AOT*) between a reference altitude  $z_0$  and z is derived from the N<sub>2</sub>-Channel by (e.g. *Royer et al.*, 2011)

$$432 \qquad AOT(z_0, z) = \frac{1}{(1+\eta_{aN})} \cdot \left| \ln \left( \frac{S_N(z_0)}{S_N(z)} \cdot \frac{\beta_N(z)}{\beta_N(z_0)} \cdot \exp\left( (1+\eta_{mN}) \int_{z_0}^z \alpha_m(z') \cdot dz' \right) \right) \right|$$
(5)

The reference altitude  $z_0$  can be taken in the upper or lower parts of the lidar profile. For this study,  $z_0$  has been chosen above the aerosol layers, between 4 and 6 km amsl. Hence, the aerosol backscatter coefficient  $\beta_a$  can be directly calculated as

436 
$$\beta_a(z) = \beta_E(z_0) \frac{S_E(z)}{S_E(z_0)} \exp\left(2 \cdot \left[ \operatorname{sgn}(z - z_0) \cdot AOT(z_0, z) + \int_{z_0}^{z} \alpha_m(z') \cdot dz' \right] \right] - \beta_m(z)$$
 (6)

437 Secondly, the AOT can be used in two ways. The first one is via a regularization approach 438 such as the Tikhonov regularization method (*Tikhonov and Arsenin*, 1977), from which the 439 vertical profiles of *LR* and  $\alpha_a$  are derived (e.g. *Royer et al.*, 2011) starting from the matrix 440 form of :

441 
$$AOT(z_0, z) = \left| \int_{z_0}^{z} LR(z') \cdot \beta_a(z') \cdot dz' \right|$$
(7)

442 The second one is via an iterative algorithm using the *Klett* (1985) approach (*Chazette*, 2003;
443 *Royer et al.*, 2011):

444 
$$\alpha_{a}(z) = \overline{LR} \cdot \left( \frac{S_{E}(z)Q(z)}{\frac{S_{E}(z_{0})}{\left(\beta_{m}(z_{0}) + \beta_{a}(z_{0})\right)} + 2 \cdot \overline{LR} \cdot \int_{z}^{z_{0}} S_{E}(z')Q(z')dz'} - \beta_{m}(z) \right)$$
(8)

445 where Q is the correction related to the differential molecular optical thickness calculated 446 from the vertical profile of the molecular scattering coefficient:

447 
$$Q(z) = \exp\left(2\left[k_f \frac{3 \cdot \overline{LR}}{8\pi} - 1\right]\int_{z}^{z_0} \alpha_m(z')dz'\right)$$
(9)

448 The columnar mean lidar ratio  $\overline{LR}$  that is derived from this second way corresponds to the 449 value of LR(z) weighted by the aerosol extinction coefficient profile between *z* and *z*<sub>0</sub>.

The depolarization of the laser beam by aerosols is also a powerful tracer to contribute to the identification of the airmass origins. Taking into account that the channel transmissions are not pure in terms of polarization, the volume depolarization ratio (VDR) is explained as (e.g. *Chazette et al.*, 2012a)

454 
$$VDR(z) \approx \frac{T_1'' \cdot S_{E2}(z)}{R_c \cdot S_{E1}(z)} - \left(1 - T_1''\right) \cdot \left(1 - T_2''\right)$$
 (10)

455  $T_1^{/\prime}$  and  $T_2^{\prime\prime}$  are the parallel transmissions of the total and cross-polarization channels. They 456 were estimated before and after the experiment in laboratory on a specific optical bench 457 (*Chazette et al.*, 2012a). The cross-calibration coefficient  $R_c$  can be assessed by normalizing 458 the lidar signals obtained in a "clean" atmospheric volume with negligible aerosol content:

459 
$$R_c \approx \frac{S_{E2}(z) \cdot T_1^{//}}{S_{E1}(z) \left[ \left( 1 - T_1^{//} \right) \cdot \left( 1 - T_2^{//} \right) + VDR_m \right]}$$
 (11)

460 where the molecular volume depolarization ratio  $(VDR_m)$  was taken equal to 0.3945% at 461 355 nm following *Collis and Russel* (1976). Therefore the particulate depolarization ratio 462 (*PDR*) is computed from

463 
$$PDR(z) = \frac{\beta_m(z) \cdot (VDR_m - VDR(z)) - \beta_a(z) \cdot VDR(z) \cdot (1 + VDR_m)}{\beta_m(z) \cdot (VDR(z) - VDR_m) - \beta_a(z) \cdot (1 + VDR_m)}$$
(12)

