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2 SPICAM on Mars Express

Multi-annual monitoring of the water vapor vertical distribution on Mars by

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9 Abstract

10 The distribution of water vapor with altitude has long remained a missing piece of the 11 observational dataset of water vapor on Mars. In this work, we present the first multi-annual survey of water vapor profile covering the altitude range from 0 to 100 km based on the SPICAM/Mars 12 Express occultation measurements. During the aphelion season, water remains confined below 40-13 60 km for all Martian years observed. The highest altitude where water vapor can be spotted is 14 between 70 and 90 km during the southern summer (Ls=240-300°; perihelion season), approaching 15 16 the transition between the middle and upper atmosphere. In this season, years without a global dust storm (GDS) show a significant moistening of the upper atmosphere (~100 ppmv) in the southern 17 hemisphere, confirming a seasonal impact on the hydrogen escape rate. The two observed GDS, 18 19 in MY28 and MY34, show a substantial disparity in water vapor response. The storm in MY28, which coincides with the southern summer solstice, creates the largest excess of water in both 20 hemispheres at >80 km. This climatology of water vapor will supply a robust statistical basis to 21 address the long-term escape processes of water from Mars. 22

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24 Plain Language Summary

The vertical distribution of water vapor in the Martian atmosphere is key to understanding water transport and its escape from the planet, which in turn helps to explain the fate of water through Mars' history. Recent studies suggest that the transport of water to 80 km can increase the hydrogen escape rate by an order of magnitude and provide evidence for the role of global dust storms (GDS) in the regulation of this process. We monitored the vertical water distribution during multiple Martian years for the first time, including two years with global dust storms. We confirm the efficiency of GDS in delivering water to 80 km and emphasize the role of a regular perihelion season, which raises the water in the southern hemisphere every year. The MY28 GDS, coincident with the southern summer solstice, demonstrated the most massive increase of water abundances at high altitudes through the observed years.

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36 Key points:

1) Through eight Martian years observed by SPICAM H₂O regularly reaches 40-60 km in the

aphelion season and 70-90 km in the perihelion season.

2) In southern summer, large H_2O vmrs (~100 ppm) are repeatedly observed up to 80 km,

40 confirming a seasonal impact on the hydrogen escape rate.

3) Out of the two observed GDS, in MY28 and MY34, the GDS of MY28, coincident with the
southern summer solstice, shows a larger water increase.

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1. Introduction

Despite the scarcity of water vapor in the Martian atmosphere it nonetheless plays a pivotal 45 role in the planet's climate. Major efforts have been made to understand how water vapor is 46 47 spatially and temporally distributed over the last few decades (Montmessin et al., 2017a). Its global seasonal cycle has been studied continuously for 20 years through nadir measurements of column 48 abundance by TES/Mars Global Surveyor (Smith 2004), three experiments onboard Mars-Express 49 (MEX), SPICAM (Trokhimovskiy et al., 2015), OMEGA (Maltagliati et al., 2011a) and PFS 50 (Fouchet et al., 2007; Wolkenberg et al., 2011), and by the CRISM instrument on the Mars 51 Reconnaissance Orbiter (MRO) (Smith 2009). The global annual average abundance of water 52 fluctuates around 10 pr. µm (~200 ppm, assuming mixed water), increasing at high latitudes when 53

the polar caps sublimate in spring and summer of either hemisphere. The northern hemisphere reaches its maximum in summer at 75°N, exhibiting a column abundance of roughly 50 pr. μ m recurrent from year to year, while the corresponding southern summer maximum is half as strong shows some interannual variability around 25 pr. μ m.

In contrast to column abundance, the climatology of water vapor vertical distribution has 58 only been scarcely documented to date. The vertical distribution gauges multiple processes 59 controlling the Martian water cycle, including the condensation/sublimation leading to cloud 60 formation/collapse, the associated sedimentation and scavenging of dust particles upon which ice 61 62 particles form, but also the sublimation and condensation from the polar caps, the photodissociation and escape processes, and surface-atmosphere exchange (Montmessin et al., 63 64 2017a). The vertical distribution also appears to be a insightful tool for revealing the details of the Martian circulation, in particular, the massive upwards motions that permit water to access 65 altitudes higher than 100 km in some extreme cases (Shaposhnikov et al., 2019; Neary et al.; 2020). 66

By analogy with the Earth's troposphere, the vertical distribution of water on Mars is 67 assumed to be controlled by vapor pressure, which is solely determined by temperature. The 68 altitude level at which water vapor should condense in theory, the hygropause, rises from 10–20 69 km in the [40°S,80°N] latitude range during the colder aphelion period to 30-60 km in the 70 [80°S,40°N] latitude range during the warmer perihelion period (Richardson and Wilson, 2002a; 71 Montmessin et al., 2004). . Saturation and further condensation of water vapor at 10 km in northern 72 73 summer is responsible for the formation of the Aphelion cloud belt (ACB), which regulates the transfer of water between hemispheres. Condensation blocks water in the northern tropics where 74 the seasonal low-level air mass convergence turns into the rising branch of the Hadley cell (Clancy 75 76 et al., 1996; Richardson and Wilson, 2002a; Montmessin et al., 2004). The seasonal trend of saturation altitude can be inferred from column-integrated H₂O measurements assuming a known 77 temperature distribution, see, e.g., Trokhimovskiy et al. (2015). 78

