Water vapor vertical profiles on Mars in dust storms observed by TGO/NOMAD

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- 20 Key Points:
- We present vertical profiles of water vapor in the Martian atmosphere during global and regional dust storms in 2018-2019.
- We show a rapid and significant increase of water vapor in the middle atmosphere (40-100 km) during both global and regional dust storms.
- Water vapor reaches very high altitudes, at least around 100 km, during the global dust storm.
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29 Abstract

It has been suggested that dust storms efficiently transport water vapor from the near-surface to 30 the middle atmosphere on Mars. Knowledge of the water vapor vertical profile during dust 31 storms is important to understand water escape. During Martian Year 34, two dust storms 32 occurred on Mars: a global dust storm (June to mid-September 2018) and a regional storm 33 34 (January 2019). Here we present water vapor vertical profiles in the periods of the two dust storms ($Ls=162-260^{\circ}$ and $Ls=298-345^{\circ}$) from the solar occultation measurements by Nadir and 35 Occultation for Mars Discovery (NOMAD) onboard ExoMars Trace Gas Orbiter (TGO). We 36 show a significant increase of water vapor abundance in the middle atmosphere (40–100 km) 37 during the global dust storm. The water enhancement rapidly occurs following the onset of the 38 storm (Ls~190°) and has a peak at the most active period (Ls~200°). Water vapor reaches very 39 high altitudes (up to 100 km) with a volume mixing ratio of ~50 ppm. The water vapor 40 abundance in the middle atmosphere shows high values consistently at 60°S-60°N at the growth 41 phase of the dust storm ($Ls=195-220^\circ$), and peaks at latitudes greater than 60°S at the decay 42 phase ($Ls=220-260^{\circ}$). This is explained by the seasonal change of meridional circulation: from 43 equinoctial Hadley circulation (two cells) to the solstitial one (a single pole-to-pole cell). We also 44 find a conspicuous increase of water vapor density in the middle atmosphere at the period of the 45 regional dust storm ($Ls=322-327^{\circ}$), in particular at latitudes greater than 60°S. 46

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48 Plain Language Summary

49 The most striking phenomenon on Mars is a planet-encircling storm, "global dust storm". Once it starts, the floating dust covers the whole atmosphere for more than several weeks. Recent studies 50 suggest that dust storms effectively transport water vapor from the near-surface to the middle 51 atmosphere. In June-September 2018 and January 2019, a strong global dust storm and a regional 52 storm occurred on Mars, respectively. This study investigates altitude profiles of water vapor in 53 the Mars atmosphere measured during the dust storms, by using brand-new measurements by 54 Nadir and Occultation for Mars Discovery (NOMAD) onboard the ExoMars Trace Gas Orbiter 55 (TGO). We confirm that the water vapor expanded into the middle atmosphere and we find that 56 57 the water vapor reached very high altitudes (up to 100 km) during the dust storms. The dust storms intensify the atmospheric dynamics and heat the atmosphere. As a result, water vapor is 58 lifted to higher altitudes and distributes along the meridional circulation. 59

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61 **1. Introduction**

Recent observations and studies have revised our understanding of water loss processes 62 on Mars. The variations of the escape rate are not dominated by the solar EUV radiation flux but 63 rather by the variable water vapor abundance in the middle atmosphere (Chaffin et al., 2014, 64 2017; Clarke et al., 2014; Fedorova et al., 2018; Heavens et al., 2018; Clarke 2018). Interestingly, 65 these recent measurements imply that global dust storms may effectively transport water vapor 66 from the near surface to the middle atmosphere and hence increase the escape rate with respect to 67 the atmospheric water loss under no-storm conditions. Heavens et al. (2018) and Fedorova et al. 68 (2018) showed the vertical profile of water vapor before/during/after the global dust storm in 69 2007. They found a significant increase of water vapor abundance in the middle atmosphere and 70 an increase in altitude of the hygropause (where the water content rapidly decreases following 71

saturation and ice cloud formation). Fedorova et al. (2018) found that the water vapor 72 enhancement is asymmetric between the northern and southern hemispheres – the increase of the 73 water vapor abundance due to the global dust storm is remarkable only in the northern 74 75 hemisphere. This suggests that meridional circulation of the atmosphere is intensified during the dust storms and transports water vapor more efficiently from the southern to the northern 76 hemisphere. Moreover, it is suggested that dust storm related increases of atmospheric 77 temperatures suppress the hygropause, hence reducing ice cloud formation and so allowing water 78 79 vapor to extend into the middle atmosphere (Heavens et al., 2018; Neary et al., 2019).

To date, vertical profiles of water vapor have been investigated by the solar occultation 80 measurements with Spectroscopy for Investigation of Characteristics of the Atmosphere of Mars 81 (SPICAM) onboard Mars Express (MEX) (Maltagliati et al., 2011; Maltagliati et al., 2013; 82 Fedorova et al., 2018) and by the limb measurements with Compact Reconnaissance Imaging 83 Spectral Mapper (CRISM) and Mars Climate Sounder (MCS) onboard the Mars Reconnaissance 84 Orbiter (MRO) onboard Mars Reconnaissance Orbiter (MRO) (Clancy et al., 2017; Heavens et 85 al., 2018). The measurements by SPICAM and MCS revealed unexpected high abundance of 86 water in the middle atmosphere that contributes the atmospheric escape of water. They also 87 found that the water vapor abundances in the middle atmosphere are further increased during the 88 global dust storm that occurred in 2007. However, even though it is proposed that an intensified 89 meridional circulation may transport water vapor efficiently, the complete picture of the water 90 91 vapor distribution during global dust storms is not yet confirmed. This is because (1) the MEX/SPICAM measurements can be performed only in a limited period because the MEX orbit 92 is not dedicated for solar occultation observations, for instance its measurements during the 93 perihelion season in MY 28 (the year of the previous global dust storm) were performed only 94 during $Ls = 255-300^{\circ}$ and no observation is available during $Ls = 200^{\circ}-255^{\circ}$ and $300^{\circ}-360^{\circ}$ 95 (Fedorova et al., 2018); (2) the previous MRO/MCS analysis (Heavens et al., 2018) did not 96 directly retrieve water vapor vertical profiles from the water vapor spectral features: water vapor 97 abundances were rather indirectly estimated from the retrieved temperature, pressure, dust, and 98 water ice; (3) the 2009-2016 MRO/CRISM water vapor profiles, derived from O₂ dayglow 99 profiles, did not encounter global dust storm conditions. In order to understand the mechanism of 100 the water vapor transport from the near-surface to the middle atmosphere, it is crucial to 101 investigate the latitudinal, longitudinal, and temporal variation of water vapor vertical profiles 102 before/during/after global dust storms. Solar occultation measurements by two new 103 spectrometers onboard TGO - NOMAD (Vandaele et al., 2018) and Atmospheric Chemistry 104 Suite (ACS) (Korablev et al., 2018) - are now able to monitor the water vapor vertical profiles 105 through the whole a Martian Year and obtain a latitudinal map for every $\sim 20^{\circ}$ of Ls since the 106 orbit of TGO is optimized for solar occultation measurements, producing 24 occultations per day 107 at a maximum (average 5-7 observations per day). 108

In 2018, for the first time after 2007, a global dust storm occurred on Mars. It lasted for 109 more than two months (from June to August). Moreover, following the global dust storm, a 110 regional dust storm occurred in January 2019. TGO began its science operations on 21 April 111 112 2018. The NOMAD and ACS observations therefore fully cover the period before/during/after the global and regional dust storms and offer a unique opportunity to study the trace gases 113 distributions during the dust storms. In this paper, we present water vapor vertical profiles from 114 April to September 2018 and from December 2018 to February 2019 retrieved from the 115 NOMAD measurements. We have analyzed those datasets and published two water vapor 116 vertical profiles as early results - one before the global dust storm and the other one during the 117

storm, which present a significantly conspicuous increase of water vapor during the global dust storm (Vandaele et al., 2019). This study presents the results of the extended datasets at the period of the dust storms. The details of the NOMAD observations and the data analysis are described in Sections 2 and 3, respectively. The observational results are discussed in Sections 4 and 5 for the global dust storm and regional dust storm, respectively. A full GCM simulation is presented in an accompanying paper (Neary et al., 2019).