#### 464 2.3 Experimental sites

To ensure its mobility, WALI was embedded onboard the Mobile Aerosol Station van 465 (Chazette et al., 2005) also equipped with a VAISALA 200 probe mounted on a mast at 466 ~10 m from the surface. Different experimental sites have been considered to calibrate and 467 test WALI under field conditions. The first one is close to the Paris area at ~30 km South of 468 Paris (48°42'50" N and 2°14'44" E). It is situated east of the Trappes meteorological station 469 where rawinsoundings are performed twice daily. The second one is close to Montpellier (43° 470 37' 14" N and 4° 4' 11" E) in the South of France close to the Mediterranean coast. This site 471 has been selected for the opportunity of launching a simultaneous rawinsounding with the 472 lidar measurements without problem for the air traffic. The third site is the one selected to 473 conduct the HyMeX/IODA-MED fall campaign in 2012 and the ChArMEx summer campaign 474

in 2013. Shown in Figure 3, it is situated on the Balearic island of Menorca (Spain) to catch 475 476 the water vapor amount before the airmasses reach the Spain and French coasts. The lidar-van was operated from a site close to Ciutadella (western part of the Menorca island, 39°60'00" N 477 and 3°50'20"E) for HyMeX and close to Mahon (eastern part of the Menorca island, 478 39°49'32" N and 4°12'30"E) for ChArMEx. Rawinsounding were performed from Palma de 479 Majorca (Majorca Island) at ~100 km southwest from the lidar location. A dedicated 480 calibration flight was also performed over Mahon in the eastern part of the Menorca Island, at 481 about 40 km east of Ciutadella. The main experimental period took place between September 482 10<sup>th</sup> and October 30<sup>th</sup> 2012. 483

484 3 Lidar calibration to retrieve the WVMR

As previously discussed, the vertical profile of the WVMR is retrieved using the ratio 485 between the H<sub>2</sub>O-Raman and N<sub>2</sub>-Raman return signals. Nevertheless, this retrieval is subject 486 to the prior assessment of both the calibration constant K and the overlap factor ratio  $\xi$ . 487 488 Because of the uncertainties on the Raman backscatter cross-section and the difficulty to exactly characterize the optical transmission of the entire lidar detection system, the 489 calibration has been performed comparatively to simultaneous vertical sounding using a 490 well-qualified meteorological probe. K is first assessed using the upper part of the 491 rawinsounding profile and  $\xi$  is then retrieved from the lower part (below 0.8 km amsl). The 492 calibration adjustments have been computed using the minimum of the mean square deviation 493 between the lidar and the rawinsounding profiles. Note that Vaughan et al. (1988) used a 494 calibration on optical bench of each optical element leading to a final precision of 12% on the 495 WVMR. 496

497 For the same purpose, atmospheric water vapor profiles have been monitored in the Paris 498 area, in the Southeast of France, and at Menorca before, during and after the HyMeX

/IODA-MED campaign of the September-October 2012 (www.hymex.org). The calibration 499 procedure has been conducted followings 3 chronological steps. 1) Lidar-derived WVMR 500 profiles have been compared with specific nighttime rawinsoundings carried out by Meteo-501 France on September 1<sup>st</sup> and October 27<sup>th</sup> 2012 close to Paris and Montpellier, respectively. 502 Hence, both K and  $\xi$  have been assessed. 2) Due to the difference of photomultipliers high 503 voltage (HV) used during nighttime (950 and 1000 V for the N<sub>2</sub>- and H<sub>2</sub>O-channels, 504 respectively) and daytime, a specific calibration function has been derived to allow continuity 505 between the lidar measurements performed during night- and daytime, as K evolved against 506 507 HV. 3) Independent rawinsoundings have been used to validate the previous calibrations using day- and night-time measurements performed from air-borne platforms equipped with 508 VAISALA probes. These checks have been made before, during and after the campaign. Note 509 that the WALI final vertical resolution has been fixed to 50 m for this calibration exercise. 510

Calibration. The rawinsounding performed near Montpellier was close to the lidar-van 511 (~100 m), and is thus considered the most relevant mean to calibrate the lidar. It has been 512 performed on October 30<sup>th</sup> 2012 during nighttime at 22:00 local time (LT). The result after 513 calibration with K = 0.066 is given in Figure 4. The presence of clouds, highlighted on the 514 elastic range-corrected lidar signal  $S_{355}$ , prevents us from verifying the agreement between the 515 instruments over 1.6 km amsl. The standard deviation (std) between the lidar- and 516 rawinsounding-derived WVMR is 0.13 g/kg (~2.3%). On the same figure, the rawinsounding 517 station of Trappes has also been used to test the calibration with the same value K = 0.066 for 518 519 a measurement performed in the Paris area (Palaiseau). The agreement is very good under 2 km amsl with a std of 0.2 g/kg as the lidar-van was downwind from the station. Over 2 km the 520 521 discrepancy increases with a std close to 0.5 g/kg. The presence of mid-altitude clouds can explain the difference between lidar and rawinsounding above 2.5 km amsl. 522

High voltage variation during daytime. The diurnal evolution of the calibration coefficient 523 K has been measured by two specific experiments over Menorca during the fall of 2012. The 524 result is shown Figure 5 against the HVs of the N<sub>2</sub>- and H<sub>2</sub>O-Raman channels. During 525 daytime the HVs were close to 850 and 650 V for the  $N_2$ - and  $H_2O$ -Raman channels, 526 respectively. With such values K significantly increases to reach  $\sim 1$ . This calibration has been 527 tested by measuring in the same airmass for HVs from 650 to 1000 V. Moreover, two areas 528 (Menorca and Paris) with different WVMR have been considered as shown in Figure 6. The 529 results are in good agreement with a std between 0.2 and 1 km amsl of ~0.8 and ~0.5 g/kg for 530 Menorca and Paris, respectively. Note that the use of lower HVs leads to a decrease in the 531 accessible altitude range because a lower PMT gain, chosen to avoid saturation by sky 532 background light, decreases the signal to noise ratio. 533

