Intercomparison of in situ water vapor balloon-borne measurements from Pico-SDLA H₂O and FLASH-B in the tropical UTLS

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- 1516 Abstract

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17 In this paper we compare water vapor mixing ratio measurements from two quasiparallel flights of the Pico-SDLA H₂O and FLASH-B hygrometers. The measurements were 18 made on February 10, 2013 and March 13, 2012, respectively, in the tropics near Bauru, Sao 19 Paulo St., Brazil during an intense convective period. Both flights were performed as part of a 20 21 French scientific project, TRO-Pico, to study the impact of the deep-convection overshoot on 22 the water budget. Only a few instruments that permit the frequent sounding of stratospheric water vapor can be flown within a small volume weather balloons. Technical difficulties 23 preclude the accurate measurement of stratospheric water vapor with conventional in situ 24 techniques. The instruments described here are simple and lightweight, which permits their 25 26 low-cost deployment by non-specialists aboard a small weather balloon. We obtain mixing ratio retrievals which agree above the cold-point tropopause to within 1.9 % and 0.5 % for the 27 28 first and second flights, respectively. This level of agreement for balloon-borne measured stratospheric water mixing ratio constitute one of the best agreement reported in the literature. 29 Because both instruments show similar profiles within their combined uncertainties, we 30 conclude that the Pico-SDLA H₂O and FLASH-B datasets are mutually consistent. 31

32 1. Introduction

Water vapor in the stratosphere plays an important role in the radiative and chemical budget (Shindell et al, 1998, Herman et al, 2002, Loewenstein et al, 2002). Changes in the stratospheric humidity can have a significant impact on the climate and the radiative balance of the Earth atmosphere (Forster et al, 2002, Solomon et al, 2010, Riese et al, 2012). Climate models show that an increase in stratospheric humidity can lead to stratospheric cooling and consequently to a more important ozone depletion (Shindell, 2001, Dvorstov and Solomon, 2001).

40 Regular radiosonde measurements are reliable only in the lower-to-middle troposphere 41 zone, whereas high-precision hygrometers must be employed for stratospheric measurements 42 because this region is so dry. Although, a variety of techniques have been developed for 43 measuring water vapor in the stratosphere, achieving high accuracy measurements of 44 humidity in the stratosphere is far from routine. Current stratospheric measurements of

humidity include: frost-point detection, light absorption using tunable diode laser 45 46 spectrometers and fluorescence (Lyman- α radiation) methods. Usually, *in situ* instruments have a higher precision and a better spatial resolution than remote sensing instruments 47 because the former measurements are performed directly inside the air mass and do not 48 require geophysical inversion. Several balloon-borne measurements to monitor the 49 50 stratospheric water vapor have been conducted since the early 1980's (Kley et al, 2000, Oltmans et al, 2000, Rosenlof et al, 2001, Vömel et al, 2002, Jensen et al, 2005, Read et al, 51 2007, Vömel et al, 2007a, Vömel et al, 2007b, Jensen et al, 2008, Weinstock et al, 2009, 52 Hurst et al, 2011, Berthet et al, 2013, Rollins et al, 2014, Kindel et al, 2015). In some cases, 53 coincident flights have been realized leading to comparisons of in situ water vapor 54 55 measurements (Jensen et al, 2005, Vömel et al, 2007a, Vömel et al, 2007b, Jensen et al, 2008, Weinstock et al, 2009, Hurst et al, 2011, Berthet et al, 2013). However, persistent 56 disagreements remain. For example, (Vömel et al, 2007a), compared in situ balloon-borne 57 measurements of water vapor from several instruments during coincident flights. Comparison 58 59 of in situ water vapor measurements from the CFH hygrometer, the NOAA/CFD aircraft hygrometer and the Harvard Lyman- α hygrometer led to considerable discrepancies up to 60 110%. Differences of ±10 % were found by comparing the FLASH-B and NOAA/CDML 61 water vapor measurements obtained at altitudes of 15 km above the polar stratosphere (Vömel 62 63 et al, 2007b). (Jensen et al, 2008) found that discrepancies between nearly simultaneous water 64 vapor measurements in the TTL (Tropical Tropopause Layer) could reach 2 to 3 ppmv. More generally, in the TTL, the measurements have shown discrepancies larger than 10 %. The 65 main problem for *in situ* measurements of water vapor is contamination by outgassing from 66 the balloon and the instrument structure. Recently, the proper selection of wall materials and 67 68 the judicious positioning of the different elements have significantly reduced this confounding effect. 69

70 The TRO-Pico project, which is funded by the French National research Agency 71 (ANR) for five years, was launched in 2010. The main objectives of TRO-Pico are to combine balloon-, ground-, and satellite-based observations as well as model simulations at 72 different scales to study the impact of deep-convection overshoots on the stratospheric 73 humidity. The balloon campaigns were realized during March 2012 and from November 2012 74 75 to March 2013 in Bauru, Sao Paulo State, Brazil and were hosted by IPMet (Instituto de 76 Pesquisas Meteorológicas). The campaigns were divided into two periods: the SMOP period 77 (six-month observation period) to study the change of water vapor during the overall convective season and the IOP campaign (intensive observation period), occurring during the 78 most intense convective period to study the troposphere-to-stratosphere transport and the 79 stratospheric moistening impact. Both comparison flights discussed here are part of the IOP 80 period. Within both periods, 31 successful water vapor flights were carried on under small 81 zero-pressure balloons from 500 m³ to 1500 m³, or 1.2 kg rubber balloons. Water vapor 82 measurements were performed using two lightweight hygrometers: Pico-SDLA H₂O and 83 FLASH-B. A forthcoming paper will present the meteorological/dynamical analysis of the 84 water vapor measurements linked to specific hydration in the lower stratosphere (S. M. 85 Khaykin, personal communication, 2015). 86

In order to validate the observations, Pico-SDLA and FLASH were launched twice on
the same day within a 3h interval close to the convection overshoot event: March 13, 2012

and February 10, 2013. These two cases will be discussed in this paper. These flights were performed using small weather balloons in order to limit the effect of water outgassing. Only a few instruments can be flown under such small volume balloons to permit regular soundings. Unlike other compact hygrometers where the speed-of-descent prevents accurate measurements, these instruments can measure stratospheric water vapor even during descent under parachutes.