However, the concept of saturation-controlled water holding capacity is presently undermined 79 by several lines of evidence. First, SPICAM solar occultation profiles measured in Martian Year 80 (MY) 29 at Ls=60–110° show considerable amounts of H₂O above the hygropause, at 30-40 km 81 82 (Maltagliati et al., 2011b). This water vapor, therefore, reached a supersaturated state with a pressure several times (up to 10) higher than the saturation pressure. Clancy et al. (2017) have also 83 found strong, though indirect, hints of the persistence of supersaturation in water at Ls=60-140° 84 using as a proxy the oxygen singlet emission measured by CRISM. The full second half of MY34 85 (Ls=165-360°) observed with ACS (Atmospheric Chemical Suite) onboard the Trace Gas Orbiter 86 87 (TGO) shows that water vapor supersaturation is deep and ubiquitous (Fedorova et al., 2020a), implying that water can be transported to the upper atmosphere much more easily than previously 88 89 assumed. High degrees of supersaturation were also predicted in simulations of water ice cloud 90 microphysics inside a Global Climate Model (Navarro et al., 2014).

Water vapor was observed at altitudes of up to 80-100 km during the Global Dust Storms (GDS) of 2007 (MY 28) and 2018 (MY34) with a volume mixing ratio in the 50-100 ppmv range (Fedorova et al., 2018, 2020a; Heavens et al., 2018; Vandaele et al., 2019; Aoki et al., 2019). The region above 60–80 km is essential in the fate of water on Mars, as the thinner CO₂ atmosphere no longer screens the solar UV light. Water photolysis here thus provides the main source of hydrogen atoms in the upper atmosphere, enabling their subsequent escape to space (Chaffin et al., 2017).

Variations in the Martian hydrogen escape rate by an order-of-magnitude were inferred from 97 98 Lyman-a airglow observations during the MY28 GDS by Mars Express and the Hubble Space Telescope (Chaffin et al., 2014; Clarke et al., 2014). Photochemical modeling tied the enhanced 99 escape rates with the presence of water molecules at 60-80 km during the GDS (Chaffin et al., 100 101 2017; Krasnopolsky et al., 2019). More hydrogen corona observations suggested that not only dust 102 storms but the perihelion season as a whole, even without a major dust event, might cause the 103 escape rate variation (Bhattacharya et al., 2015). Instead, during perihelion and outside a GDS, significant amounts of water vapor have been observed in the southern hemisphere during MY29 104

105 (Ls=240-260°) above 60 km (Maltagliati et al., 2013), and by ACS in MY34 (Ls=270-290°, in between the dust events) up to 100 km (Fedorova et al., 2020a). GCM simulations explain the 106 seasonal upward transport of water vapor by atmospheric dynamics lifting water in the upward 107 108 branch of the pole-to-pole meridional circulation cell. Shaposhnikov et al. 2019 demonstrated the upward water flux maximizes around perihelion between Ls= 220° and 300°, in a symmetric 109 fashion about Ls=260° when the global mean temperature reaches annual maximum and the 110 111 circulation is the most intense because of hemispheric dichotomy (Richardson and Wilson, 2002b). Even at this moment, the upward water flux is minimal at ~ 60 km, where water can penetrate into 112 113 the upper atmosphere only in the region between 20°S and 70°S and transported further upward and across latitudes northward. At other seasons this flux is negligible. 114

Better seasonal coverage of the vertical water distribution in the lower atmosphere (<100 km) of Mars and of the Lyman- α airglow in the upper atmosphere (>200 km) should help disentangle the respective roles between the intense but rare GDS and the weaker but recurrent seasonal increase of dust in modulating the escape of hydrogen from Mars. The monitoring of water vapor profiles is also necessary to understand whether the high-altitude water, which constitutes a small fraction of the whole atmospheric water, is an essential component in the transport of water across the globe.

122 Here, for the first time, we present observations of H₂O vertical distribution in the Martian atmosphere obtained by SPICAM infrared spectrometer onboard Mars Express during eight Mars 123 124 years, from MY27 to MY34, including two GDS. The instrument measures the water density and mixing ratio at the Mars limb in solar occultations (Korablev et al., 2006; Fedorova et al., 2009, 125 2018; Montmessin et al., 2017b; Maltagliati et al., 2011b, 2013). The details of the SPICAM 126 127 observations and the data analysis are described in sections 2 and 3, respectively. The retrieved climatology is presented in section 4, where we discuss the average dataset with a focus on the 128 second, dusty half of the Martian year, from Ls=180° to 360°, and the two GDS. 129

131 2. SPICAM observations.