124 **2. Observations: NOMAD Solar occultation**

125 **2. 1. Instrument - NOMAD onboard TGO**

NOMAD is a spectrometer operating in the spectral ranges between 0.2 and 4.3 µm 126 onboard ExoMars TGO. NOMAD has 3 spectral channels: a solar occultation channel (SO -127 Solar Occultation; 2.3–4.3 µm), a second infrared channel capable of nadir, solar occultation, and 128 limb sounding (LNO – Limb Nadir and solar Occultation; 2.3–3.8 µm), and an ultraviolet/visible 129 channel (UVIS - Ultraviolet and Visible Spectrometer, 200-650 nm). The infrared channels (SO 130 131 and LNO) have high spectral resolution ($\lambda/d\lambda \sim 10,000-20,000$) provided by an echelle grating used in combination with an Acousto Optic Tunable Filter (AOTF) which selects diffraction 132 orders (Neefs et al., 2015). The concept of the infrared channels are derived from the Solar 133 Occultation in the IR (SOIR) instrument (Nevejans et al., 2006) onboard Venus Express (VEx). 134 The sampling rate for the solar occultation measurement is 1 second, which provides better 135 vertical sampling step (~1 km) with higher resolution (~2 km) from the surface to 200 km. 136 Thanks to the instantaneous change of the observing diffraction orders achieved by the AOTF, 137 the SO channel is able to measure five or six different diffraction orders per second in solar 138 occultation mode. One of the most remarkable capabilities of NOMAD is its high spectral 139 140 resolution in the near infrared range. It allows us (1) to investigate vertical profiles of the atmospheric constituents (such as carbon dioxide, carbon monoxide, water vapor, and their 141 isotopic ratio), and (2) to perform sensitive search of organic species (such as CH₄, C₂H₄, C₂H₆, 142 H₂CO) and other trace gases (such as HCl, HCN, HO₂, H₂S, N₂O, OCS) by solar occultation 143 measurements with the SO channel. 144

145 **2. 2. Dataset**

In this study, we analyze the solar occultation measurements acquired by the NOMAD 146 SO channel during the period from 21 April to 30 September 2018 (corresponding to Ls = 162-147 260° in Martian Year (MY) 34) and from 1 December 2018 to 23 February 2019 (Ls = 298-345° 148 in MY 34). Measurements of diffraction order 134 (3011–3035 cm⁻¹) and 168 (3775–3805 cm⁻¹) 149 are analyzed. Observations of these orders have been regularly conducted since they include 150 strong H_2O bands (both in orders 134 and 168) and CH_4 Q-branch (in order 134). In the period 151 above, a total of 987 occultations that operated with both diffraction orders 134 and 168 were 152 acquired. Figure 1 shows the latitudinal coverage of these occultations. As shown in this figure, 153 the latitude shifts from orbit to orbit, at the same time, the longitudinal coverage is dispersed 154 155 over the whole planet. It demonstrates that this dataset allows us to obtain a latitudinal map for every $\sim 20^{\circ}$ of Ls. The local solar time is generally around 6 AM or 6 PM outside of the polar 156 regions. Note that the gaps in data between $Ls = 171-179^\circ$, $201-210^\circ$, and $327-333^\circ$ are due to 157 the orbital geometry, which prevents solar occultations from being measured when the orbital 158 nadir track is close to the terminator and the Sun is never occulted by Mars. 159

160 **2. 3. Data reduction**

Solar occultation is a powerful technique for investigating vertical structure of 161 atmospheres. It observes a strong light source - the Sun - through the atmosphere from very high 162 altitude (typically from 250 km altitudes for NOMAD) down to the surface. The absolute 163 calibration of solar occultation spectra is relatively easy since transmittances are obtained by 164 dividing the spectra measured through the atmosphere by the reference solar spectrum recorded 165 outside the atmosphere, which basically removes systematic instrumental effects (except small 166 changes occurring during the occultation). For the calculation of transmittances, we employ the 167 algorithm developed for the reduction of the SOIR data (Trompet et al., 2016). This algorithm 168 does not simply average the spectra recorded outside the atmosphere but calculates its linear 169 regression with altitude for each pixel. The algorithm first calculates the pixel-by-pixel linear 170 regressions with the solar spectra between 150 and 250 km, it then applies them to the spectra 171 between 120 and 150 km where no absorption due to Mars atmosphere is expected. If more than 172 80 % of the transmittances at 120-150 km are equal to one sigma, the algorithm applies the linear 173 regression to the spectra recorded through the Mars atmosphere (below 120km) and calculates 174 the transmittances. If not, the algorithm re-calculates the linear regression by excluding the solar 175 spectrum at the highest altitude. This iterative process continues until accepted. Such a pixel-by-176 pixel linear extrapolation allows us to reduce the residual instrumental systematic due to small 177 178 changes during an occultation (such as small deviation of the center wavenumber of the AOTF transfer function, tiny spectral shift due to grating movement/expansion/contraction because of 179 instrumental oscillations). The instrumental noise is given by 180

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where I is the spectrum through the atmosphere, F is the reference solar spectrum, and I/F is thus 182 the transmittance. The noise in the reference solar spectrum F_{err} is given by the standard 183 deviation of the solar spectra (normalized) that are used to create the reference solar spectrum. 184 The noise in the spectrum through the atmosphere I_{err} is given by the sum of dark noise (we use 185 the standard deviation of the signal recorded when the radiation reaching the detector is similar 186 to the noise level when the planet is in the Line-Of-Sight (LOS)) and estimated noise from the 187 one in the reference solar spectrum (Vandaele et al., 2013). Typical Signal to Noise Ratio (SNR) 188 for a single spectrum recorded in diffraction orders 134 and 168 is 1500-2500. Figure 2 shows 189 an example of calculated transmittances of the diffraction order 168 during an occultation. When 190 the LOS to the Sun transects the atmosphere, the slant optical depth along the LOS gradually 191 increases owing to the presence of aerosols and molecules, until the atmosphere becomes 192 completely opaque at some tangent altitude. For the particular example in Figure 2, the 193 transmittance drops to zero around 5 km. It usually occurs because of the large amount of dust in 194 the lowermost part of the atmosphere. 195

196 **2. 4. Calibration**

197 Calibration of the NOMAD infrared channels from the first in-flight data was 198 summarized in Liuzzi et al. (2019) and has been improved since then. Based on the work by 199 Liuzzi et al. (2019), in this analysis, the AOTF transfer function of the NOMAD SO channel is 200 characterized as a sinc square function whose side lobes are multiplied by 1.3 and 1.8 for 201 diffraction order 134 and 168, respectively. The instrumental line shape is assumed to be a 202 Gaussian function with the full width at half maximum (FWHM) of 0.228 cm⁻¹ ($\lambda/\Delta\lambda$ ~13250) for diffraction order 134, and 0.338 cm⁻¹ ($\lambda/\Delta\lambda \sim 11220$) for diffraction order 168. The spectral calibration is first performed based on the results in Liuzzi et al. (2019) and then refined using the solar lines at 3014.960 cm⁻¹ (order 134) and 3878.865 cm⁻¹ (order 168) for each spectrum in the retrieval process.