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96 The purpose of this study is to thoroughly evaluate the accuracy of the water vapor 97 measurements preformed during this campaign and to quantify the consistency of the data 98 produced by the two hygrometers. Both Pico-SDLA and FLASH hygrometers are described in 99 the Sec. 2 and the flight train is described in Sec. 3. The *in situ* water vapor measurements in 100 the TTL and lower stratosphere are compared for each of the flights in the Sec.4.

101 2. Instrumentation

102 2.1. The Pico-SDLA H₂O hygrometer

Pico-SDLA H_2O (hereafter Pico-SDLA) is a lightweight spectrometer which measures water vapor using laser absorption spectroscopy (Durry et al, 2008). The probe laser emits at a wavelength of 2.63 µm and has a 1-m path length through ambient air. This hygrometer was flown during a coincident flight with the ELHYSA frost-point hygrometer in March 2011, leading to a stratospheric water vapor measurement comparison (Berthet et al, 2013). Both hygrometers agreed to within 3.5% in the polar stratosphere, which is well below their combined instrumental uncertainties.

The mass of the Pico-SDLA is less than 9 kg, making it suitable as a payload for small 110 stratospheric balloons (500 and 1500 m³). Its design was improved in 2012 in order to meet 111 the requirements of TRO-Pico campaigns. The electronic components are now integrated into 112 a Rohacell box on the top of the cell, which makes the instrument more compact. Figure 1 113 shows the new version of the hygrometer. It uses a distributed feedback (DFB) diode laser 114 115 emitting at 2.63 µm. The water vapor absorption line is scanned by tuning the laser current at fixed temperature. After passing through the ambient-air sample, the laser beam is focused 116 onto an InAs detector using a sapphire lens. The mechanical structure of the sensor comprises 117 carbon fiber tubes to strengthen the overall instrument, especially for the landing with 118 119 parachutes. The instrument is equipped with a TM/TC antenna to transmit the spectrum data 120 to the ground during the flight and to control instrument parameters in case intervention is required. The sensor is able to measure water vapor from the ground to altitudes of 35 km for 121 concentrations ranging from 15000ppmv to less than 1 ppmv. 122

Two different rotation-vibration absorption transitions of water vapor are probed 123 because of the large variation in mixing ratio occurring between the troposphere and the 124 stratosphere. For measurements from the ground to around 200 hPa pressure level, we used 125 the $4_{13} \leftarrow 4_{14} H_2^{16}O$ line at 3802.96561 cm⁻¹. Above 200 hPa pressure level, we use the 126 $2_{02} \leftarrow 1_{01} H_2^{16}O$ line at 3801.41863 cm⁻¹. During in-flight measurements, the switch from one 127 line to the other is automatically driven. Both sets of line parameters are obtained from 128 129 HITRAN 2012 database (Rothman et al, 2013). In HITRAN, the line intensities for these two lines is based on the work by R. A. Toth at JPL (Jet Propulsion Laboratory, NASA) with a 130

relative uncertainty of 2% (see "Linelist of water vapor parameters from 500 to 8000 cm⁻¹" at
 <u>http://mark4sun.jpl.nasa.gov/h2o.html</u>). The water vapor transition is determined prior to the
 launch, thus allowing for automatic selection during in-flight measurements.

The mixing ratio is extracted from the measured spectra using a non-linear least 134 squares fitting algorithm applied to the measured line shape. We use the Beer-Lambert law to 135 136 model the spectrum and use a Voigt profile (VP) to describe the molecular line shape. We found that fitting the VP to the measured spectra vielded residuals consistent with the 137 instrument noise. No evidence of systematic residuals caused by higher-order line shape 138 effects were observed for stratospheric pressures (our region of interest). Figure 2 shows an 139 example of three atmospheric spectra of the H₂O $2_{02} \leftarrow 1_{01}$ line recorded during the February 140 10, 2013 flight in Bauru, at different altitudes in the lower stratosphere (24.24 hPa = 25.2 km; 141 73.60 hPa = 18.4 km; 101.05 hPa = 16.6 km). During this flight, the cold point tropopause 142 143 (hereafter CPT) altitude was approximately 16.7 km. In the upper panel, the black and red lines represent the measurement and fitted results, respectively. The corresponding fit 144 residuals (meas.- fit) are shown in the bottom panel. The standard deviation of the residuals is 145 around 2×10^{-4} and corresponds to the noise level of the measured beam transmission. These 146 residuals do not show any W structure which has been observed when the VP is fit to 147 transitions exhibiting non-Voigt effects such as Dicke narrowing and/or speed-dependent 148 149 effects (Dicke, 1953, Rautian and Sobel'man, 1967, Tran et al, 2007, Boone et al, 2007). Defining the spectrum signal-to-noise ratio (SNR) as the peak absorbance divided by 150 the baseline standard deviation, we find a maximum SNR of approximately 65:1. For the 151 relatively low pressures (20 hPa to 120 hPa) and hence low absorbances encountered in the 152 TTL and in the lower stratosphere the VP provides an accurate representation of the measured 153 spectrum for the noise levels of this spectrometer. At higher pressures (in the troposphere) a 154 more sophisticated line shape may be necessary because the spectrum SNR may reveal 155 systematic deviations from the VP. 156

Several tests were conducted to determine the sensitivity of the fitting procedure to the
baseline interpolation, as well as to the temperature- and pressure-measurement uncertainties.
These tests were realized using a synthetic spectrum with a noise level equivalent to the inflight spectra. Details of these tests are given below.