132 SPICAM IR is an AOTF (Acousto-Optic Tunable Filter) infrared spectrometer. It works in the spectral range of 1–1.7 μ m with a spectral resolution of 3.5–4 cm⁻¹. Near the H₂O 1.37- μ m 133 134 band the resolving power corresponds to ~2000. The spectrometer operates both in nadir and solar occultation modes; the present work is based on occultation data. Two detectors of the 135 spectrometer working in orthogonal polarizations have different signal-to-noise ratios (SNR). This 136 corresponds to ~120 in channel 0 and ~220 in channel 1 for a pure solar spectrum outside the 137 138 atmosphere in the range of 1.2–1.55 µm. Thus we use the data of channel 1 for the analysis. With the FOV of 4.2 arcmins (0.07°) the vertical resolution in occultations varies from 1 to 12 km (4–5 139 140 km on average) due to the elliptical orbit of the Mars-Express spacecraft. More details of the instrument and its calibrations related to occultations can be found in papers by Korablev et al. 141 (2006a, b) and Fedorova et al. (2009). 142

SPICAM can measure the H₂O and CO₂ density and the aerosol density and properties. 143 144 The AOTF implies a sequential acquisition of spectra, but the measured spectral range can be arbitrarily chosen. In solar occultation we use 609 spectral points in the range of 1.34–1.47 µm to 145 record the strongest CO₂ 1.43-µm band, the H₂O 1.37-µm band and 55 points distributed between 146 1 and 1.7 µm to measure transmission outside gaseous absorption bands ("reference wavelengths") 147 and characterize the optical properties of aerosols (see also Fedorova et al., 2009, 2018). One 148 149 spectrum is recorded in 4 s. Taking into account that the vertical projection of the spacecraft speed on the limb varies from 0.5 to 3 km/s, the change in altitude over one spectrum varies from 2 to 150 151 10 km depending on occultation.

From 2005 to March of 2019 Mars Express has completed 18 occultation campaigns. Figure 1 shows the coverage of solar occultations for eight Martian years from MY27 to MY35. This dataset includes ~1500 occultations. During the two first occultation campaigns in MY27 the spectral coverage was not optimized, which meant that only the H₂O density could be retrieved, but not the CO₂ density. Through the whole dataset, ~15% of data are not suitable for the retrieval 157 of gaseous density profiles due to pointing instabilities, problems with the reference solar 158 spectrum, etc.



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Figure 1. Seasonal-latitudinal coverage of SPICAM observations in solar occultations for eight
 Martian years. The open circles mark faulty observations inapplicable for retrieval. Morning
 observations are marked by triangles and evening observations by stars.

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164 **3. Data processing**

The details of the SPICAM IR data processing in occultations is described in Fedorova et al. (2009, 2018) and Maltagliati et al. (2011, 2013). A summary of the retrieval process and a few recent amendments to it are presented below.

Occultation observations are self-calibrated, yielding transmittance of the atmosphere from the ratio of a spectrum through the atmosphere to the solar reference collected high enough where the atmosphere negligibly absorbs. Above 120 km for any season and location the CO_2 1.43 μ m absorption band is too weak to be detected by SPICAM; we therefore averaged the reference spectrum between 120 and 170 km. Fig. 2 shows an example of transmittances obtained in orbit 18560 of MY34.





Figure 2. SPICAM transmittance spectra measured between 0 and 120 km for orbit 18560A1 (Ls=242°, 59.7°N, 301.5°E) in the spectral range of 1346–1462 nm. The color indicates the average altitude of a spectrum. Data of detector 1 are shown. The black curve is the model spectrum with the CO₂ and H₂O absorption bands at the target altitude of 20 km. Solid lines bound the spectral ranges used for the retrievals. A broad and shallow absorption appearing at 1350– 1390 nm (~0.6 transmission level) is likely a signature of a thin detached aerosol layer, manifested as a change of the slant optical depth during the spectrum record.

Because each spectral point in an occultation spectrum is measured sequentially, each point corresponds to a different altitude. The aerosol opacity, which changes with altitude, results in an artificial spectral slope (see Fig. 2). To correct it, we normalized the spectra around the studied absorption bands to the continuum outside the bands. For CO_2 , the continuum is computed with a linear interpolation between averages of two intervals on both sides of the band, 1423–1426 nm and 1456–1459 nm. The H₂O band at the short-wavelength boundary extends beyond the acquired spectral window, and the continuum is estimated in between the absorption lines within the bandas described in Maltagliati et al. (2013).

To reduce the noise in measured spectra, we applied the Savitzky-Golay smoothing filter (Savitsky and Golay, 1964) to ensure a strong noise reduction while preserving the spectral resolution. No averaging of spectra along the occultation sequence is performed, because of the too coarse vertical sampling, with typically 5–25 useful spectra during an occultation between 0 and 100 km. We also limit the H₂O fit to the strongest part of the band, between 1360 and 1409 nm.