3. Data analysis: retrievals of H₂O vertical profiles

208 **3. 1. Forward model**

For the forward calculation of the simulated spectra and inversion of the H_2O abundance, we employ the ASIMUT-ALVL radiative code developed at Royal Belgian Institute for Space Aeronomy (BIRA-IASB) (Vandaele et al., 2006). The code solves the radiative transfer equation for nadir or solar occultation geometries. It was originally developed for the Earth atmosphere and then extended for Venus (e.g., Vandaele et al., 2008) and Mars (e.g., Vandaele et al., 2019) atmospheres. The code has been widely used in the data analysis of the solar occultation measurements by VEx/SOIR.

The radiative transfer calculation is performed in the spectral ranges between ± 4 diffraction orders from the main order to properly model the contributions from adjacent orders (Vandaele et al., 2008). CO₂ and H₂O molecules absorption are taken into account in the calculation. The absorption coefficients of CO₂ and H₂O molecules are calculated based on the line-by-line method by using the following spectroscopic database: HITRAN 2016 database (Gordon et al. 2016) for CO₂ and the water line list for CO₂-rich atmospheres by Gamache et al. (2016) for H₂O. A Voigt function is adopted for the line shape function.

The observing geometry is calculated based on the SPICE kernel of the TGO orbits. 223 Based on the geometry (i.e., latitude, longitude, solar longitude, and the local solar time at the 224 tangential point), we extract vertical profiles of the temperature, pressure, and CO₂ volume 225 mixing ratio predicted by the general circulation model (GCM), Global Environmental 226 Multiscale Mars model (GEM-Mars) (Neary et al., 2019; Daerden et al., 2019; Neary and 227 Daerden, 2018). This is performed for each spectrum corresponding to the probed tangent 228 altitude. The modeled atmosphere starts from the tangent altitude of the measurement and up to 229 150 km altitude with a uniform thickness of 5 km. Note that the tangent altitude is calculated as 230 the shortest distance between the LOS of the center of the field of view and the MGM1025 231 232 areoid (i.e., the Mars geoid) (Lemoine et al., 2001). In addition, we note that the atmospheric state predicted by GEM-Mars takes into account the effects of the dust storms in MY34 (Neary 233 et al., 2019). Neary et al. (2019) described multiple dust storm simulations. We used the run 234 235 where dust is scaled to the MY34 climatology and the vertical profile was determined using an adjusted Conrath profile (Conrath parameter=0.0008; "GDS0008" simulation). 236

237 **3. 2. Retrievals**

We performed the retrieval of H_2O abundance for each spectrum at each tangential altitude independently (Vandaele et al., 2019), i.e., using the classical "onion peeling" method. The retrievals start at the top of atmosphere (120 km altitude for this study). At the highest altitude, the initial guess for the H_2O volume mixing ratio is set a small value (typically 1 ppm). Once H_2O absorption features are detected, the retrieved value is used as the initial guess for the next layer below. Since the slant optical depth integrated along the LOS is relatively large for solar occultation measurements comparing to nadir observations, strong H_2O lines (line intensity S larger than $\sim 10^{-20}$ cm⁻¹/ (molecule cm⁻²)) are easily saturated. In our retrieval scheme, if lines are saturated (i.e., the total optical depth at infinite resolution is greater than 1), their weights are reduced as follows:

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where $\sigma(v)$ is the instrumental noise, $\sigma'(v)$ is the de-weighted error used in the retrievals, and $\tau(v)$ is the total slant optical depth integrated along the LOS. De-weighting pixels that contain lines whose optical depth at line center before considering the instrument exceeds 1 would be a strong constraint, however, it is useful to reduce possible biases and uncertainties associated with imperfect knowledge of instrumental functions.

The retrievals are performed using the Optimal Estimation Method (OEM) (Rodgers, 254 2000) implemented in a Gauss-Newton iterative scheme. The spectral ranges for the retrieval are 255 the full range of diffraction order 134 (3011–3035 cm⁻¹) and a confined spectral range (3783– 256 3803 cm⁻¹) for diffraction order 168, where more than 50 % of the recorded signal originate from 257 the main diffraction order. This is because the side-lobes in the AOTF transfer function are 258 relatively large for diffraction order 168. Since the side-lobes are not yet perfectly characterized, 259 limiting the retrieval to the spectral region where the majority of signal comes from the main 260 order reduces the uncertainties associated with imperfect knowledge of the AOTF function. The 261 free parameters in the retrievals are a factor multiplying the initial guess of H₂O density, and the 262 parameters of the 5th order polynomial function used to model the continuum of each spectrum. 263 Note that most of the information comes from the sounded tangent altitude (about 70% of the 264 slant number density integrated over the LOS is within 4 km from the tangent height), thus the 265 retrieved local H₂O abundances at the tangential altitudes of the measurements can be considered 266 as its vertical profiles. The continuum established by the polynomial function removes the effect 267 of extinctions due to the presence of aerosols (dust and water ice clouds) along the LOS as well 268 as instrumental features caused by physical changes of the instrument during the occultation 269 (such as small deviation of the center wavenumber of the AOTF transfer function). Figure 3 270 shows examples of the fitting results. The intensity of the H₂O lines in the diffraction order 168 271 is about 100 times stronger ($S \sim 10^{-19} \text{ cm}^{-1}$ / (molecule cm⁻²)) than those in the diffraction order 272 134 (S ~ 10^{-21} cm⁻¹/ (molecule cm⁻²)), which allows us to investigate water vapor abundance 273 from the near-surface to high altitudes. As shown **Figure 3b**, we firmly detect H_2O with a good 274 SNR up to at least 100 km during the global dust storm. The retrievals are conducted for each 275 diffraction order (i.e., 134 and 168) independently in order to evaluate the consistency between 276 orders. Lastly the vertical profiles of H₂O abundances and their errors are calculated from the 277 278 weighted averages:

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280 where H_2O_{134} , H_2O_{168} , H_2O_{134err} and H_2O_{168err} are retrieved H₂O volume mixing ratio and 281 their error values from the diffraction order 134 and 168, respectively.

282 **3. 3. Uncertainties**

There are three sources of error in the retrieved H_2O volume mixing ratio: (1) instrumental noise on the measured spectra, (2) uncertainty in the vertical temperature and atmospheric density profile used in the radiative transfer calculation, and (3) uncertainty of imperfect knowledge of the NOMAD instrumental functions.

The first error is directly derived from the covariance matrix of the optimal fit parameters in the retrievals. If both diffraction orders 168 and 134 are available, the error values obtained from each diffraction order are weighted averaged, as described above. The median value of this error in the retrievals is about 5 %.

The second error can be evaluated by retrievals with temperature profiles shifted by their 291 accuracy. We estimate that the accuracy of the GEM-Mars temperature predictions is about ± 10 292 K. The difference between the water vapor profiles retrieved with the temperature profiles 293 294 uniformly shifted by ± 10 K and those with retrieved with the original temperature profiles are about 5-8 %, which can then be considered as the error in the retrieved H₂O volume mixing ratio 295 due to uncertainty in the GEM-Mars temperature. Moreover, since the H₂O volume mixing ratio 296 297 is the ratio between the water vapor number density and the total atmospheric number density that is based on the predictions by GEM-Mars in this study, the uncertainty in the GEM-Mars 298 total atmospheric number density directly affects the retrieved H₂O volume mixing ratio. We 299 estimate that the accuracy of the atmospheric total number density by GEM-Mars is about 10-15 300 %, thus we have another 10-15 % of error in the retrieved H₂O volume mixing ratio. 301