The absorption spectrum is extracted from the atmospheric spectra by removing 161 structure in the baseline which is induced by optical components and vibrations of the optical 162 cell. The baseline is interpolated using a polynomial combined with a sinusoid term which 163 164 takes into account commonly observed interference fringes caused by Fabry-Pérot effects between optics. The quality of the fitting procedure is influenced by the spectrum SNR, the 165 polynomial order and the number of points chosen for the interpolation. The combined 166 uncertainty introduced by these different factors varies with the peak absorbance of the line 167 and consequently with the pressure level from 4.5 % at 50 hPa to 0.7 % at 150 hPa. 168

The air pressure is measured using a Honeywell absolute pressure sensor, which operates between -40°C and +85 °C with a manufacturer-specified relative uncertainty of 0.05% full scale (0.7 hPa). The pressure measurements are corrected for drift caused by changes in temperature. During the TRO-Pico campaign flights, the atmospheric temperature ranged from -85 °C to +35 °C. In order to eliminate measurement error caused by being outside the instrument's temperature operating range, the pressure sensor is placed inside an enclosure having a minimum temperature of 0 °C. The uncertainty in the fitted water vapor
concentration caused by temperature-dependent sensitivity of the pressure sensor temperature
is estimated to be ~0.05%.

The temperature is measured using three SIPPICAN thermistors which are coated to 178 limit solar radiation effects. These sensors are located on each end and at the center of the 179 180 optical cell, providing an average temperature for the measurements. The rotation of the optical cell during the flight induces a temperature difference between the three thermistors, 181 which varies from 0 to 5 °C. This depends on the solar exposure of the thermistors (in the case 182 of daytime flights). For this reason, we select the lowest measured temperature for the data 183 processing. Each sensor was calibrated independently by the manufacturer between -90 °C 184 and +50 °C. The uncertainty of the temperature is specified to be 0.3 °C, yielding a 0.25 % 185 uncertainty in the measured sample concentration. 186

By taking into account all sources of error that we can estimate (i.e spectroscopic and experimental errors, as well as error due to spectra processing), the combined relative standard uncertainty ranges from 7.5 % to 3.5 % in the TTL and the lower stratosphere, depending on the local conditions. Since temperature and pressure are input variables for the mixing ratio retrievals, we investigated the consistency of these measurements. We compared the Pico-SDLA measurements with those of a Vaisala RS-92 radiosonde during one coincident flight on January 18, 2013. Details of this work are provided in the next section.

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2.1.1. Temperature and pressure measurements comparison on January 18, 2013

On January 18, Pico-SDLA has been launched at 22:11 UTC under a 1500 m³ balloon. The time is recorded in UTC using a GPS-disciplined clock located onboard the Pico-SDLA. One measurement is made every 300ms to 500 ms depending on the signal-to-noise ratio of the measurements and on the vertical speed of the payload during the flight. The measurements start as soon as they are requested by the operator, independently of launch time. The RS-92 radiosonde, attached to the same balloon, detects the launch time and records it as *t*=0. Thereafter, it takes one measurement every 10 seconds.

The data were synchronized by applying a small temporal offset to the time stamps. This offset was determined from the cross-correlation of the temperature profiles from both sensors and corresponded to the maximum of the cross-correlation.

We calculated the mean temperature difference (mean ΔT), the mean pressure 205 difference (mean ΔP) as well as the standard deviations of the differences $\sigma(\Delta T)$ and $\sigma(\Delta P)$. 206 We only used the ascent measurements for the comparison. Although the descent of the Pico-207 SDLA occurs under parachute, this is not the case of the radiosonde which remains attached 208 to the balloon. The vertical speeds of both sensors are consequently different therefore 209 210 precluding correlation with time. Since only the descent measurements of Pico-SDLA are usable, the radiosonde is never attached to Pico-SDLA during this time. Indeed, the RS-92 211 telemetry system at 403 MHz induces a modulation of the laser emission, which creates two 212 sidebands on the spectrum rendering them unusable. 213

The temperature uncertainty on the RS-92 is 0.5 ° C while the pressure uncertainty is quoted by the manufacturer for two pressure ranges : 1080hPa to100 hPa and 100hPa to 3 hPa, for which the combined standard uncertainty is 1.5 Pa, and 0.6 hPa, respectively.

The mean ΔT for this flight is 0.12 ° C with a standard deviation $\sigma(\Delta T)$ of 0.28° C. 217 The mean ΔT is less than both uncertainties. The ΔT is always lower than 0.5 ° C except 218 above 23 km where the RS-92 exhibited large spikes in the measured temperature. Therefore 219 for this flight, we concluded that the RS temperature was unreliable above this altitude. The 220 SIPPICAN and the RS-92 measurements agree well with the observations of (Nash et al, 221 222 2010; Bower and Fitzgibbon, 2004) which were obtained by comparing different types of temperature sensors. In these studies, the comparison of temperature measurements, using 223 corrected data, lead to temperature differences up to 0.4 ° C during night flights and 1 ° C for 224 daytime flights. The differences are usually higher above the tropopause, which is probably 225 226 due to icing of the sensor.