198 The whole profile of the local H₂O or CO₂ density is retrieved using the Levenberg-Marquardt 199 iterative algorithm (Levenberg, 1944; Marquardt, 1963). The Tikhonov regularization is applied 200 after the fit, in order to smooth the profile and minimize the errors (Ceccherini, 2005; Ceccherini 201 et al., 2007). The uncertainty in the densities is given by the covariance matrix of the solution errors. The H₂O detection limit is estimated as $7-9\times10^9$ molecules/cm³. This detection limit 202 203 corresponds to the volume mixing ratio (vmr) accuracy better than 1 ppm below 35 km, better than 204 10 ppm at 50-55 km and better than 70 ppm at 80 km. The measurement accuracy varies with 205 season and location due to variations of atmospheric density. The CO₂ detection limit is below 10^{12} molecules/cm³ which corresponds to the pressure level of 2×10^{-6} - 6×10^{-5} mbar. The 1.43 µm 206 absorption band is visible up to 110-115 km depending on location and season. 207

208 A line-by-line synthetic spectrum of gaseous absorption was calculated using the HITRAN 209 2012 spectroscopic database (Rothman et al., 2013). There are only a few laboratory measurements 210 of the CO₂-broadened H₂O half-widths in this band for some lines (Langlois et al., 1994). Based on theoretical calculations by Gamache et al., (1995) and measurements in thermal IR by Brown 211 212 et al., (2007), we correct the air-broadening half-widths for the CO₂-broadening in the Martian atmosphere by multiplying by a factor of 1.7 (see also discussion in Fedorova et al., 2010). In case 213 214 of occultation measurements, the Doppler broadening begins to dominate above 30-40 km and the sensitivity to this coefficient is minimal. 215

216 Information about the pressure and temperature profile in the Martian atmosphere is a critical aspect of SPICAM's water retrieval, to simulate spectra for the forward model, as well as to 217 compute the mixing ratio. As described in Fedorova et al. (2018), while the retrieved H₂O density 218 219 is not very sensitive to the temperature profile, the CO_2 density could be mistaken by a factor of 2 220 when the temperature deviates by as much as 50 K. This translates into a 50% error in the water mixing ratio. For the forward model we used the temperature and pressure profiles from the recent 221 222 European Martian Climate Database (EMCD) [http://www-mars.lmd.jussieu.fr/, version MCD V5.3; Forget et al. 1999; Millour et al. 2019]. To mitigate the differences between the model and 223 224 the observations we choose the individual Mars years scenarios available in the MCD for MY 24 225 through 34, which use the observed daily dust column maps for each year to constrain the dust 226 distributions in the model and hence reproduce observations to the greatest extent possible 227 (Montabone et al., 2015, 2020). These scenarios reproduce the temperature profiles by year reasonably well, with inaccuracy being far below 50K outside of a GDS. In the case of CO₂, the 228 density profile predicted by MCD was used as an *a priori* vector in the minimization procedure. 229 230 For H₂O, we used a constant volume mixing ratio of 1 ppmv at all altitudes as an initial assumption. Examples of H₂O fits can be found in Fedorova et al. (2009; 2018). 231

Even if individual dust scenarios of MY25, 28, and 34 in the MCD include the extreme 232 global dust events, modeling of a GDS remains challenging. As shown in Fedorova et al. (2018), 233 during the MY28 GDS a difference between MCD temperature profiles and measured MCS/MRO 234 235 profiles sometimes reached 40 K. Such a bias can be critical for our retrievals. We resolved to use MCS temperature profiles (Kleinböhl et al., 2009) close in time/space for these special cases when 236 available. MCS measures at 03:00 and 15:00 Martian local time, out of phase with the Mars-237 Express occultations, but many observations within 1° of Ls and 2° of latitude and 5° of longitude 238 could be found. The selection of the MCS data was done at local times closest to the SPICAM 239 240 observations and performed additional checking using the daily temperature cycle of the MCD to find the difference between morning-evening SPICAM observations and day-night MCS profiles, 241

as detailed in Fedorova et al., 2018. The share of available collocated MCS temperature profiles
during two GDS (in MYs 28 and 34) used to retrieve the H₂O densities is high, reaching 90% of
all observations.

The vertical profile of the H₂O volume mixing ratio (f_{H2O}) is the ratio between the water 245 vapor number density and the atmospheric number density. To find the atmospheric density profile 246 we divide the CO_2 profile by the CO_2 mixing ratio, assumed to be 0.9543. As the CO_2 and the H_2O 247 248 bands were not acquired at the same altitude (as described above) we find the atmospheric density at the H₂O altitude with a log-linear interpolation. The error in the H₂O mixing ratio is given by 249 250 the quadratic sum of the uncertainties of the two density profiles. The main systematic error on 251 H₂O vmr results from the temperature profile uncertainty, in particular during the dusty part of the 252 Martian year, due to a strong sensitivity of the CO₂ band, as described above. The efforts taken to 253 minimize this uncertainty reduce the error in the H_2O vmr to <20-30%. All the details of the 254 retrieval and the associated sensitivity analysis are presented in previous works (Maltagliati et al., 2011b, 2013, Fedorova et al., 2009, 2018). 255