The third error can be roughly estimated by comparing the retrieved H₂O abundances in 302 each diffraction order (168 and 134). Figure 4 shows examples of the retrieved vertical profiles 303 of the H₂O volume mixing ratio from diffraction orders 134 and 168. The retrieved profiles from 304 each diffraction order are predominantly consistent within 3-sigma (as shown in Fig. 4a), 305 however, the retrieved values are sometimes inconsistent beyond 3-sigma of the standard 306 retrieval error (as shown in Fig. 4b). The median value of the difference between order 168 and 307 134 retrievals is about twice larger than 1-sigma of the standard retrieval error, and inconsistent 308 results above 3-sigma occur in around 35% of occultations. We consider this is mainly due to the 309 fact that instrumental characterization such as the AOTF transfer function is not fully achieved, 310 however this does not invalidate the conclusions of this study. 311

312 **4. The global dust storm in 2018**

313 **4. 1. Overview of the dust storm**

The global picture of the 2018 global dust storm was summarized in Guzewich et al. 314 (2019) and references therein. The storm started regionally at the middle/end of May ($Ls \sim 180$ -315 185°) across Acidalia Planitia (30-60°N, 300-360°E) and Utopia Planitia (30-60°N, 80-140°E). 316 Then, it merged with substantial dust lifting occurring independently in the southern hemisphere 317 by the beginning of June ($Ls \sim 190^\circ$), and expanded globally by the middle of June ($Ls \sim 195^\circ$). 318 This most active period of the global dust storm lasted until the beginning of July ($Ls \sim 205^{\circ}$), 319 and started a long decay phase that ended at the middle of September ($Ls \sim 250^{\circ}$). As shown in 320 321 Figure 1a, the measurements by the NOMAD-SO channels cover the whole period of the global dust storm except for some gaps due to the geometry of the spacecraft's orbit, including the most 322 active period of the dust storm ($Ls = 202-210^{\circ}$). The colors in **Figure 1a** denote the highest 323 tangent altitude at which the slant-optical depth along the LOS is less than 1, which is basically 324

representative of the top altitude of the most opaque dust region in the atmosphere. The dust top altitudes reaches ~50 km in the seasonal range between $Ls = 197-202^{\circ}$, which corresponds to the most active period of the global dust storm.

4. 2. Seasonal variation of the water vapor vertical profiles

Figure 5a-b shows the seasonal variation of the water vapor vertical profiles at *Ls*=162– 329 260° retrieved from the NOMAD measurements taken in the northern hemisphere (Fig. 5a) and 330 the southern hemisphere (Fig. 5b). The top panels show the latitude and local time of the 331 measurements as references. It would be interesting to explore the changes in the water vapor 332 vertical distribution during the 2018 dust storm and for the same period in a non-dust storm year. 333 However, as the 2018 storm occurred in the first year of operations of NOMAD, we have no 334 direct self-consistent reference for the non-dust storm conditions. Previous directly and indirectly 335 retrieved water vapor profiles (see introduction) are sparse and the match in space and time with 336 337 the NOMAD profiles is in general poor. Therefore, we prefer to use a GCM as tentative reference for the non-dust storm water vapor distribution, as it provides a complete coverage for 338 all times and allows for a full interpolation of the model output to the location and time of all the 339 340 NOMAD profiles. The vertical water vapor distribution simulated in the GEM-Mars GCM was evaluated by both the water vapor vertical profiles and total water columns retrieved from 341 CRISM in Daerden et al. (2019). Figure 5c-d shows the same water maps as Fig. 5a-b but 342 simulated by GEM-Mars for non-dust storm conditions (Daerden et al., 2019). Figure 5e-f 343 presents the differences between the measured water vapor vertical profiles (Fig. 5a-b) and the 344 345 predictions by GEM-Mars for non-dust storm conditions (Fig. 5c-d).

The abundance of water vapor in the middle atmosphere suddenly increased around Ls =346 190° in both hemispheres. This is not seen in the GCM data for non-dust storm conditions (Fig. 347 5c-d). In contrast, around $Ls = 210^{\circ}$, the water vapor abundance in the lower atmosphere seems 348 to have decreased with respect to non-dust storm conditions. However, the retrieval accuracy and 349 data coverage is poorer at low altitudes. Also, a comparison of the GCM water vapor profiles 350 with those derived from CRISM (Clancy et al., 2017; Daerden et al., 2019) in non-dust storm 351 conditions shows that the GCM water vapor is too abundant at low latitudes. Therefore, it 352 remains hard to estimate the effect of the dust storm on the behavior of water vapor in the lower 353 atmosphere using the current GCM results. GCM simulations for the MY34 dust storm that 354 reproduce the observed profiles reasonably well are presented in the accompanying paper Neary 355 et al. (2019) and allow for a theoretical assessment of the impact of the dust storm. 356

The variation of the water vapor abundances occurs very rapidly: the water abundance in 357 the middle atmosphere in both hemispheres increases by an order of magnitude in just a few days 358 (around $Ls = 195^{\circ}$). Since the timing of this phenomenon corresponds to the onset of the global 359 dust storm and this is not predicted by the GCM for non-dust storm conditions, we conclude that 360 the rapid enhancement of the water vapor in the middle atmospheres is due to the effects of the 361 global dust storm. The water abundances in the middle atmosphere have maximum values 362 around $Ls = 200^{\circ}$. At that period, we find large water vapor abundances in the middle 363 atmosphere, exceeding 200 ppm. Such large water vapor abundances are similar to 364 MEx/SPICAM observation during the global dust storm in MY 28 (Fedorova et al., 2018). 365 Moreover, we detect water vapor at very high altitude, reaching at 100 km with a volume mixing 366 ratio of ~50 ppm. After these peaks, the enhanced water vapor in the middle atmosphere 367 gradually returned to the typical climatological levels. A small local maximum in water vapor 368

also appears around $Ls = 235-240^{\circ}$ in the northern hemisphere (Fig. 5a). This is due to the fact 369 370 that the measurements are performed at equatorial region where more water vapor is present. While in contrast, the water vapor abundances in the southern hemisphere have a small local 371 372 maximum around $Ls = 230^\circ$, which can be explained by the fact that the water vapor in the southern hemisphere does not have a maximum at equatorial but at high latitude (see Section 4.3. 373 in detail). Finally, we note that it is difficult to distinguish local time variation from the seasonal 374 one since either sunrise or sunset measurements last for 10-20° of Ls as shown in the top panel of 375 Fig. 5a-b. 376

4. 3. Latitudinal variation of the water vapor vertical profiles

Figure 6 shows the seasonal variation of latitudinal maps of the water vapor vertical 378 profiles observed by NOMAD (the top panel) and predicted by GEM-Mars for non-dust storm 379 conditions (the bottom panel) during $Ls = 160-195^{\circ}$ (before the global dust storm, Fig. 6a), Ls =380 381 195–202° (growth phase of the dust storm, Fig. 6b), $Ls = 210-220^{\circ}$ (mature phase of the dust storm, Fig. 6c), $Ls = 220-240^{\circ}$, and $Ls = 240-260^{\circ}$ (decay phase of the dust storm, Fig. 6d and 382 6e). The latitudinal distributions of the water vapor vertical profiles before and after the onset of 383 384 the global dust storm are quite different (Fig. 6a, b). After the onset of the global dust storm, the water vapor abundance in the middle atmosphere is significantly and consistently increased at 385 60°S-60°N. At the mature phase of the dust storm (Fig. 6c), the water vapor abundance is 386 decreased compared to the growth phase, however the latitudinal distribution of the water vapor 387 is similar. At the growth/mature phase of the dust storm, the season on Mars is just following the 388 northern autumn equinox and it is expected that the Hadley circulation (the equatorial cell) 389 extends to 60° latitude in both hemispheres (e.g., Forget et al., 1999; Takahashi et al., 2003). This 390 explains that the large water vapor abundance is confined between 60°S-60°N. Under non-dust 391 storm conditions, water vapor abundances decrease rapidly by ~40 km altitude due to saturation 392 conditions at 30-50 km. Our results suggest that during the dust storm water vapor was 393 transported from the lower to the middle atmosphere by the intensified circulation and expanded 394 above 40 km due to the heating of the middle atmosphere by dust absorption that prevents water 395 vapor from condensation (Neary et al., 2019). In addition, it has been proposed that strong 396 397 convective transport processes at the mesoscale can create high altitude dust layers (Spiga et al., 2013; Daerden et al., 2015; Wang et al., 2018; Heavens et al., 2018). Such processes will also 398 transport water vapor and may also contribute to the formation of local high altitude water vapor 399 maxima. At the decay phase of the dust storm (Fig. 6d, 6e), the water vapor abundance is similar 400 to that at the mature phase, however the latitudinal distribution has a gradient with a maximum 401 value at latitudes greater than 60°S. It is well known that the symmetry between two meridional 402 403 "equinox" cells is significantly reduced at $Ls = 220^\circ$, as the formation of a single pole-to-pole "solstice" Hadley circulation develops (e.g., Forget et al., 1999; Takahashi et al., 2003). The local 404 maximum observed at latitudes greater than 60°S may be explained by a new theoretical study 405 performed by Shaposhnikov et al. (2019). Although their study focused on the MY 28 global dust 406 storm which occurred later in the season ($Ls = 250^{\circ} - 270^{\circ}$), the authors suggest that transport of 407 water vapor to the upper atmosphere by the strong upward branch of the meridional circulation at 408 perihelion occur only at latitudes greater than 60°S, which corresponds to the location of the 409 peak of water vapor abundances observed by NOMAD. 410