227 The mean pressure difference (mean ΔP) and the standard deviation of this difference $\sigma(\Delta P)$ are -0.024hPa and 0.163hPa respectively. This pressure difference is below the 228 uncertainties of both the Pico-SDLA and RS-92 pressure sensors. Between the ground and 2.6 229 km, the pressure differences are as large as 0.5 hPa. This behavior was also observed in the 8th 230 231 WMO High Quality Radiosonde Intercomparison (Nash et al, 2010). During this campaign, the performance of radiosonde systems' pressure measurements was investigated. It was 232 found that the pressure differences ranged from 0 to 1.4 hPa and correlated with the altitude of 233 the balloon. The biggest differences occurred near the ground. 234

We determined the consistency of measurement pairs using the GRUAN (Reference Upper-Air Network) analysis approach detailed by (Immler et al, 2010). Given two independent measurements m_1 and m_2 and their respective uncertainties u_1 and u_2 , these two measurements can be considered as consistent if: $|m_1 - m_2| < k\sqrt{u_1^2 + u_2^2}$. Here, *k* is the statistical significance factor. For *k*=1 and if the condition is true, the measurements are consistent.

For measurements of temperature and taking into account each sensor uncertainty, we find that $k\sqrt{u_1^2 + u_2^2} = 0.58$. Thus, to be consistent, the measurements of absolute difference, expressed as $|m_1 - m_2|$, must be lower than 0.58. The mean temperature difference, calculated from in situ measurements, is 0.05° C ±0.15 ° C. Likewise for pressure, the mean ΔP has to be less than 0.92 hPa. In our case, the mean pressure difference is 0.02hPa ± 0.11 hPa. For both parameters, the condition is satisfied and therefore, the measurements are consistent following the GRUAN approach.

248 2.1.2. Water

2.1.2. Water vapor outgassing

Contamination of water vapor measurements caused by outgassing from the balloon 249 envelope or instrument surfaces was first observed in the in situ measurements of 250 (Mastenbrook, 1968) and (Zander, 1966). For the TRO-Pico campaign, the use of small-251 volume weather balloons (1500 m^3 or 500 m^3) is expected to reduce the water vapor 252 outgassing from the balloon envelope. We found that the ascent mixing ratio reached as high 253 as 25 ppmv, whereas the mean stratospheric mixing ratio was 4 ppmv. Therefore, we used 254 only the measurements obtained during descent. We compared these measurements from 255 Pico-SDLA with those of FLASH-B to determine whether or not outgassing of water vapor 256 contaminated the data. As described in detail in the following section, we found that the 257 FLASH-B descent measurements did not suffer from outgassing contamination. During the 258

beginning of the descent, a small contamination (up to 0.5 ppmv and visible up to 3 km below
the float altitude) of the Pico-SDLA data was observed. Therefore, we considered the PicoSDLA data below the altitude where the contamination is observed.

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263 2.2. The FLASH-B hygrometer

The balloon version of FLASH is a compact lightweight sonde developed at the 264 Central Aerological Observatory, Russia, for balloon-borne water vapour measurements in the 265 upper troposphere and stratosphere (Yushkov et al, 1998). The instrument is based on the 266 267 fluorescent method (Kley and Stone, 1978; Bertaux and Delannoy, 1978), which uses the photodissociation of H₂O molecules exposed to vacuum ultraviolet radiation (λ < 137 nm) 268 followed by the measurement of the fluorescence of excited OH radicals using a Hamamatsu 269 photomultiplier in photon-counting mode. The intensity of the fluorescent light sensed by the 270 271 photomultiplier is directly proportional to the water vapor mixing ratio under stratospheric conditions (10-150 hPa). The H₂O measurement range is limited to pressures lower than 272 300hPa to 400 hPa because of strong Lyman-alpha absorption in the lower troposphere. The 273 274 instrument uses an open optical layout, where the analyzed volume is located outside the 275 instrument. This design allows reduction of the instrument size to that of a small sonde with a total mass(including batteries) of about 1 kg. This arrangement restricts the use of the 276 277 instrument to night-time only.

278 Each FLASH-B instrument is calibrated in the laboratory against a reference dewpoint hygrometer, MBW 373L. A description of the procedure can be found in (Vömel et al, 279 2007b). The detection limit for a 4-s integration time at stratospheric conditions is 280 approximately 0.1 ppmv, while the accuracy is limited by the calibration error amounting to a 281 relative uncertainty of 4 %. The typical measurement precision in the stratosphere is 5 % to 6 282 %, whereas the combined relative uncertainty in water vapor concentration is less than 10 % 283 throughout the stratosphere. The FLASH-B has been successfully used in a number of balloon 284 campaigns (e.g., LAUTLOS-WAVVAP, SCOUT-AMMA,TC4, LAPBIAT-II) which 285 included simultaneous measurements of stratospheric water vapor by different measurement 286 techniques. In particular, point-by-point comparison with the frost-point hygrometer from the 287 288 NOAA CMDL showed a mean deviation of 2.4 % with 3.1 % standard deviation (1σ) (Vömel et al., 2007a), and comparison with CFH showed a mean deviation of 0.8 % with a 4 289 % relative standard deviation (Khaykin et al., 2013). 290

The flight configuration of the FLASH-B, in which the analyzed volume is located 291 beneath the downward-looking optics 2-3 cm away from the lens, caused noticeable self-292 contamination during balloon ascent because of water outgassing from the instrument surfaces 293 and balloon. The contamination effect is observed as a quasi-exponential growth of water 294 vapour readings above about 70 hPa during the ascent. This occurs because the relative 295 296 contribution of water carried on the sounding equipment surfaces becomes more significant as the number density of ambient water molecules decreases with altitude. In contrast, the 297 FLASH-B measurements during the descent at the bottom of the flight train in undisturbed air 298 are free of contamination as shown by the reduction in water vapour readings immediately 299 300 after the burst of balloon. Here we use the contamination-free descent profiles along with the 301 clean ascent profiles below 75 hPa.