534 Validation using independent rawinsoundings. The validation of the previous calibration has been carried out using measurements from balloon and aircraft. Figure 7 gives 535 536 comparisons between WVMR retrieved from lidar and rawinsounding over the same previous sites of the Paris area, before, and several months after the IODA-MED campaign. The first 537 (second) one is during nighttime (daytime). On September 5<sup>th</sup> 2012 the lidar and 538 rawinsounding comparison leads to a std of 0.83 g/kg for WVMR between 0.3 and 5 km amsl. 539 The stronger discrepancy is mainly due to the airmass variability in the lower part of the 540 profile. The agreement is significantly better on February 19<sup>th</sup> 2013, with measurements 541 performed during daytime. The std is equal to 0.29 g/kg between  $\sim$ 0.5 and 1.2 km. 542

A specific flight was performed above Mahon on October 27<sup>th</sup> 2012 between 08:30 and 09:30 LT. The meteorological probe used on the plane was a VAISALA PTB110-Veriteq SP2000. It delivers the thermodynamic temperature with an uncertainty of 0.15 K, the pressure with an uncertainty of 0.6 hPa and the relative humidity with a relative uncertainty of

5% for the atmospheric conditions encountered in the low and middle troposphere. This leads 547 548 to an absolute uncertainty of 0.67 g/kg on the WVMR. As shown in Figure 8, when compared to the lidar-derived WVMR, the std is 0.55 g/kg for altitudes between 0.2 and 1.2 km amsl, 549 which is close to the error bars. Note that for the lidar, the std is also due to the atmospheric 550 fluctuations during a diurnal average of one hour. Nighttime comparison with the 551 rawinsounding of Palma de Majorca leads to a similar std of 0.48 g/kg. Figure 8 also includes 552 553 comparison to operational modeling. The first output is from ECMWF (European Center for Medium-Range Weather Forecasts, www.ecmwf.int) analyses. The 9 closest grid points from 554 Ciutadella have been considered, showing that the WVMR below 2 km is not fluctuating 555 556 much with a std of 0.22 g/kg. The second model is AROME WMED whose WVMR forecast has been extracted above the ground-based lidar location. It is a mesoscale model based on a 557 three dimensional variational data assimilation system with a horizontal resolution of 2.5 km, 558 559 centered over the western part of the Mediterranean basin for real-time and case-study uses. It has been developed for the preparation of the experimental HyMeX special observation 560 period. It is derived from the operational version of the AROME model (Seity et al., 2011) 561 which is centered over France. Lateral boundary conditions are provided by the global model 562 ARPEGE (Action de Recherche Petite Echelle Grande Echelle). As shown in Figure 8 and 563 still for altitudes from 0.2 to 1.2 km amsl, the comparison to lidar-derived WVMR for this 564 specific case leads to std of 0.51 and 0.81 g/kg for ECMWF and AROME-WMED, 565 respectively. 566

## 567 4 Error estimation

The different sources of uncertainty playing a major role in both the WVMR and the aerosol optical properties retrievals will be analyzed in this section. For the latter, we will consider the results already published by Chazette et al. (2012b) showing the entire methodology for the same type of lidar. The uncertainties in the determination of the WVMR are related to 3 main sources: (i) the shot noise characterized by the signal to noise ratio (SNR<sub> $\lambda$ </sub>) of the lidar system, (ii) the calibration related to rawinsoundings, and (iii) the molecular and aerosol contributions. At the first order, the relative error  $\varepsilon_H$  on  $r_H$  is then given by

576 
$$\varepsilon_{H} \approx \sqrt{\underbrace{\frac{1}{SNR_{N}^{2}} + \frac{1}{SNR_{H}^{2}}}_{\text{Shotnoise}} + \underbrace{\varepsilon_{K}^{2} + \varepsilon_{\xi}^{2} + \varepsilon_{HV}^{2}}_{\text{Calibration}} + \underbrace{\varepsilon_{m}^{2} + \varepsilon_{a}^{2}}_{Atmosphere}}_{Atmosphere}}$$
 (13)

where  $\varepsilon_K$ ,  $\varepsilon_{\xi}$  and  $\varepsilon_{HV}$  are the relative errors due to the calibration constant *K*, the overlap factors and the HV variation, respectively. The relative error associated to the spectral dependency of the extinction properties of molecules (aerosols) is given by  $\varepsilon_m$  ( $\varepsilon_a$ ).

Shot noise. An accurate assessment of the shot noise contribution requires a precise 580 characterization of the SNR. During nighttime such assessment is easier because the photon 581 582 counting mode is active. In that mode, the associated standard deviation is the square root of the returned signal (Measures, 1984). An example is given on Figure 9 for a lidar signal 583 acquired during the night of October 19th 2012 over Menorca with a vertical resolution of 584 15 m. The SNR is assessed for an average lidar profile over 1000 laser shots. The SNR for a 585 larger number of laser shots p can be easily calculated knowing that it is proportional to  $\sqrt{p}$ . 586 For a lidar signal averaged over 20 minutes (20000 laser shots) and using a Monte Carlo 587 approach as in Royer et al. (2011), the uncertainty on the WVMR has been assessed as close 588 to 0.08 (0.32) g/kg between 0 and 2 km (2 and 5 km) amsl. Figure 10 shows an example 589 obtained during the same day for a representative WVMR vertical profile. Such uncertainties 590 are a little lower than the deviations measured during the inter-comparison between lidar 591 592 measurements and rawinsoundings.