411 **5. The regional dust storm in 2019**

412 **5. 1. Overview of the dust storm**

A 2019 regional dust storm started around 7 January ($Ls \sim 320^{\circ}$) in the southern 413 hemisphere, peaked around 15 January, and declined into the middle of February ($Ls \sim 340^\circ$) 414 (Chaffin et al., 2019). This seasonal dust event occurs with significantly variable amplitude in 415 every Mars year, and can present vertically deep increases in atmospheric temperatures and dust 416 over low-to-mid latitudes (e.g., Kass et al., 2016), as it did in 2019. As shown in Figure 1b, the 417 measurements by the NOMAD-SO channel cover the whole period of the dust storm except for a 418 gap due to the geometry of the spacecraft's orbit at the active period of this regional dust storm 419 $(Ls = 327 - 333^{\circ})$. The dust top altitude reaches ~50 km in the seasonal range between Ls = 322-420 327° in the southern hemisphere, which corresponds to the period and the region of the dust 421 storm (reddish colors in Fig. 1b). 422

423 **5. 2. Seasonal variation of the water vapor vertical profiles**

424 Figure 7a-b shows the seasonal variation of the water vapor vertical profiles at Ls=298-345° retrieved from the NOMAD measurements taken in the northern hemisphere (Fig. 7a) and 425 southern hemisphere (Fig. 7b). Figure 7c-d shows the same water maps as Fig. 7a-b but 426 simulated by GEM-Mars for non-dust storm conditions (Daerden et al., 2019). Figure 7e-f 427 presents the differences between the measured water vapor vertical profiles (Fig. 7a-b) and the 428 predictions by GEM-Mars for non-dust storm conditions (Fig. 7c-d). In the time before the storm 429 (Ls~300° in the north and Ls~305-320° in the south) and also at Ls~240-260° in the southern 430 hemisphere (Fig. 5b), the GCM water mixing ratios between 10 to 40 km are considerably higher 431 (by a factor ~2) than those measured by NOMAD. However total water columns from the GCM 432 match very well with observations from TES and CRISM (Neary and Daerden, 2018; Smith et 433 al., 2018; Daerden et al., 2019). This suggests that water vapor was much more confined to the 434 lowest scale height than currently predicted by models. The water vapor abundances in the 435 middle atmosphere suddenly increased around $Ls = 321^{\circ}$ in both hemispheres and reached 436 maximum values around $Ls = 325^{\circ}$. The enhancement lasted at least until $Ls = 327^{\circ}$, before a 437 data gap due to orbital geometry. The GCM data also show a distinct increase of water vapor 438 abundances up to 40 km in this period. However, the increase of water vapor seen in the GCM is 439 due to the latitude of the measurements (low latitudes) and does not expand above 40 km 440 altitude. Since the timing of the enhancement of the water vapor in the middle atmosphere seen 441 by NOMAD corresponds to the period of the regional dust storm and since it expands into the 442 whole middle atmosphere, it is reasonable to attribute that enhancement to the regional dust 443 storm. At the period of this storm, water vapor abundances in the middle atmosphere exceed 150 444 ppm and water vapor is present up to (at least) 90 km. The water vapor abundances in the middle 445 atmosphere are smaller than those in the 2018 global dust storm and the top altitude is lower. 446

447 5. 3. Latitudinal variation of the water vapor vertical profiles

Figure 8 shows the latitudinal map of the water vapor vertical profiles measured by NOMAD and predicted by GEM-Mars for non-dust storm condition at $Ls = 300-320^{\circ}$ (Fig. 8a), $Ls = 320-330^{\circ}$ (Fig. 8b, at the time of the regional dust storm), and $Ls = 330-345^{\circ}$ (Fig. 8c). It is found that the water vapor is significantly increased in the middle atmosphere at the time of the regional dust storm with the maximum values at latitudes greater than 60°S (Fig. 8b). The meridional circulation over the period of this regional dust storm ($Ls = 322-327^{\circ}$) is still expected to be a single pole-to-pole "solstice" cell. Thus, the mechanism may be the same as for the decay phase of the 2018 global dust storm, i.e., the strong upward branch of the meridional circulation, as proposed by Shaposhnikov et al. (2019), may be responsible for the local maximum observed at latitudes greater than 60°S.

458 **6.** Conclusions

We have analyzed a selection of the first year solar occultation measurements by TGO/NOMAD and have presented variations in the vertical profile of water vapor on Mars, including periods of the global dust storm in 2018 and the following regional storm in 2019. The main results are:

- 463 1. We find a rapid and significant increase of water vapor in the middle atmosphere 464 following the onset of the global dust storm (around $Ls = 190^{\circ}$). The enhancement of 465 water vapor at the most active period of the dust storm (around $Ls = 200^{\circ}$) is 466 remarkable: the water vapor reaches very high altitudes (100 km).
- 467 2. The latitudinal variation of water vertical profiles during the growth and mature 468 phases of the global dust storm ($Ls = 196-202^{\circ}$ and $210-220^{\circ}$) shows that the water 469 vapor abundance in the middle atmosphere is consistently increased at 60° S- 60° N. 470 The overall increase in middle atmospheric water vapor likely reflects a breakdown 471 in the 30–50 km hygropause trapping of water vapor due to dust driven atmospheric 472 temperature increases. A full simulation in a GCM supporting this explanation is 473 presented in an accompanying paper (Neary et al., 2019).
- 474 3. The latitudinal variation of water vertical profiles during the decay phases of the 475 global dust storm ($Ls = 220-240^{\circ}$ and $240-260^{\circ}$) shows that the water vapor 476 abundance in the middle atmosphere has peaks at latitudes greater than 60° S. At this 477 time, the global circulation is beginning its transition from an equinoctial to a 478 (southern summer) solstitial Hadley circulation pattern. Strong upward branch of the 479 circulation at latitudes greater than 60° S (Shaposhnikov et al., 2019) may be the 480 responsible for the local maximum.
- 481 4. We also find a conspicuous enhancement of water vapor in the middle atmosphere 482 during the regional dust storm in 2019 (around $Ls = 325^{\circ}$). The magnitude of the 483 water vapor increase is not as large as in the global dust storm but still remarkable: 484 water vapor increases reach ~90 km. Again, this behavior is indicative of dust storm 485 heating conditions.
- 486 5. Water vertical profiles observed during this regional dust storm ($Ls = 322-327^{\circ}$) 487 exhibit peak upper level water vapor abundance at high southern latitudes (60°S), 488 which can be explained by the upward branch of the solstitial Hadley circulation. 489 This annually reoccurring dust storm behavior exhibits strong interannually 490 variability (Kass et al., 2016), and appears to have been particularly intense in this 491 Mars Year (34).