302 **3. Balloon Flight trains**

The flights have been realized under small zero pressure balloons of 500 m³ and 1500 m³ volume for Pico-SDLA instruments and 1.2 kg rubber balloons for the FLASH instruments. The launch of these balloons was realized by the French scientific team, assisted by staff from IPMet.

307 During the SMOP period, regular soundings of the UT-LS using the Pico-SDLA H_2O 308 spectrometer were conducted by the technicians of IPMet without the presence of the French 309 scientific teams. The hygrometer operation was simplified to permit its deployment by non-310 specialists. During this period, the hygrometer was deployed under 500 m³ zero pressure 311 Aerostar balloons.

For all flights, the flight train includes a parachute, a cutter device and a balloon telemetry/remote control system (E-track iridium), a strobe light and a radar reflector. The cutter device is used to separate the payload from the balloon, with the payload descending under the parachute. The E-Track iridium allows one to follow the flight train during the ascent and the descent and to initiate separation from the balloon. The scientific instrument is connected to the flight train by a nylon driss. The flight trains were easy to implement and permitted quick deployment of the instruments with respect to larger balloons.

For the water vapor flights of Pico-SDLA, the instrument was located at least 15 m below the balloon to limit outgassing from the balloon envelope. On March 13, 2012, the instruments of the flight train, from bottom to top, were the Pico-SDLA H₂O, and the LOAC Optical Particle Counter. The total payload weight for this flight was 15 kg under a 500 m³ balloon. On February 10, 2013, the instruments of the flight train, from bottom to top, were the Pico-SDLA H₂O and the Pico-SDLA CH₄, The total payload mass was 25 kg under a 1500 m³ balloon.

For flights of the FLASH-B, the E-Track box and cutter device were not included in the flight train. The instruments of the flight train were from bottom to top, the FLASH-B and the COBALD (Compact Optical Back-scatter Aerosol detector) backscatter sonde, on March 13, 2012, and FLASH-B, COBALD and LOAC on February 11, 2013. The overall payload masses were 7.4 kg and 9.4 kg, respectively.

331 4. Comparison of mixing ratio retrievals

332 4.1. Flight conditions

The flights of February 10-11, 2013 and March 13, 2012 were intended to capture the signature of the overshoots in water vapor profiles. The launch site was located on the UNESP Bauru Campus, at the outskirts of town (Coordinates: 22.36 °S, 49.03 °W).

On February 10, 2013, the Pico-SDLA was launched at 21:03 UTC with overshooting conditions observed by the IPMet S-band radar located 200 km east of Bauru. Subsequently, a convective cell reached an altitude of >16 km, which was about 150 km east of the launch site position. On this day, the most intense convective events occurred between 18:06 and 21:15 UTC. The FLASH-B hygrometer was launched at 0:09 UTC, 3 hours later than Pico-SDLA.

On March 13, 2012, Pico-SDLA H₂O was launched at 19:20 UTC in convective conditions and FLASH-B was launched 3 hours later. On this day, strong convection was observed until 21:00 UTC with convective cells reaching altitudes exceeding 18 km. Both
 instruments were able to catch the signature of an overshooting cell reaching 19.2 km.

345 During the descent, the vertical speed of the instruments ranged from 60 m/s (just after 346 the flight train separates from the balloon) to 20 m/s in the TTL. In this condition, the Pico-347 SDLA spectra were recorded without any averaging or a maximum average of 5 spectra in 348 order to achieve good vertical resolution and to avoid excessive overlapping of mixing ratio 349 measurements from different layers of the TTL.

350 *4.2. February 10-11, 2013*

351 Figure 3 shows the balloon trajectories of both instruments. On this plot we show the 352 descent trajectory of Pico-SDLA and both the ascent and descent trajectory of FLASH-B wherever the ascent measurements of FLASH can be considered. The altitude of the 353 trajectories is color coded. Altitudes between 14 km and 28 km are considered, representing 354 the TTL and lower stratosphere, which are our regions of interest. For both instrument 355 trajectories, the time is indicated in UTC. The ascent of Pico-SDLA lasted 1h41 min followed 356 by a float of 7 min at 27.4 km before a 37-min-long descent. The ascent of FLASH-B lasted 357 1h 31 min followed by a descent of 47 min. The maximum altitude reached by the balloon 358 was 28.75 km. We can see in Fig. 3 that Pico-SDLA flew 25 km south of FLASH-B which 359 360 resulted in some small differences in the observed water vapor enhancements.