**Calibration.** The relative uncertainty on the assessment of the overlap factor *F* is close to 3% 593 594 and comparable to the previous assessment of 5% performed by Chazette (2003) when using the same approach. This leads to a relative uncertainty  $\varepsilon_{\xi} \sim 4\%$  for altitudes between 0.3 and 595 0.80 km amsl. The accuracy and precision of the calibration constant K is closely related to 596 the rawinsounding error that is directly linked to the type of radiosonde used for the 597 rawinsounding. It is not easy to obtain such information from meteorological services. 598 599 Fortunately some papers give the relative uncertainty for some meteorological probes (e.g. *Bock et al.*, 2009). The rawinsoundings performed over Palma de Majorca used VAISALA 600 601 RS92 probes. A discussion on various VAISALA probes has been presented by Agusti-Panareda et al. (2009) following the African Monsoon Multidisciplinary Analysis (AMMA) 602 field experiment in 2006 where numerous rawinsoundings were performed. They used the 603 results of the WMO rawinsounding intercomparison experiment (Nash et al., 2005) and the 604 correction used by Ciesielski et al. (2003) for modeling applications. Such a correction has its 605 606 own uncertainties as explained by Wang et al. (2002; 2008) because it does not take into account the solar heating effect, which affects the measurement during daytime. Moreover, 607 the accuracy is affected by wet and dry biases. The magnitude of the humidity correction is up 608 609 to 5% in the lower troposphere but can reach 20% in the upper troposphere. Ferrare et al. (1995) claim an accuracy of 2-3% with a precision of 2%. Such results have been confirmed 610 by Fujiwara et al. (2003) and Bock et al. (2009) for VAISALA RS80 and RS92 probes. 611 Accounting for all these considerations, we consider here that the relative error on the 612 rawinsounding-derived WVMR is about 6% between 0 and 5 km amsl. Associated with the 613 std between the lidar- and rawinsounding-derived WVMR, the calibration error is  $\varepsilon_K \sim 6.5\%$ . 614 During daytime the effect of the HV variation has to be considered. The uncertainty is here 615

616 mainly due to the atmospheric fluctuations during the HV scanning (~30 minutes). For mean

617 lidar profiles of 1000 laser shots (Figure 6), the additional relative error is high (~10%).
618 During daytime the number of laser shots has to be enhanced (60000 for 1 hour) and this
619 uncertainty should decrease but it is difficult to quantify it. The easiest approach is to compare
620 the lidar-derived WVMR to the one retrieved from daytime rawinsounding as shown in
621 section 3. The calibration error is then around 7%.

Molecules and aerosols spectral dependencies. The third error source is negligible compared to the others. Indeed, the residual molecular contribution can be easily corrected using a climatologic model as in *Chazette et al.* (2003) leading to a very low uncertainty, i.e. less than  $10^{-3}$  g/kg (0.3 g/kg when not corrected). The presence of aerosol layers leads to an error on  $r_H$  close to 0.01 g/kg (for A = 1). Nevertheless, such a contribution can be accounted for after the retrieval of the AOT derived from N<sub>2</sub>-Raman channel.

Synthesis on the WVMR error. Taking into account all the main error sources, the relative 628 VWMR error can be established for 3 different altitude ranges. During nighttime and for a 629 temporal integrated sampling of 20 minutes (20000 laser shots), the relative error on the 630 WVMR is ~8% within the first kilometer (0-0.8 km amsl). It reaches 11% between 2 and 631 5 km amsl. The smaller relative error is between 0.8 and 2 km amsl with a value of  $\sim$  7%. Of 632 course, the transitions are gradual and these values may change depending on the presence of 633 more or less moist air masses in the middle troposphere. During daytime, the same relative 634 error can be reached in the first kilometer but with 1 hour integration time. Actually, for 635 operational purposes, the error on the WVMR can be calculated for each averaged profile 636 knowing the SNR for both the N<sub>2</sub>- and H<sub>2</sub>O-channels. The main error source that could be 637 reduced is the one due to the calibration, which is entirely dependent of both the 638 rawinsounding measurement accuracy and precision. 639

Aerosol optical properties. Uncertainties on the retrieval of aerosol optical properties from similar detection channels have already been well discussed in the scientific literature (e.g. *Chazette et al.*, 2010; *Royer et al.*, 2011). For SNR > 20 as encountered with WALI, the relative uncertainty on the LR is ~5% (~10%) during nighttime (daytime). The relative uncertainty on the VDR and PDR are close to 10% for the encountered AOT > 0.2. The relative uncertainty on the AOT is less than 2%.