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- 511 al., 2019).

512 **References**

- Aoki, S., Vandaele, A. C., & Daerden, F. Vertical profiles of water vapor in the Martian atmosphere during dust storms in 2018-2019 observed by TGO/NOMAD, presented in Aoki et al., JGR, 2019. Royal Belgian Institute for Space Aeronomy. https://doi.org/10.18758/71021054.
- Chaffin, M. S., J.-Y. Chaufray, I. Stewart, F. Montmessin, N. M. Schneider, and J.-L. Bertaux,
 (2014), Unexpected variability of Martian hydrogen escape. Geophysical Research
 Letters 41, 314-320. doi: 10.1002/2013GL058578.
- Chaffin, M. S., J. Deighan, N. M. Schneider, and A. I. F. Stewart (2017), Elevated atmospheric
 escape of atomic hydrogen from Mars induced by high-altitude water, Nature geoscience,
 10, 174-178, doi: 10.1038/ngeo2887.
- Chaffin, M. S., D. M. Kass, S. Aoki, A. A. Fedorova, J. Deighan, J.-Y. Chaufray, K. Connour, N.
 G. Heavens, A. Kleinböhl, S. K. Jain, M. Mayyasi, J. T. Clarke, N. M. Schneider, B.
 Jakosky, G. Villanueva, G. Liuzzi, F. Daerden, I. R. Thomas, J.-J. Lopez-Moreno, M. R.
 Patel, G. Bellucci, A.C. Vandaele, A. Trokhimovskiy, F. Montmessin, and O. I. Korablev
 (2019), Mars climate controls atmospheric escape: dust-driven escape from surface to
 space with MRO/MCS, TGO/NOMAD, TGO/ACS, and MAVEN/IUVS, The Ninth
 International Conference on Mars abstract #6312.
- Clancy, T., R., M. D. Smith, F. Lefèvre, T. H. McConnochie, B. J. Sandor, M. J. Wolff, S. W. Lee,
 S. L. Murchie, A. D. Toigo, H. Nair, and T. Navarro (2017), Vertical profiles of Mars 1.27
 µm O₂ dayglow from MRO CRISM limb spectra: Seasonal/global behaviors,
 comparisons to LMDGCM simulations, and a global definition for Mars water vapor
 profiles, Icarus, 293, 132-156, doi: 10.1016/j.icarus.2017.04.011.
- Clarke, J. T., J.-L. Bertaux, J.-Y. Chaufray, G. R. Gladstone, E. Quemerais, J. K. Wilson, and D.
 Bhattacharyya, (2014), A rapid decrease of the hydrogen corona of Mars: the Martian

- 537
 Hydrogen
 Corona.
 Geophysical
 Research
 Letters
 41,
 8013-8020,
 doi:

 538
 org/10.1002/2014GL061803
 org/10.1002/2014GL061803
- Clarke, J. T. (2018), Dust-enhanced water escape, Nature Astronomy, 2, 114-115, doi:
 10.1038/s41550-018-0383-6.
- Daerden F., J. A. Whiteway, L. Neary, L. Komguem, M. T. Lemmon, N. G. Heavens, B. A.
 Cantor, E. Hébrard, and M. D. Smith (2015), A solar escalator on Mars: Self-lifting of dust layers by radiative heating, Geophysical Research Letters, Volume 42, Issue 18, pp. 7319-7326, doi: 10.1002/2015GL064892
- 545 Daerden, F., L. Neary, S. Viscardy, A. García Muñoz, R. T. Clancy, M. D. Smith, T. Encrenaz,
 546 and A. Fedorova (2019), Mars atmospheric chemistry simulations with the GEM-Mars
 547 general circulation model, Icarus, 326, 197-224, doi: 10.1016/j.icarus.2019.02.030.
- Fedorova, A., J.-L. Bertaux, D. Betsis, F. Montmessin, O. Korablev, L. Maltagliati, and J. Clarke
 (2018), Water vapor in the middle atmosphere of Mars during the 2007 global dust storm.
 Icarus, 300, 440-457, doi: 10.1016/j.icarus.2017.09.025.
- Forget, F., F. Hourdin, R. Fournier, C. Hourdin, O. Talagrand, M. Collins, S. R. Lewis, P. L.
 Read, and J. and Huot (1999), Improved general circulation models of the Martian atmosphere from the surface to above 80 km. Journal of Geophysical Research Planets 104, 24155-24176.
- Gamache, R. R., M. Farese, and C. L. Renaud (2016), A spectral line list for water isotopologues
 in the 1100-4100 cm⁻¹ region for application to CO₂-rich planetary atmospheres, Journal
 of Molecular Spectroscopy, 326, 144-150, doi: 10.1016/j.jms.2015.09.001.
- Gordon, I. E., L. S. Rothman, C. Hill, R. V. Kochanov, Y. Tan, P. F. Bernath, M. Birk, V. Boudon, 558 A. Campargue, K. V. Chance, B. J. Drouin, J.-M. Flaud, R. R. Gamache, J. T. Hodges, D. 559 Jacquemart, V. I. Perevalov, A. Perrin, K. P. Shine, M.-A. H. Smith, J. Tennyson, G. C. 560 Toon, H. Tran, V. G. Tyuterev, A. Barbe, A. G. Császár, V. M. Devi, T. Furtenbacher, J. J. 561 Harrison, J.-M. Hartmann, A. Jolly, T. J. Johnson, T. Karman, I. Kleiner, A. A. Kyuberis, 562 J. Loos, O. M. Lyulin, S. T. Massie, S. N. Mikhailenko, N. Moazzen-Ahmadi, H. S. P. 563 Müller, O. V. Naumenko, A. V. Nikitin, O. L. Polyansky, M. Rey, M. Rotger, S. W. 564 Sharpe, K. Sung, E. Starikova, S. A. Tashkun, J. Vander Auwera, G. Wagner, J. 565 Wilzewski, P. Wcisło, S.Yu, and E. J. Zak (2017), The HITRAN2016 Molecular 566 Spectroscopic Database, Journal of Quantitative Spectroscopy and Radiative Transfer, 567 203, 3-69, doi: 10.1016/j.jqsrt.2017.06.038. 568
- Guzewich, S. D., M. Lemmon, C. L. Smith, G. Martínez, Á. de Vicente-Retortillo, C. E.
 Newman, M. Baker, C. Campbell, B. Cooper, J. Gómez-Elvira, A.-M. Harri, D. Hassler,
 F. J. Martin-Torres, T. McConnochie, J. E. Moores, H. Kahanpää, A. Khayat, M. I.
 Richardson, M. D. Smith, R. Sullivan, M. de la Torre Juarez, A. R. Vasavada, D.
 Viúdez-Moreiras, C. Zeitlin, and Maria-Paz Zorzano Mier (2019), Mars Science
 Laboratory Observations of the 2018/Mars Year 34 Global Dust Storm, Geophysical
 Research Letters, 46, 1, 71-79, doi: 10.1029/2018GL080839.
- Heavens, N., G., A. Kleinböhl, M. S. Chaffin, J. S. Halekas, D. M. Kass, P. O. Hayne, D. J.
 McCleese, S. Piqueux, J. H. Shirley, and J. T. Schofield (2018), Hydrogen escape from