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4. Results

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H₂O density distribution

The SPICAM water density profiles have been obtained for eight Martian years from MY 27 to 34 with two occultation campaigns per year. The vertical distribution of water vapor is highly variable with season and altitude. To obtain the latitude vs. altitude distribution seasonal distribution through regular years without dust storms, we averaged all observations into $12 \times 30^{\circ}$ Ls bins (Figure 3) and then averaged each hemisphere separately to compare their respective Ls vs. altitude distributions (Figure 4). The two GDS of MY28 (Ls=267–302°) and MY34 (Ls=190– 230°) characterized by a high elevation of water vapor are excluded from the averaged maps.

The detected density varies from 5×10^9 cm⁻³ to 10^{13} cm⁻³. Water vapor is less abundant and more confined into the lower atmosphere during northern spring/summer, as expected, due to the colder aphelion climate and correspondingly the lower level of the hygropause. Noticeable amounts of water are observed at 30-40 km, while profiles can be traced as high as 40–60 km in the aphelion season at Ls=30–120°. Around perihelion, at Ls=240–270° the highest water is observed at 70–90 km. To first order then, water transport to higher altitude is dictated by atmospheric temperature as the Aphelion-to-Perihelion transition corresponds to >20K temperature change (Smith et al., 2017).

At Ls=30–90° during the northern spring-summer, the maximal water density was observed in the middle and high northern latitudes corresponding to the sublimation at the northern polar cap and subsequent water release. In the southern equinox (Ls=150–210°) timeframe, the water distribution is very symmetric between the two hemispheres with low values at high latitudes. Sparse detections at Ls=0–150° are explained by the confinement of the water vapor near the surface, associated with a less confined aerosol layer and Aphelion intertropical clouds that both impair detection.

Figure 5 shows the maximal altitude where the H_2O density reached 10^{10} (close to SPICAM's 281 detection limit, but still dependably measured). The black open circles mark orbits where no water 282 was detected at all, meaning the density at all altitudes was below $5-7 \times 10^9$ cm⁻³. This happened 283 for some of the low-latitude orbits associated with the aphelion clouds at $Ls=0-150^{\circ}$. Detections 284 are also absent at polar latitudes, poleward of 60°S in southern winter (Ls=20–180°), and poleward 285 of 60°N in northern spring (Ls=330-30°) and autumn (140-200°). The 10¹⁰ cm⁻³ detection 286 threshold altitude varies from 15 to 40 km (30 km on average) in the aphelion season, rising to 60-287 90 km in the perihelion season. 288



Figure 3. The H₂O density distribution (in cm^{-3}) with altitude and latitude for 12 Ls bins

averaged over the MY27-34 timeframe. The altitudes and latitudes have been arranged into 5km

292 \times 5° bins. The dataset for the GDS of MY28 and 34 were excluded. The y-axis shows the altitude

above the areoid. Black curves mark averaged MOLA altimetry for the bins.

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Figure 4. The H₂O density distribution (in cm⁻³) with altitude as a function of Ls for the northern and southern hemispheres. The Ls scales are shifted to align summer and winter seasons in both hemispheres Altitudes and Ls are arranged into 5 km \times 5° bins. Two GDS of MY28 and 34 are excluded from the binning.

300 In contrast, the warmer southern spring appears associated with higher vertical mobility of water vapor. From Ls=210° the maximum of water density moves to middle-to-high southern 301 302 latitudes, marking the dissipation of the polar cap. The water amount increases after $Ls = 240^{\circ}$ to solstice, when the density reaches 10^{12} cm⁻³ above 40 km and water extends to 90 km (Fig. 3 and 303 304 4). Several periods when the high-altitude water is observed in the dusty season can be identified in Figure 5. Two periods, marked by stars, are related to the GDS of MY28 and 34 at Ls=190-240° 305 and 270-300°, respectively. During these times, water attains altitudes up to 90 km in both 306 hemispheres. A pronounced solsticial maximum at Ls=240-300° corresponds to water lofted up 307 308 to 70-90 km in the southern hemisphere every year. In the northern hemisphere, water reaches only

60-80 km and thus the picture looks asymmetric around the equator in the corresponding panel ofFig. 3.

We also note a moderate, compared to perihelion and GDS, increase of water up to 60-80 km 311 at $Ls = 315 - 330^{\circ}$ in both hemispheres (Fig. 4), at least for three occultation campaigns MY27, 31 312 and 32 (Fig. 5). This increase is correlated with regional dust activity, which occurs at this Ls 313 almost every year (Kass et al., 2016). This regional dust activity started between Ls=305° and 320° 314 and lasted 3–15° of Ls. Interestingly, water increased in both hemispheres in middle to high 315 latitudes from 40° to 70° in accord with the recurrent northern temperature response (Kass et al., 316 2016). A similar increase of water was observed during the MY34 GDS by TGO (Aoki et al., 317 318 2019; Fedorova et al., 2020a).