646 5 A case study analysis during the HyMeX campaign

WALI was operated during the 2012 fall campaign of the HyMeX program (Special 647 Observing Period 1), between September 17<sup>th</sup> and October 27<sup>th</sup>. After presenting the lidar 648 calibration and the assessment of the different error sources, we will present a study of a 649 typical meteorological situation using a synergy between the WVMR and aerosols lidar 650 measurements. As shown in Figure 11, an intense dust aerosol event was observed from 651 October 17<sup>th</sup> to 20<sup>th</sup>. The VDR highlights two maxima, one on October 18<sup>th</sup> and the other on 652 October 19<sup>th</sup>. This event has been sampled to follow its evolution along time. The different 653 time periods considered are given in Table 2 with the corresponding dust layers and their 654 main optical characteristics. The values of the WVMR are also given, showing a strong link 655 between dust layers and significant water vapor contents. 656

Such an application uses the entire capability of the lidar but needs complementary information. Hence, exogenous modeling material has been used. Airmass backtrajectories have been computed to determine the corresponding transport routes (Figure 12) using the NOAA Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) model (*Draxler and Rolph*, 2003) with 6-hourly archived meteorological data provided from the US National Centers for Environmental Prediction (NCEP) Global Data Assimilation System (GDAS) at the horizontal resolution of 0.5°. The altitude of the trajectory starting point was selected primarily from the lidar/in-situ observation of aerosol layer height. The WVMR along the airmass trajectory was retrieved by using the HYSPLIT model, which calculates the main meteorological parameters (i.e., temperature, relative humidity, pressure) along its trajectories. Note that the WVMRs given by the HYSPLIT model were in good agreement with those of balloon-borne data observed at an adjacent rawinsounding site for each time step of the trajectory (*Yoon et al.*, 2006). The outputs of the ECMWF re-analysis (http://www.ecmwf.int) have also been considered for illustrating the horizontal wind-field.

Before the arrival of the dust event, winds in the lower troposphere are southwesterly with 671 low speeds, of the order of 2-5 ms<sup>-1</sup> (Figure 13). They are associated with a low-pressure area 672 situated in the South-western part of Ireland. They transport an aerosol layer above the marine 673 boundary layer (MBL) (Figure 14) from the Spanish coast. In this layer, the mean LR (PDR) 674 is  $\sim 77$  sr (1%) as it can be expected for this type of pollution particles. The VDR is close to 675 the value of its molecular contribution on the entire sampled atmospheric column (Figure 14), 676 no desert dust aerosol is present. The higher values of the WVMR are located in the MBL 677 (~9-10 g/kg), whose top altitude remained below 0.5 km amsl during all the experiment. A 678 wet layer (>7 g/kg) is also present above the MBL where the polluted aerosols are trapped. 679 680 From the airmass backtrajectories shown in Figure 12, it appears that this layer might be mainly off the Balearic Islands. Note that the rawinsounding from Palma de Majorca shows 681 strong similarities with the lidar-derived WVMR profile between the surface and 5 km amsl 682 (Figure 14). It is therefore very likely that the same air mass was sampled above the two sites. 683 Above 2 km amsl the free troposphere is reached with a wet layer (WVMR ~2-3 g/kg) 684 between 2 and 3.5 km amsl. The aerosol load in this layer is very low and non-depolarizing. 685 During the night of October 17<sup>th</sup>-18<sup>th</sup> 2012, the strong prevailing winds veer to the South, 686

bringing relatively warm and humid airmasses from Sahara to Menorca, because of the

presence of a cut-off over Ireland, which moves East during the event. Thus, Saharan 688 airmasses penetrate over the Mediterranean from the Algerian coast (Figure 12 and Figure 689 13). The Saharan region is the world's major source of natural wind-blown mineral dust 690 aerosol (e.g. *Hamonou et al.*, 1999; *Mona et al.*, 2012) and thus aerosol column burden may 691 be enhanced when wind blows from the African coast. Indeed, the AOT (PDR) increases 692 significantly from 0.1 to 0.18 (0.01 to 0.10) whereas the LR decreases to reach ~ 45 sr , which 693 is a typical lidar ratio value for dust related aerosols (e.g. *Müller et al.*, 2007; *Mona et al.*, 694 2012). Stronger winds (> 10 m.s<sup>-1</sup>) are linked to a higher WVMR (~10 g/kg) in the lower 695 tropospheric layers. Dust aerosols are present in the MBL but the main dust layer is between 696 0.5 and 2.2 km amsl associated with  $r_{H}$ ~6 g/kg that contrasts sharply with the content of the 697 free troposphere. Similar observations can be made for the following day. The presence of 698 important amounts of water vapor in the dust aerosol layers, between 4 and 6.7 g/kg, may 699 contribute to maintain the particles in well-defined vertical structures along their transport for 700 a longer period of time. The static stability of the layer can thus be enhanced as described by 701 702 Kim et al. (2007; 2009). In return, dust plumes can act on the high precipitation events that 703 occurred during the experimental period, by leading to the destabilization of the air masses coming from the sea that crossed the regions of Valencia and Tarragona (Spain) and upstream 704 of Lourdes (France), where 24h accumulated rainfall of ~50 mm occurred on October 20<sup>th</sup>. 705 706 Indeed, the presence of dust aerosol presence in the atmospheric column impacts the radiative balance and by this way modifies the vertical equilibrium by increasing the convection. 707 The atmospheric transport for October 18<sup>th</sup> and 19<sup>th</sup> is presented on Figure 12, Figure 13 and 708 Figure 15. It confirms what has been described for the morning of October 18<sup>th</sup>. As the low 709 moved eastward, the wind weakened and the event ended on October 20<sup>th</sup> (Figure 12 and 710 Figure 15). The end of the event is associated with intense rainfalls related to the high 711 712 humidity of the Saharan airmasses, which underwent subsidence over the Mediterranean Sea, Page 27 sur 56