- 578 Mars enhanced by deep convection in dust storms, Nature Astrononomy, 2, 126-132, doi: 579 10.1038/s41550-017-0353-4.
- Kass, D. M., A. Kleinboehl, D. J. McCleese, J. T. Schofield, and M. D. Smith (2016),
 Interannual similarity in the Martian atmosphere during the dust storm season, Geophys.
 Res. Lett., 43, 6111-6118, doi:10.1002/2016GL068978.
- 583 Korablev, O., F. Montmessin, A. Trokhimovskiy, A. A. Fedorova, A. V. Shakun, A. V. Grigoriev, B. E. Moshkin, N. I. Ignatiev, F. Forget, F. Lefèvre, K. Anufreychik, I. 584 Dzuban, Y. S. Ivanov, Y.K. Kalinnikov, T.O. Kozlova, A. Kungurov, V. Makarov, F. 585 Martynovich, I. Maslov, D. Merzlyakov, P.P. Moiseev, Y. Nikolskiy, A. Patrakeev, D. 586 Patsaev, A. Santos-Skripko, O. Sazonov, N. Semena, A. Semenov, V. Shashkin, A. 587 Sidorov, A. V. Stepanov, I. Stupin, D. Timonin, A. Y. Titov, A. Viktorov · A. Zharkov, 588 F. Altieri, G. Arnold, D. A. Belyaev, J.-L. Bertaux, D. S. Betsis, N. Duxbury, T. 589 Encrenaz, T. Fouchet, J.-C. Gérard, D. Grassi, S. Guerlet, P. Hartogh, Y. Kasaba, I. 590 Khatuntsev, V. A. Krasnopolsky, R. O. Kuzmin, E. Lellouch, M. A. Lopez-Valverde, M. 591 Luginin, A. Määttänen, E. Marcq, J. Martin Torres, A. S. Medvedev, E. Millour, K. S. 592 Olsen, M. R. Patel, C. Quantin-Nataf, A. V. Rodin, V. I. Shematovich, I. Thomas, N. 593 Thomas, L. Vazquez, M. Vincendon, V. Wilquet, C.F. Wilson, L.V. Zasova, L.M. 594 Zelenyi, M.P. Zorzano (2016), The Atmospheric Chemistry Suite (ACS) of Three 595 596 Spectrometers for the ExoMars 2016 Trace Gas Orbiter, Space Science Reviews, 214, 1, article id. 7, 62, doi: 10.1007/s11214-017-0437-6. 597
- Lemoine, F. G., D. E. Smith, D. D. Rowlands, M. T. Zuber, G. A. Neumann, D. S. Chinn, and D.
 E. Pavlis (2001), An improved solution of the gravity field of Mars (GMM-2B) from Mars Global Surveyor, Journal of Geophysical Research, 106, E10, 23359-23376, doi: 10.1029/2000JE001426.
- Liuzzi, G., G. L. Villanueva, M. J. Mumma, M. D. Smith, F. Daerden, B. Ristic, I. Thomas, A. C.
 Vandaele, M. Patel, J.-J. Lopez-Moreno, G. Bellucci, and the NOMAD Team (2019),
 Methane on Mars: New insights into the sensitivity of CH₄ with the NOMAD/ExoMars
 spectrometer through its first in-flight calibration, Icarus, 321, 671-690, doi:
 10.1016/j.icarus.2018.09.021.
- Maltagliati, L., F. Montmessin, A. Fedorova, O. Korablev, F. Forget, and J.-L. Bertaux (2011),
 Science, 333, 6051, 1868-, doi: 10.1126/science.1207957
- Maltagliati, L. F. Montmessin, O. Korablev, A. Fedorova, F. Forget, A. Määttänen, F. Lefèvre,
 and J.-L. Bertaux (2013), Annual survey of water vapor vertical distribution and water aerosol coupling in the martian atmosphere observed by SPICAM/MEx solar
 occultations, Icarus, 223, 2, 942-962, doi: 10.1016/j.icarus.2012.12.012
- Neary L., and F. Daerden (2018), The GEM-Mars general circulation model for Mars:
 Description and evaluation, Icarus, 300, 458-476, doi: 10.1016/j.icarus.2017.09.028.
- Neary L., F. Daerden, F. Daerden, S. Aoki, J. Whiteway, R. T. Clancy, M. Smith, S. Viscardy, J.
 T. Erwin, I. R. Thomas, G. Villanueva, G. Liuzzi, M. Crismani, M. Wolff, S. R. Lewis,
 J. A. Holmes, M. R. Patel, M. Giuranna, C. Depiesse, A. Piccialli, S. Robert, L.
 Trompet, Y. Willame, B. Ristic, and A. C. Vandaele (2019), Explanation for the increase
 in high altitude water on Mars observed by NOMAD during the 2018 global dust storm,
 Geophysical Research Letters, *accepted*, doi: 10.1029/2019GL084354.

Neefs, E., A. C. Vandaele, R. Drummond, I. Thomas, S. Berkenbosch, R. Clairquin, S. 621 622 Delanoye, B. Ristic, J. Maes, S. Bonnewijn, G. Pieck, E. Equeter, C. Depiesse, F. Daerden, E. Van Ransbeeck, D. Nevejans, J. Rodriguez, J.-J. Lopez-Moreno, R. Sanz, R. 623 Morales, G.P. Candini, C. Pastor, B. Aparicio del Moral, J.M. Jeronimo, J. Gomez, I. 624 Perez, F. Navarro, J. Cubas, G. Alonso, A. Gomez, T. Thibert, M. R. Patel, G. Belucci, L. 625 De Vos, S. Lesschaeve, N. Van Vooren, W. Moelans, L. Aballea, S. Glorieux, A. Baeke, 626 D. Kendall, J. De Neef, A. Soenen, P.Y. Puech, J. Ward, J.F. Jamoye, D. Diez, A. 627 Vicario, and M. Jankowski (2015), NOMAD spectrometer on the ExoMars trace gas 628 orbiter mission: part 1 - design, manufacturing and testing of the infrared channels, 629 Applied Optics, 54 (28), 8494-8520, doi: 10.1364/AO.54.008494 630

- Nevejans, D., E. Neefs, E. Van Ransbeeck, S. Berkenbosch, R. Clairquin, L. De Vos, W.
 Moelans, S. Glorieux, A. Baeke, O. Korablev, I. Vinogradov, Y. Kalinnikov, B. Bach, J.P. Dubois, and E. Villard (2006), Compact high-resolution spaceborne echelle grating
 spectrometer with acousto-optical tunable filter based order sorting for the infrared
 domain from 2.2 to 4.3 μm, Applied Optics, 45 (21), 5191-5206, doi:
 10.1364/AO.45.005191.
- Rodgers, C. D. (2000), Inverse Methods for Atmospheric Sounding Theory and Practice,
 Inverse Methods for Atmospheric Sounding Theory and Practice. Series: Series on
 Atmospheric Oceanic and Planetary Physics, ISBN: 9789812813718. World Scientific
 Publishing Co. Pte. Ltd., Edited by Clive D. Rodgers, vol. 2, doi:
 10.1142/9789812813718.
- Shaposhnikov, D. S., A. S. Medvedev, A. V. Rodin, and P. Hartogh (2019), Seasonal Water
 "Pump" in the Atmosphere of Mars: Vertical Transport to the Thermosphere, Geophysical Research Letter, 46, 8, 4161-4169, doi: 10.1029/2019GL082839
- Smith, M. D., B. J. Conrath, J. C. Pearl, and P. R. Christensen (2002), NOTE: Thermal Emission
 Spectrometer Observations of Martian Planet-Encircling Dust Storm 2001A, Icarus, 157,
 1, 259-263, doi: 10.1006/icar.2001.6797.
- Spiga A., J. Faure, J.-B Madeleine, A. Määttänen, and F. Forget (2013), Rocket dust storms and
 detached dust layers in the Martian atmosphere, Journal of Geophysical Research:
 Planets, Volume 118, Issue 4, pp. 746-767, DOI: 10.1002/jgre.20046.
- Takahashi, Y. O., H. Fujiwara, H. Fukunishi, M. Odaka, Y.-Y. Hayashi, and S. Watanabe (2003),
 Topographically induced north-south asymmetry of the meridional circulation in the
 Martian atmosphere, Journal of Geophysical Research (Planets), 108, E3, doi:
 10.1029/2001JE001638.
- Trompet, L., A. Mahieux, B. Ristic, S. Robert, V. Wilquet, I. R. Thomas, A. C. Vandaele, and J. L. Bertaux (2016), Improved algorithm for the transmittance estimation of spectra obtained with SOIR/Venus Express, Applied Optics, 55, 32, 9275, doi: 10.1364/AO.55.009275.
- Vandaele, A. C., M. Kruglanski, and M. De Maziere (2006), Simulation and retrieval of
 atmospheric spectra using ASIMUT, paper presented at Atmospheric Science Conference,
 Eur. Space Agency, Frascati, Italy.