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Figure 5. The altitude of water density of 10¹⁰ cm⁻³ (close to SPICAM's detection limit). Black open circles mark orbits where water is below the detection limit everywhere in the profile. Stars correspond to the GDS of MY28 and 34.

Figures 6 and 7 show the H₂O vmr latitude-altitude cross-sections for 12 seasonal bins and for 326 the two hemispheres, respectively, similar to Figures 3 and 4. In the aphelion season at Ls=30-327 120°, the H₂O vmr does not exceed 40 ppmv. A minimum of water is found in middle-to-high 328 southern latitudes during southern winter (Ls= $60-90^\circ$), where the vmr was below 1-2 ppmv. The 329 confinement of water vapor towards the surface during the aphelion season suggested by the 330 present observations (as the lowest part of the profile is inaccessible to occultation) appears in 331 agreement with the conclusions drawn from nadir observations (Smith, 2004; Trokhimovsky et 332 al., 2015). Nonetheless, a sharp decrease of the mixing ratio from 50 to 5 ppmv occurs in the 333 334 middle northern latitudes at Ls=30-120° (the most prominent at Ls=90-120°), indicating a rise of the hygropause from 15 km at 60° N to 25–30 km at 40° N. 335

The water mixing ratio in low-to-middle latitudes starts to grow in the middle atmosphere of 336 both hemispheres after Ls=180°, reaching 50 ppmv between 20 and 60 km. Higher northern and 337 southern latitudes are exempt from this moistening and remain dry with vmr $\leq 5-10$ ppmv. In the 338 Ls=210-240° period, humid air progressively fills all latitudes, and reaches altitudes up to 70-80 339 km. The most abundant water (>100 ppm) can be observed at Ls=240-270° in both hemispheres, 340 spanning the whole middle atmosphere (from 30 to 70-80 km) exhibiting an uniform distribution. 341 At high southern latitudes the water layer expands vertically higher than 90 km. A similar pattern 342 is observed at Ls=270–300°, even if our occultation dataset does not cover this season well enough. 343 The mixing ratio decreases only after Ls \sim 300° in the north, as tracked by the altitude of the 10¹⁰ 344 density level in Figure 5. However, the mixing ratio in the low-to-mid southern latitudes still 345 346 reaches 50-100 ppmv at 70 km. Remains of the southern summer water maximum can still be 347 spotted at low latitudes throughout the Ls=330-360° period.



Figure 6. Same as Figure 3 except the H₂O mixing ratios are displayed instead of the number

351 density.



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Figure 7. Same as Figure 4 except the H₂O mixing ratios are displayed instead of the number density.

4.3 Comparing years with and without GDS during the dusty season

The SPICAM survey period includes the two GDS that happened in 2007 (MY28) and 357 2018 (MY34). Wang and Richardson (2015) described the development of the MY 28 GDS as 358 observed by MARCI (MARs Color Imager) on MRO. The storm reached its global phase on 359 Ls=269.3° (13 sols after onset) at southern middle-to-high latitudes between 35–80°S (Cantor et 360 al., 2008). By the end of the expansion phase (Ls~275.2°), the dust encircled the planet from 361 approximately 90°S to 58°N, reaching an optical depth of 4.4-4.6 (Smith, 2009b; Lemmon et al., 362 2015). The decay phase started at Ls \sim 310° and ended at Ls \sim 310-320°. 363 The 2018 GDS has been reviewed by Guzewich et al. (2019), Montabone et al. (2020), and 364 references therein. The storm started regionally in the northern hemisphere in the second half of 365

366 May 2018 (*Ls* ~186°) and then merged by the beginning of June (*Ls* ~190°) with another regional

dust lifting occurring independently in the southern hemisphere. It became global by the middle of June (*Ls* ~197°). This most active period of the GDS lasted until the beginning of July (*Ls* ~205°) followed by a long decay, which ended in the middle of September (*Ls* ~240–250°).

370 The process of dust lifting was uneven across the globe for both GDSs, and even at the peak of the storm northern latitudes (>50°N) and southern polar areas were mostly clear of dust and 371 suitable for solar occultations (Montabone et al., 2015; 2020). The intercomparison of the two 372 373 GDS (Smith, 2019; Wolkenberg et al., 2020) showed that even though they happened in different seasons, their globally-averaged peak optical depths were similar. The MY 28 GDS reached dust 374 375 optical depth greater than 1.5 (at 9 μ m) rapidly, within 10°–15° of Ls, and then maintained this dust loading for an extended period, before decay. According to THEMIS observations the MY 376 377 34 GDS was characterized by a more gradual build-up with a two-stage increase in dust optical 378 depth. Dust optical depth increased rapidly at the start of the storm at the same rate as during MY 379 28 but then remained nearly constant for several degrees of Ls before increasing again to its maximum value (Smith, 2019). 380