while it caught water above the sea. Thus, the WVMR may have reached more than 15 g/kg at 713 the ground level (Figure 14). These important amounts of water vapor are to put in parallel 714 with the higher aerosol extinction coefficients, likely due to the hygroscopic growth of certain 715 716 particle types. A mix of various aerosol types could be present in the MBL because the PDR remains high in this layer. During all the events, the PDR varied from 0.1 to 0.19 in the 717 aerosol layers (Table 2). Such values are very close to the ones derived by Müller et al. (2007) 718 with the Raman lidar POLIS in the frame of the Saharan Mineral Dust Experiment (SAMUM) 719 720 with  $PDR = 0.25 \pm 0.08$ . By cons, the LR marks more significant differences between the aerosol layers. In the MBL, the LR derived from our study is found to be generally higher 721  $(\sim 70 \text{ sr})$ , which is in contrast to what can be expected for marine aerosols with a coarse mode 722 of sea salt whose LR ~ 25 sr (e.g. Flamant et al., 2000). Our derived LR value can be 723 criticized for two reasons. Firstly, the altitude range of the MBL is more sensitive than the 724 725 upper ones to the assessment of the lidar overlap function. Secondly, the boundary effects of the regularization method used to retrieve the aerosol optical properties during the night can 726 727 strongly impact the derivation of the optical properties in the lower layers. Note that higher value of the LR was also retrieved on October 17th when no dust event occurred. Again, the 728 result could be questionable because the AOT is low, close to 0.07, and the inversion 729 procedure could not be well constrained. However, error studies show uncertainty of about 730 30% in such cases (e.g. Chazette et al., 2012), which confirms the likely higher values of the 731 LR in the MBL. 732

For the upper layers that contain more probably Saharan dust aerosols only, the LR ranges from 47 to 63 sr. It is significantly variable over the four days sampled. This indicates a high variability of aerosol optical properties and possibly of the particle nature. Different dust sources are activated along the days, as the low moves eastward, resulting in different types of dust particles transported. Moreover, human activities located close to the coast may also Page 28 sur 56

explain a part of this variability. The lidar-derived LR range can be retrieved from the 738 literature. During the African Monsoon Multidisciplinary Analysis (AMMA), Chazette et al. 739 (2007) found LR between 40 and 67 sr at 355 nm for the Harmattan dust layer above Niamey. 740 During the same project, Kim et al. (2009) analyzed the CALIOP measurements and reported 741 a value of  $\overline{LR} \sim 36-38$  sr at 532 nm. Note that the LR generally increases when wavelength 742 decreases. Cattrall et al. (2005) also reported  $\overline{LR}$  values close to 43 sr using supphotometer 743 measurements. Dulac and Chazette (2003) found a  $\overline{LR}$  of 59 sr at 532 nm for a multilayer 744 745 structure with desert, anthropogenic and marine aerosols over the Mediterranean. Moreover, Mattis et al. (2002) used the Raman lidar technique to measure the LR value of elevated dust 746 layers during two episodes over Germany. They report LR values between  $\sim 50$  and 77 sr at 747 532 nm. Again with a Raman lidar, Balis et al. (2004) give LR mainly between 45 and 55 sr at 748 355 nm for dust event over Thessaloniki. 749

750 6 Conclusion

Raman lidar systems are powerful tools for the atmospheric sampling of both water vapor and 751 aerosols with high vertical resolution (between 15 and 50 m). Recent technology 752 improvements of detectors, optics and electronics enable precise and reliable instruments and 753 answer the increasing need to study the cycle of these atmospheric components. Aiming at 754 new scientific and operational capabilities unavailable with current large instruments, the eye-755 safe transportable Raman lidar WALI has been developed with compact refractive telescopes 756 for versatile measurements in the whole troposphere. This paper focused on the simultaneous 757 758 retrieval of the WVMR and aerosol optical properties from the WALI instrument. It insists on the calibration procedure and on the error budget for deriving the WVMR. 759

The original design of the WALI system leads to very good capabilities in terms of low altitude overlap and WVMR retrieval during nighttime, that is to say, with an absolute

deviation from rawinsoundings of less than 0.5 g/kg. The calibration procedure is the main 762 error source for the lidar-derived WVMR, when dealing with a large SNR values. This error 763 is very dependent on the rawinsounding accuracy and precision. It reaches 11% in the MBL 764 and decreases to 7% below 5 km range for a temporal averaging of 20 minutes and a vertical 765 resolution of 15 m. The precision of measurements can deteriorate very quickly thereafter due 766 to the decreasing SNR with altitude. The determination of the water vapor is more difficult 767 768 during daytime, but the measurements have been performed with the same uncertainty for altitude ranges below 1 km using a temporal averaging over ~1 hour. 769

The uncertainties linked to the retrieval of aerosol optical properties are comparable to the
ones of previous Raman mobile lidars developed by our team with a relative error less than
10% for the LR or the PDR retrieval.