In the case of Pico-SDLA, we use the water vapor measurements below 23 km 361 because a small outgassing effect (~0.4 ppmv) is observed above this height. The balloon 362 carrying the FLASH-B flight-train is much smaller than the 1500 m³ balloon used for Pico-363 SDLA. Since such a small balloon limits water vapor outgassing, we can also consider the 364 FLASH-B ascent profile up to approximately 18 km of altitude, above which a small 365 outgassing effect starts to be observed. This leads to the comparison shown in Fig.4. In this 366 figure, we compare in situ water vapor measurements between 15 km and 24.5 km from Pico-367 SDLA H₂O and FLASH-B. The lower boundary of the TTL is defined in (Fueglistaler et al, 368 2009) as the area above the level of the mean convective outflow (~ 14 km). The upper 369 boundary is set at 70 hPa (18.8 km), above which the atmosphere is governed mainly by 370 stratospheric processes. In Fig.4, the upper boundary of the TTL (green dot line) corresponds 371 to an altitude of ~18.8 km. The CPT of each instrument is determined from the descent 372 373 temperature profiles and is shown by the orange and brown dashed lines. In the case of Pico-SDLA, the CPT is 16.63 km (-74.15 °C) and for FLASH it is 16.98 km (-75.2 °C). This 374 altitude corresponds to the level of the minimum temperature and has an important role in the 375 troposphere-to-stratosphere coupling and exchange. The water vapor transport from the 376 troposphere to the stratosphere is partially dependent on the thermal characteristics of the CPT 377 (Holton et al, 1995, Mote et al, 1996, Kim and Son, 2012, Randel and Jensen, 2013). Indeed, 378 the coldest temperature encountered during the slow ascent partially determines the amount of 379 dehydration of the air mass entering the stratosphere. The amount of water vapor and the 380 temperature determine the relative humidity with respect to ice (RHi). At a given specific 381 humidity, the coldest temperature will correspond to the highest RHi, thus the highest 382 potential to nucleate ice particles that can fall, leading to a dehydration of the air entering the 383 stratosphere. The altitude difference between the CPT altitudes from Pico-SDLA and FLASH 384 can be attributed to three different factors : a natural temporal and spatial temperature 385

variability in the TTL, the measurements uncertainties and the temperature profile behavior in
the TTL which complicates the determination of the CPT : for this flight and the March 13,
2012 flight, the temperature profile in the TTL is quite constant with temperature variations of
less than 0.3 °C, within the sensors uncertainties. Then the CPT altitude is determined using
small structures in the profile.

391 Analyzing the profile comparison in more detail, we find that the main structures are well captured by both instruments above and around the CPT, although the amplitude of the 392 local maxima/minima sometimes varies slightly. Three water vapor enhancement structures 393 appear on the descent profile of Pico-SDLA at altitudes of 16.5 km, 17.2 km and 18 km. The 394 structure at 16.5 km is captured by FLASH during the ascent but not during the descent and is 395 396 shifted downwards by about 90 m in altitude compared to the Pico-SDLA. The amplitude of the enhancement is, in the case of FLASH, about 0.5 ppmv and around 0.68 ppmv for the 397 Pico-SDLA. During the descent, the structure at 17.2 km was captured by FLASH-B at the 398 same altitude and shifted up by 50 m. The descent profile of FLASH-B does not show any 399 400 structure at this altitude. For both instruments, the amplitude of the enhancement is similar but the structure is slightly thicker in the case of FLASH (nominally 560 m) instead of 500 m for 401 Pico-SDLA. The structure at 18 km was captured by FLASH-B at the ascent but not at the 402 descent. Because of a small amount of outgassing, the profile above 17.7 km cannot be 403 404 considered. Nevertheless, structures are visible. The small altitude difference is of the same 405 order of magnitude as the GPS height uncertainty. It also must be considered that the hygrometers did not fly at exactly the same time. 406

Over the altitude range between 15 km and 23 km, comparison between the ascent of 407 FLASH-B and the descent of Pico-SDLA leads to a mean difference of (0.13 ± 0.33) ppmv. In 408 409 the same altitude range, the comparison between the descent profiles of both instruments yields a mean mixing ratio difference of (0.08 ± 0.39) ppmv. FLASH-B is dryer than Pico-410 SDLA by 0.08ppmv at the descent. Considering the 4.1 ppmv mean mixing ratio over the 15 411 to 23 km altitude range, the differences observed correspond to 1.9 % (with a 1- σ standard 412 deviation of 9.5%). Restricting our comparison to above the CPT, the mean difference is then 413 (-0.13 ± 0.15) ppmv (1- σ standard deviation: 3.7 %). We clearly see the impact of the 414 humidity variability in the lower TTL region on the statistical results. The strong humidity 415 variability induces a larger standard deviation and therefore less precise results. Although 416 417 both instruments were flown 3 hours apart, the measurements are in good agreement.

418 4.3. March 13, 2012 flight

Figure 5 shows the trajectory plot for Pico-SDLA and FLASH flights on March 13, 2012. As in Fig. 3, the altitude of the trajectories is color coded and the time is in UTC. Pico-SDLA flew22 km to the west of FLASH. The ascent of Pico-SDLA lasted 1h49 min followed by a float of 14 min at 23.6 km before a 40-min-long descent. The ascent of FLASH-B lasted 1h 15 min followed by a descent of 1h 12 min. The maximum altitude reached by FLASH was 21.6 km.

The comparison of water vapor mixing ratio profiles from FLASH and Pico-SDLA between 21.3 km and 15 km is shown in Fig.6. For this case, FLASH-B water vapor measurements are usable up to 21.3 km. Up to this altitude, Pico-SDLA measurements do not show any outgassing effects. In this figure, the CPT from Pico-SDLA (orange dashed line) and FLASH (brown dashed line), are located at 17.95 km and 17.44 km respectively. The
upper boundary of the TTL is shown with a green dotted line at 70 hPa pressure level,
corresponding to an altitude of 18.6 km. The temperature profiles are also shown in orange
and brown lines. The CPT is much colder in this case than for the February 10 flight (-79 °C
in average instead of -74.6 °C).