The water vapor distribution during MY28 was described in detail by Fedorova et al. 381 (2018). Here we compare the behavior of water during the two GDS, highlighting the contrast with 382 regular perihelion seasons, unaffected by GDS. Figures 8 and 9 present the H₂O density and mixing 383 ratio distribution for altitudes of 60, 70 and 80 km and the lowest usable tangent height for 384 SPICAM occultations, limited by transmission levels below this point being <10 percent which 385 386 corresponds to aerosol optical depth of \sim 3 on the line of sight and indicating the aerosol loading. In the northern hemisphere from Ls=268° to Ls=285° the H₂O density increased by an order of 387 magnitude at 60-80 km (fig.8). During the dust storm, the profiles extended up to 80 km, with a 388 389 H₂O mixing ratio \geq 100 ppmv (fig.9). We note two maxima of the H₂O density. The first and largest maximum was observed above 60°N, from Ls=269 to 275° and does not directly correlate with 390 391 the aerosol loading. It likely relates to the downwelling branch of the meridional circulation, transporting water from the southern to the northern hemisphere, which intensified during the 392

- 396 During the MY34 GDS, SPICAM observations in the northern hemisphere began at very high
- latitudes, $>75^{\circ}$ N. Only from Ls=200° was a water density of $>10^{10}$ cm⁻³ detected with a mixing
- ratio up to 100 ppmv at altitudes of 60-70 km and well-correlated with increase of aerosol loading
- (Fig. 8). Compared to the MY28 GDS, water density at 60 to 80 km was one order of magnitude
- 400 lower. In the southern hemisphere, water density peaked at 10^{11} cm⁻³ at 70 km, and 5×10^{10} cm⁻³
- 401 at 60 km, with mixing ratios of 50-100 ppmv but still 2–3 times lower compared to MY28.
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- 403



405 Figure 8. Seasonal distribution of observations for two hemispheres, (A), the lowest altitude

attained in occultation (a measure of aerosol loading), (B), the H₂O density at 60, 70 and 80 km
(C, D, E, respectively) for MY27-MY34. The altitudes above the areoid are presented. The two
GDS are marked by the pink shaded Ls periods.

409

Compared to a "regular" year, H₂O at 60-70 km during MY28 is several times more abundant in the North. In the South, high H₂O density is also observed in MY29, 32, and 33, but still only half as large as in MY28. The density increase for the same Ls during MY34 in the southern hemisphere exceeds other years by 2-3 times, comparable with perihelion regular density, but lower than during the MY28 GDS. Despite the observational selection, we conclude that the MY28 GDS was exceptional in terms of delivering water to high altitudes (80 km), exceeding at least by 2 times the perihelion water maximum and the water supply during GDS of MY34.

417 As one of possible explanations is the existence of a seasonal water "pump" mechanism responsible for the upward transport of water vapor suggested by Shaposhnikov et al. (2019). A 418 combination of the mean vertical flux with variations induced by solar tides facilitates penetration 419 420 of water through the condensation at ~ 60 km. This mechanism becomes efficient at high southern latitudes (>60°S) at perihelion. That time the upward branch of the meridional circulation is 421 422 particularly strong. The recent supersaturation observation in this region (Fedorova et al., 2020a) even facilitates this process. Water is lifted up by the upward branch and the meridional circulation 423 424 then transports it to the northern hemisphere. The GDS MY28, overlapping the upward branch of 425 the meridional circulation that happened southern summer Ls=250-280° in perihelion, made the lifting of water to extremely high altitudes and transports to middle-to-high northern latitudes. 426



429 Figure 9. Same as in Figure 8 for the H₂O mixing ratio.

431 Since April 2018, two TGO infrared spectrometers (ACS and NOMAD) perform vertical profiling of water vapor. A nearly polar, not sun-synchronized circular 400-km orbit of TGO is 432 well adapted for solar occultations which began from Ls=163° of MY34. ACS and NOMAD 433 measure the vertical distribution of H₂O in several bands (Korablev et al., 2018; Vandaele et al., 434 2018). Aoki et al. (2019) reported the H_2O distribution during the GDS and the regional storm at 435 Ls=315–330° based on NOMAD SO observations in the 2.56-µm band. Fedorova et al. (2020a) 436 reported the water and temperature for the full duration of the dusty season from Ls=165° to 360° 437 with detection of supersaturation of water in this period based on ACS NIR observation in the 1.38 438 439 μm band.

ACS NIR is an AOTF echelle spectrometer working in the range of $0.76-1.7 \mu m$ with spectral resolving power of 25,000–27,000 (Korablev et al., 2018). In occultation, CO₂ density and temperature profiles are retrieved from 1.57- μm CO₂ band and H₂O density from 1.38- μm band (Fedorova et al., 2020a). Simultaneous measurement of temperature minimizes the uncertainties, discussed in Section 3 for the case of SPICAM retrievals. The vertical resolution of NIR is ~1 km, its sensitivity is better than 1 ppm between 10 and 75 km and ~20 ppm at 100 km, which surpasses the SPICAM characteristics.