To demonstrate its performances for measuring the WVMR and the aerosol optical properties, 773 774 the WALI system has been implemented in the Menorca Island during fall 2012 in the frame of the Mediterranean projects HyMeX and ChArMEx. It has allowed highlighting a strong 775 event of desert dust aerosols associated with high water vapor contents between the 17<sup>th</sup> and 776 20<sup>th</sup> of October 2012. Both the LR and PDR attributed to dust particles are very variable but 777 stay in the range of the variability reported in the literature, between  $\sim$ 45-63±6 sr and 778 0.1-0.19±0.01, respectively. These dust aerosol layers are associated with significant WVMR 779 of  $\sim$ 4-6.7±0.4 g/kg, which may contribute to the important rain falls observed during this 780 period over the Southwestern Europe. 781

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965 Dust Layer: Entrainment Processes and Implication for Aerosol Optical Properties,
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Laser	Nd:Yag				
Energy	60 mJ at 355 nm				
Frequency	20 Hz				
	Elastic total 354.67 nm				
D	Elastic $\perp 354.67$ nm				
Reception channels	Raman-N <sub>2</sub> 386.63 nm				
	Raman-H <sub>2</sub> O 407.5 nm				
Reception diameters	15 cm				
Field of view	~2.3 mrad				
Full overlap	~300 m				
Detector	Photomultiplier tubes				
Filter bandwidths	0.2 - 0.3 nm				
Vartical compline	0.75 m (analog)				
vertical sampling	15 m (photon counting)				
Vertical resolution	~30 m				
Acquisition system	PXI technology at 200 MHz				

967 Table 1: Main technical characteristics of the WALI instrument.

Table 2: Analysis of the dust event of October 17<sup>th</sup> to 20<sup>th</sup> over Menorca: different dust layers
and their LIDAR derived WVMR and aerosol optical properties for 7 different time periods.

Date	Altitude range (km)	AOT at 355 nm (0.25 to 5 km)		$\overline{r_H}$ (g/kg)	IR     (sr)		PDR
17.00.00.02.00.17	0.3-1.0	~0.07	0.04	8.8±0.6	74	67±8	0.01±<0.01
17 00:00-03:00 LT	2-3.5	0.07	<0.01	2.8±0.3	/4	-	-
18.00.00.02.00.17	0.3-0.5	~0.17	0.03	10.0±3	45	66±3	0.1±0.01
18 00:00-03:00 L1	0.5-2.0	~0.17 -	0.09	6.4±0.4	45	47±6	0.1±0.01
19 10:20 14:20 IT	0.3-1	~0.29	0.06	7.9±2	50	-	0.15±0.02
18 10:30-14:30 LT	1.5-3	0.38	0.21	-	59	-	0.16±0.01
19 21:00 24:00 LT	0.5-1.5	~0.20	0.09	5.8±0.4	71	69±7	0.16±0.01
18 21.00-24.00 LT	1.5-4	0.29	0.12	4.1±0.4	/1	63±2	0.19±0.01
10.00.00.02.00.17	0.5-1.5	~0.29	0.09	6.3±0.8	62	58±8	0.12±0.01
19 00:00-03:00 LT	2-3.5	0.38	0.12	4.0±0.2	03	53±2	0.18±0.01
10.02:00.06:00.17	0.3-1.0	~0.46	0.07	7.2±2	60	69±15	0.15±0.02
19 03:00-06:00 LT	1.0-3.5	<sup></sup> 0.46	0.32	4.7±1	60	60±4	0.18±0.01
20.00:00.02:00.17	0.5-1.0	~0.14	0.03	7.6±0.2	E 2	59±7	0.10±0.01
20 00:00-03:00 LT	1.0-2.5	0.14	0.07	6.7±0.5	55	40±2	0.10±0.01

971 The main dust layers are highlighted in gray.



974 Figure 1: Schematic representation of the WALI. The receiver refractive telescopes, located on each path before the field diaphragm, were omitted for clarity. The 3 separate detection 975 976 boards are highlighted with their main components. The emission size is adapted using a beam expander to fulfill eye-safe conditions. The elastic (354.67 nm) detection board is 977 composed of 2 polarization channels: total and cross-polarized. The separation of the radiation 978 over the 2 channels is done using a beam-splitter plate. The N2-Raman detection board 979 (386.63 nm) is equipped with a 386.63 nm working-wavelength Brewster plate to get rid of 980 981 half of the sky-background. The H<sub>2</sub>O-channel detection board (407.5 nm) is equipped with an additional dichroic plate to ensure a total rejection of the fundamental radiation at 354.67 nm. 982





Figure 2: WALI Overlap factors for the N<sub>2</sub>-, H<sub>2</sub>O-channels (left) and elastic channel (right) as
experimentally measured. The colored areas represent the standard deviations of the overlap
factors.