434 The RS-92, integrated into FLASH, measures the geopotential altitude whereas the GPS onboard Pico-SDLA measures the geometric altitude, inducing a shift of 378 m in 435 altitude. To correct for this difference, we used the altitude measurements from the COBALD 436 backscatter sonde which are obtained from a GPS. Thus, we were able to reconstruct the 437 FLASH altitude scale by interpolating the COBALD data with respect to the time into flight. 438 439 In this case, a (188 \pm 7) m altitude difference is still observed between Pico-SDLA and FLASH water vapor mixing ratio profiles. Although the origin of the shift is not fully 440 understood, one possible explanation is an initialization on FLASH or COBALD error at 441 launch time. Because the Pico-SDLA and the E-track iridium GPS measurements agree to \pm 442 443 20 m between the CPT and 21.3 km, this excludes an error coming from Pico-SDLA GPS altitude measurements. In Fig. 6, a 188 m shift was applied to the FLASH profile. This shift 444 was determined maximize the correlation coefficient between both profiles. We emphasize 445 that the March 13, 2012 case was the only one where such a high difference in altitude was 446 447 observed.

448 Applying the 188 m shift leads to a mean mixing ratio difference of (0.02 ± 0.21) 449 ppmv between 15 km and 21.2 km between descent profiles. In this case, Pico-SDLA H₂O is 450 dryer by 0.02 ppmv. Considering the mean mixing ratio, around 4.3 ppmv, the relative 451 difference represents ~ 0.5 % (with 1- σ standard deviation of 4.6 %). This shows the excellent 452 agreement between the FLASH and Pico-SDLA measurements, which were always within 453 instrumental uncertainties despite the fact that both instruments were flown 3 h apart.

454 This profile comparison showed identical structures (at 17.4 km, 18.1 km and 18.7 km of altitude) and mostly with a similar amplitude. Also, the altitude ranges of these structures 455 are very close. The local maximum at 18.1 km (Fig. 6) stands out with a mixing ratio of 4.09 456 ppmv in both Pico-SDLA and FLASH measurements. The structure is a little bit thicker for 457 Pico-SDLA (300 m) than for FLASH (200 m). Also, besides the maximum value being 458 identical for both instruments, the amplitude of the water vapor enhancement is slightly 459 460 higher for Pico-SDLA(about 0.8 ppmv) whereas FLASH-B shows a 0.65 ppmv enhancement. An airmass trajectory analysis by (S. M. Khaykin, personal communication, 2015) shows that 461 this enhancement is caused by a hydration from overshooting convection, which is about 65 462 km away from the balloons. The differences in the amplitude of the signal by both 463 instruments can easily be explained by the difference of time of the flights with respect to 464 very local/short duration process. As a result, the instruments cannot sample the same process 465 amplitude. Figure 5 shows the trajectory of both balloons, highlighting the relatively close 466 trajectories which are slightly shifted in space. This helps account for the slight differences 467 between the two profiles. Investigating another large water vapor enhancement at 18.7 km, 468 both instruments measure the same local maximum of 4.19 ppmv. Both the vertical amplitude 469 of the signal (500 m) and the amplitude of the enhancement based on the difference between 470 471 the bottom of this layer and the local maximum is very similar ~ 1 ppmv. (S. M. Khaykin, 472 personal communication, 2015) shows that this enhancement is due to large scale mid-latitude air intrusion, bringing higher mixing ratios of water into the tropical regions. However it
should be noted that the shape of this enhancement is sharper for Pico-SDLA than for
FLASH-B. No significant patterns are highlighted above this layer (~19 km) and both
instruments report very similar mixing ratios.

477 5. Pico-SDLA/FLASH-B correlation

Figure 7 shows a scatter plot comparison of Pico-SDLA versus FLASH water vapor measurements for the flights of March 13, 2012 and February 10, 2013. The data are color coded by pressure in the altitude range from the CPT altitude up to the altitude free of outgassing for each flight. A linear fit of the Pico-SDLA versus FLASH data is shown as a solid line.

We have calculated the Pearson's *r* coefficient from 15 km and from the CPT altitude. This coefficient is calculated from the linear least-squares fitting of the scatter plot data and represents the correlation coefficient.

For the February 10, 2013 flight results, r=0.92 for the 15 km to 23 km range and r=0.95 for the CPT (16.7 km to 23 km) range. In this case, the water vapor enhancements at 17.2 km and 18 km, which are seen by Pico-SDLA but not by FLASH as well as the humidity variability in the lower TTL region, have a significant impact on the correlation.

In the case of the March 13, 2012 flights, the correlation coefficient is mainly affected by the two large water vapor enhancements observed at 18.1 km and 18.7 km and which do not have exactly the same thickness and amplitude. Within 15 km to 21.2 km, the *r* is equal to 0.98. Surprisingly, *r* decreases to 0.89 between the CPT (17.7 km) and 21.2 km. The statistical weight of the two structures at 18.1 km and 18.7 km is larger in the calculation when only altitudes above CPT are considered.

Because the two sensors did not fly at the same time, the correlation is strongly 496 affected by the variability in the water vapor enhancement structures shown by the two 497 different hygrometers. This effect is clearly visible through the changes in r between the two 498 altitude ranges. In the case of March 13, 2012 r is strongly affected by the two enhancement 499 structures (one is even present above the TTL upper limit). Despite the evident impact of the 500 vertical structures on the results, the present comparison exhibits some of the best agreement 501 502 found in the literature for studies realized from coincident flights (Weinstock et al, 2009; Khaykin et al, 2013). In each case, the water vapor enhancements are of much larger 503 amplitude than the difference between the two instruments. FLASH and Pico-SDLA are 504 therefore able to see, with good accuracy, the impact of dynamical process on water vapor 505 506 concentrations.

507 6. Summary and conclusions

508 This work compares *in situ* water vapor measurements from two hygrometers: Pico-509 SDLA H₂O and FLASH-B, obtained during the TRO-Pico balloon campaign held in Brazil 510 between 2012 and 2013. It serves as the basis for a future paper (S. M. Khaykin, personal 511 communication, 2015), centered on the meteorological analysis of the measurements.