447 In order to validate the multi-year SPICAM dataset, we compare the data of SPICAM IR and 448 ACS NIR during the MY34 GDS from Ls=160° to Ls=260° in Figure 10. The data were binned in 449 the same manner for both experiments into 5 km of altitude and 5° of Ls. We note a much larger 450 altitude range, in which the ACS profiles can be retrieved. Although the occultations as observed from the two spacecraft occur at different latitudes, the retrieved water trends are similar. Also, 451 452 the retrieved values are generally close, with the exception of cases with evident latitudinal mismatch. In the northern hemisphere the increase of water to >50 ppm as observed by SPICAM 453 began at Ls=195–200° whereas in NIR data the growth started one bin earlier (Ls=190–195°). This 454





Figure 10. The comparison of SPICAM and ACS NIR observations of the MY34 GDS. Seasonal
distribution with altitude is shown for two hemispheres. Altitudes and Ls are arranged into
5 km×5° bins. Top panels: Distribution of occultations for both instruments; Middle panels:
SPICAM observations; Bottom panels: ACS NIR observations.

462 **5.** Conclusions

Here we present the first multiyear survey of water vapor vertical distribution obtained from occultation measurements performed by the SPICAM spectrometer onboard the Mars Express spacecraft. The water vapor density profiles have been retrieved for 8 Martian Years MY27–MY34 and the water vapor mixing ratio for 7 Martian Years MY28–MY34, because the CO₂ density reference was not measured during the first year (MY27). Data reveal seasonal and spatial variations, and the impact of two global dust storms.

The detected water density varies from 5×10^9 cm⁻³ to 10^{13} cm⁻³. The upper level of detected 469 water density is located at 40–60 km in the aphelion season and 70–90 km in the perihelion season. 470 At Ls= $30-90^{\circ}$, during the northern summer, the maximal water density was observed in the 471 472 middle and high northern latitudes, corresponding to sublimation and subsequent release of vapor from the northern polar cap. Most of the aphelion detections (Ls=30–120°) are located above the 473 hygropause with H₂O mixing ratio <20 ppmv. Nevertheless, the hygropause was detected in the 474 middle northern latitudes at Ls=30–120°, manifested with a sharp decrease of the mixing ratio 475 from 50 to 5 ppmv, at the level of 15 km at 60°N, to 25–30 km at 40°N. Around the southern 476 equinox, in the Ls=150-210° timeframe, low water amounts are distributed symmetrically 477 between high latitudes of both hemispheres. 478

The perihelion season is characterized by an increase of the water amount after Ls=240° to 479 solstice, when the density reaches 10^{12} cm⁻³ and mixing ratio 100 ppm above 40 km. Water extends 480 481 to 90 km in the southern hemisphere confirming a seasonal impact on the hydrogen escape rate (Bhattacharya et al., 2015). In the northern hemisphere water reaches 60–80 km and the solstice 482 pattern looks asymmetric. The largest mixing ratio of >100 ppmv at 40–90 km was observed at 483 Ls=240-300° in high and middle southern latitudes. At the end of the dusty season, during the 484 485 regional dust activity at Ls=310-330°, which occurs at this Ls in almost every year (Kass et al., 486 2016), an abrupt increase of water was observed in both hemispheres in middle-to-high latitudes.

Intercomparison of water during the two global dust storms of MY28 and 34 shows that during 487 the MY28 GDS, coincident with summer solstice (Ls=268°-285°) SPICAM observed a two- to 488 three-fold increase of the H₂O density at 60-80 km, exceeding that in regular years and in MY34 489 490 (when this period followed a GDS), especially in the northern hemisphere. The water density 491 increase during the MY34 GDS is comparable with the regular solstice water in the southern hemisphere. In the northern hemisphere, even though our data for MY34 sample very high 492 493 latitudes only, the increase of water is prominent, marking an intensified circulation during global dust events. We conclude that the MY28 GDS was exceptional in terms of delivering water to high 494

altitudes (80 km), exceeding by at least a factor of two the perihelion water maximum and thewater supply during the MY34 GDS.

497 Comparison of SPICAM/MEX and ACS NIR/TGO observations during the MY34 GDS shows498 a good quantitative agreement and similar trends in both hemispheres.

The seasonal middle atmosphere water distribution presented here will help us to understand in detail the water transport between hemispheres, to study its interannual variations, and to separate the effect of seasonal water variation from the global and regional storm contribution, in order to quantify better the hydrogen escape from Mars.

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512 Competing interests

513 The authors declare no competing interests

514 **Data and materials availability**

515 SPICAM raw and calibrated data are available in the ESA Planetary Science Archives (PSA)

516 <u>https://archives.esac.esa.int/psa/#!Table%20View/SPICAM=instrument</u>. The H₂O density and

- 518 <u>http://dx.doi.org/10.17632/vx4gks6bx7.1</u> (Fedorova, A. 2020b). The full IR Solar Occultation
- 519 Level-2B data will also be publicly available on the ESA PSA.

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