Figure 3: Southern experimental sites selected for both the HyMeX/IODA-MED and the
ChArMEx campaigns. The ground-based lidar-van is shown on the bottom-left. The main
experimental sites are indicated on the map (courtesy of Google Inc.).





Figure 4: WALI WVMR retrieval calibration by comparison to rawinsounding: in Montpellier
on October 30<sup>th</sup> 2012 22:00 LT (up) and in Palaiseau (Paris area) on September 1<sup>st</sup> 2012 01:00
LT (down). The red (blue) areas give the standard deviations around the VWMR mean value
derived from the rawinsoundings (lidar). PTU stands for the ground-based measurements at
~10 m from the surface.





Figure 5: Photomultiplier High Voltage (HV) dependent calibration coefficient K with respect
to N<sub>2</sub> channel and H<sub>2</sub>O channel HVs. The black triangle locates the usual night-time setup.
The white dots represent the automatic selected HVs during daytime.



Figure 6: Test of the WVMR calibration for HVs varying between 650 and 1000 V over the Menorca site (on October 21<sup>st</sup> 2012 from 20:00 to 20:30 LT) (left) and the Paris area (November 8<sup>th</sup> 2012 from 17:45 to 18:15 LT). Both the mean value and the standard deviation (gray area) computed on the WVMR retrieval are derived from lidar profiles associated with the different HVs.







Figure 7: Calibration validation by comparing lidar derived WVMR to rawinsounding in Paris area before IODA-MED experiment on September 5<sup>th</sup> 2012 00:50-01:15 LT (up) and after IODA-MED on February 19<sup>th</sup> 2013 12:30-13:00 LT (down). The red (blue) areas give the standard deviations around the VWMR mean value derived from the rawinsoundings (lidar). PTU stands for the ground-based measurements at ~10 m from the surface.



Figure 8: Comparison of lidar-derived WVMR in Ciutadella to airborne in-situ measurements during a flight over Mahon (with green error bars), Palma rawinsounding, ECMWF (with the standard deviation for the 9 closest modeled profiles from the lidar location) and AROME-WMED model outputs on October 27<sup>th</sup> 2012 08:30 to 09:30 LT. The gray (blue) areas give the standard deviations around the WVMR mean value derived from the rawinsoundings (lidar). PTU stands for the ground-based measurements at ~10 m from the surface.



Figure 9: WALI Signal to Noise Ratio (SNR) as a function of altitude for 3 channels (Elastic,
N<sub>2</sub>-Raman and H<sub>2</sub>O-Raman) for a 1000 shots average lidar profile obtained on October 19<sup>th</sup>
2012 during nighttime over Menorca.



Figure 10: WALI derived-WVMR profile (black) and its associated standard deviation (gray)
averaged over 20 minutes (20000 laser shots) during the night of October 19<sup>th</sup> 2012 over
Menorca.





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Figure 11: WALI lidar derived VDR (up) and WVMR (down) from October 17th to October 1046 20<sup>th</sup> over Menorca. The gray solid line represents ground-based WVMR measurements from a 1047 meteorological probe at  $\sim 10$  m from the surface. 1048



Figure 12: Backtrajectories between the 17<sup>th</sup> and 20<sup>th</sup> October 2012. They have been computed using the Hysplit model (courtesy of NOAA Air Resources Laboratory; http://www.arl.noaa.gov). The wind fields are from GDAS (Global Data Assimilation System, http://www.ncep.noaa.gov/) at the horizontal resolution of 0.5°. The terminal location of the

- 1054 air masses is the site of Ciutadella for the altitudes: 1, 1.5 and 2.5 km amsl. The color bar
- 1055 represents the WVMR along the trajectories.



Figure 13: Wind-field (wind barbs) and WVMR ( $r_H$ ) (color plot) from ECMWF OPERA 0.5° horizontal resolution analysis at 850 hPa level for October 17<sup>th</sup> at 02:00 LT (a), October 18<sup>th</sup> 02:00 LT (b), and October 18<sup>th</sup> 14:00 LT (c).

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Figure 14: Comparison between lidar derived WVMR ( $r_H$ ) and rawinsounding on October 1060 17<sup>th</sup> 00:00–03:00 LT (a), and WALI derived parameters: extinction coefficient ( $\alpha_a$ ), VDR, 1061 WVMR ( $r_H$ ) and LR for sampled times of Table 2, i.e. October 17<sup>th</sup> 00:00–03:00 LT (b), 1062 October 18<sup>th</sup> 00:00–03:00 LT (c), October 18<sup>th</sup> 10:30–14:30 LT (d), October 18<sup>th</sup> 21:00–00:00

- 1063 LT (e), October 19th 00:00-03:00 LT (f), October 19th 03:00-06:00 LT (g), October 20th
- 1064 00:00–03:00 LT (h). The time periods from a) to f) correspond to the ones of Table 2.



Figure 15: Wind-field (wind barbs) and WVMR ( $r_H$ ) (color plot) from ECMWF OPERA 0.5° horizontal resolution analysis at 850 hPa level for October 19<sup>th</sup> at 02:00 LT (a), October 19<sup>th</sup> 08:00 LT (b), and October 20<sup>th</sup> 02:00 LT (c).