The hygrometers were deployed on March 13, 2012 and February 10, 2013 when an overshooting convection event was observed in the vicinity of the flight paths. The impact of overshoots on water vapor mixing ratios is visible on March 13, 2012, by the presence of
three vertical structures at 17.4 km, 18.1 km and 18.7 km. A detailed analysis of this profile
will be given in the forthcoming paper (S. M. Khaykin, personal communication, 2015).

The water vapor profiles were compared within two altitude ranges : above 15 km and 517 above the CPT. The comparison above 15 km shows larger deviations (up to 9.5 %) than 518 those above the CPT (around 4%) because of humidity variability in the uppermost 519 troposphere. On March 13, 2012 and February 10, 2013, the mean difference of mixing ratios 520 is 0.5 % and 1.9 %, respectively, above the CPT altitude; differences which are well below 521 both instrument uncertainties. The differences are then much lower than the amplitude of the 522 water vapor enhancements (between 0.5 ppmv and 0.8 ppmv) permitting us to reliably detect 523 524 these overshoot signatures. Because the hygrometers were not flown at the same time, the humidity variability through the TTL had an important impact on the correlation coefficient 525 and on the mixing ratio differences between the two instruments. Nevertheless, the 526 differences observed in this study are well below the majority of in situ comparisons in the 527 528 TTL and constitute one of the best intercomparison results by comparison to the work of (Weinstock et al, 2009; Khaykin et al, 2013).In these previous studies, the mixing ratio 529 differences for in situ measurements ranged between 0.8 % and 5% and were obtained for 530 coincident flights. In the context where persistently large disagreements exist between in situ 531 532 measurements, the present work shows that Pico-SDLA H₂O and FLASH-B are suitable for accurate in situ water vapor measurements over a variety of conditions, such as those 533 including strong convection and high vertical speed. Furthermore, given the small differences 534 observed among the profiles of each instrument, it can be concluded that the H₂O data 535 536 provided by the TRO-Pico campaign made of Pico-SDLA and FLASH-B measurements are 537 mutually consistent. The compactness of these instruments permits their deployment under small weather balloons and therefore allows frequent soundings of the upper troposphere and 538 539 lower stratosphere to be performed.

540 Acknowledgements

This work and the TRO-Pico project were supported by Agence Nationale de la Recherche (ANR) under Contract ANR-2010-BLAN-609-01 and by the region Champagne-Ardenne in France. The authors are grateful for the logistical and infrastructural support provided by IPMet/UNESP under a collaborative agreement with the French CNRS. We also thank IPMet's technicians, Hermes França, Bruno Biazon and Demilson Quintão for their assistance during the TRO-Pico campaigns and the Dr. Joseph T. Hodges from NIST for his greatly appreciated help reviewing this paper.

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550

Figure captions :

Fig. 1: Description of the Pico-SDLA H_2O hygrometer, improved for the TRO-Pico campaign (2012-2013).

Fig. 2: Atmospheric spectra of the $2_{02} \leftarrow 1_{01}$ line of $H_2^{16}O$ from Pico-SDLA H_2O measurements on February, 10, 2013 during the descent of the flight. The top panel shows three experimental spectra(black line) and the results from fitting procedure(red line). These spectra were recorded at 25.2 km (24.24 mbar), 18.4 km (73.6 mbar) and 16.5 km (101.05 mbar) of altitude. The bottom panel shows the fit residuals for each spectrum.

Fig. 3: Balloon trajectories of Pico-SDLA and FLASH flights on February 10and February 11, 2013. The trajectories are color coded with altitude. The time is given in UTC. The ascent and descent time stamps correspond to time when balloon was passing an altitude of 14 km.

Fig. 4: Comparison of water vapor in situ measurements from Pico-SDLA H_2O and FLASH-B hygrometers in the TTL and lower stratosphere for the flight of February 10, 2013. The descent water vapor vertical profile of Pico-SDLA is represented by the solid black line. The ascent and descent water vapor profiles from FLASH-B are shown as solid blue and red lines respectively. The temperature profiles from Pico-SDLA and FLASH are shown in orange and brown lines. The CPT altitude is given by the orange and brown dashed lines for Pico-SDLA and FLASH respectively. The upper boundary of the TTL is shown is given by the green dotted line.

Fig. 5: Balloon trajectories of Pico-SDLA and FLASH flights on March 13, 2012. The trajectories are color coded with altitude. The time is given in UTC. The ascent and descent time stamps correspond to the time when the balloon passed an altitude of 14 km.

Fig. 6: Comparison of water vapor *in situ* measurements from Pico-SDLA H₂O and FLASH-B hygrometers in the TTL and lower stratosphere for the flight of March 13, 2012. The descent water vapor vertical profile of Pico-SDLA is represented by a solid black line. The ascent and descent water vapor profiles from FLASH-B are shown in solid blue and red lines respectively. The temperature profiles from Pico-SDLA and FLASH are shown in solid orange and brown lines. The CPT altitude is given by the orange and brown dashed lines for Pico-SDLA and FLASH respectively. The upper boundary of the TTL is shown by the green dotted line.

Fig. 7 : Scatter plot comparison of Pico-SDLA versus FLASH water vapor measurements between the CPT and the free-of-outgassing altitude (21.3 km on March 13, 2012 and 23 km on February 10, 2013). The linear fit of the data is represented with solid blue and black lines for the March 13, 2012 and February 10, 2013 flights respectively. The data are color mapped by the pressure.

Figure 1











Figure 4







Figure 6





Figure 7