- Vandaele, A. C., M. De Mazière, R. Drummond, A. Mahieux, E. Neefs, V. Wilquet, O.
 Korablev, A. Fedorova, D. Belyaev, F. Montmessin, and J.-L. Bertaux (2008),
 Composition of the Venus mesosphere measured by Solar Occultation at Infrared on
 board Venus Express, J. Geophys. Res., 113, E00B23, doi: 10.1029/2008JE003140.
- Vandaele, A. C., A. Mahieux, S. Robert, S. Berkenbosch, R. Clairquin, R. Drummond, V.
 Letocart, E. Neefs, B. Ristic, V. Wilquet, F. Colomer, D. Belyaev, and J. -L. Bertaux
 (2013), Improved calibration of SOIR/Venus Express spectra, Optics Express, 21 (18),
 21148-21161, doi: 10.1364/OE.21.021148.
- Vandaele, A. C., J.-J. Lopez-Moreno, M.R. Patel, G. Bellucci, M. Allen, G. Alonso-Rodrigo, F. 670 Altieri, S. Aoki, D. Bolsée, T. Clancy, E. Cloutis, F. Daerden, C. Depiesse, R. 671 Drummond, A. Fedorova, V. Formisano, B. Funke, A. Geminale, J.-C. Gérard, M. 672 Giuranna, N. Ignatiev, J. Kaminski, O. Karatekin, Y. Kasaba, M. Leese, F. Lefèvre, S. 673 Lewis, M. López-Puertas, M. López-Valverde, A. Mahieux, J. Mason, J. McConnell, M. 674 Mumma, L. Neary, E. Neefs, E. Renotte, B. Ristic, S. Robert, J. Rodriguez-Gomez, G. 675 Sindoni, M. Smith, A. Stiepen, I. R. Thomas, A. Trokhimovsky, J. Vander Auwera, G. 676 Villanueva, S. Viscardy, J. Whiteway, Y. Willame, V. Wilquet, M. Wolff, and the 677 NOMAD Team (2018), NOMAD, an integrated suite of three spectrometers for the 678 ExoMars Trace Gas mission: technical description, science objectives and expected 679 performance, Space Science Reviews, 214, 5, article id. 80, 47 pp, doi: 10.1007/s11214-680 018-0517-2. 681
- Vandaele, A. C., O. Korablev, F. Daerden, S. Aoki, I. R. Thomas, F. Altieri, M. López-Valverde, 682 G. L. Villanueva, G. Liuzzi, M. D. Smith, J. T. Erwin, L. Trompet, A. A. Fedorova, F. 683 684 Montmessin, A. Trokhimovskiy, D. A. Belyaev, N. I. Ignatiev, M. Luginin, K. S. Olsen, L. Baggio, J. Alday, J.-L. Bertaux, D. Betsis, D. Bolsée, R. T. Clancy, E. Cloutis, C. 685 Depiesse, B. Funke, M. Garcia-Comas, J. -C. Gérard, M. Giuranna, F. Gonzalez-Galindo, 686 A. V. Grigoriev, Y. S. Ivanov, J. Kaminski, O. Karatekin, F. Lefèvre, S. Lewis, M. López-687 Puertas, A. Mahieux, I. Maslov, J. Mason, M. J. Mumma, L. Neary, E. Neefs, A. 688 Patrakeev, D. Patsaev, B. Ristic, S. Robert, F. Schmidt, A. Shakun, N. A. Teanby, S. 689 690 Viscardy, Y. Willame, J. Whiteway, V. Wilquet, M. J. Wolff, G. Bellucci, M. R. Patel, J.-J. López-Moreno, F. Forget, C. F. Wilson, H. Svedhem, J. L. Vago, D. Rodionov, NOMAD 691 Science Team, and ACS Science Team (2019), Martian dust storm impact on atmospheric 692 H₂O and D/H observed by ExoMars Trace Gas Orbiter, Nature, 568, 7753, 521-525, doi: 693 10.1038/s41586-019-1097-3. 694
- Wang, C., F. Forget, T. Bertrand, A. Spiga, E. Millour, and T. Navarro (2018), Parameterization
 of Rocket Dust Storms on Mars in the LMD Martian GCM: Modeling Details and
 Validation, Journal of Geophysical Research: Planets, Volume 123, Issue 4, pp. 982-1000,
 DOI: 10.1002/2017JE005255.

699 Figures

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Figure 1. Solar longitude (x-axis) and latitude (y-axis) of the solar occultation measurements taken from (a) 21 April to 30 September 2018 and (b) from 1 December 2018 to 23 February 2019 by TGO/NOMAD used in this study. The color denotes the highest altitude at which the mean transmittance of spectra at the diffraction order 168 is less than exp(-1.0) (i.e., the optical depth is less than 1.0), which basically corresponds to the top altitude (relatively) free from the dust suspended in the Mars atmosphere.



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Figure 2. Examples of spectra obtained during one occultation (21 April 2018; the first date of the solar occultation measurements by TGO/NOMAD) in the spectral range between 3775 and 3805 cm⁻¹ (diffraction order 168). Each transmittance is obtained by dividing the spectra measured through the atmosphere by the reference spectrum recorded outside the atmosphere. The selection of a spectral interval is achieved through the AOTF. The absorption features presented in the spectra are mainly H₂O lines. Differences in colors represent the tangential altitude of the measurements.

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Figure 3. Examples of the data analysis of NOMAD spectra to retrieve H₂O abundance. Spectra 731 of (a) diffraction order 134 taken at 60 km altitude (latitude=51S°, longitude=22E°) on 28 June 732 2018 (Ls=201°) and (b) diffraction order 168 taken at 97 km altitude (latitude=38°N, 733 longitude=138W°) on 24 June 2018 (Ls=199°). The black and red curves show the measured 734 NOMAD spectrum and the best-fit synthetic spectrum calculated by the radiative transfer model, 735

respectively. The retrieved H₂O abundance are 201 (\pm 4) and 233 (\pm 8) ppm, respectively. The bottom figures show the residuals of the fits in red and the error bars in black. Note that larger residuals around the H₂O line at 3026 cm⁻¹ shown in Panel (a) are due to the fact that the spectral calibration is not achieved with a sub-pixel accuracy. The H₂O line at 3801.4 cm⁻¹ shown in Panel (b) has larger error bars because the core of the line is saturated.



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Figure 4. Examples of the water vapor vertical profiles retrieved from solar occultation measurements by TGO/NOMAD taken on (a) 19 June 2018 ($Ls=196^\circ$, latitude=82°S, longitude=159E° at the highest altitude), and on (b) 28 June 2018 ($Ls=201^\circ$, latitude=15°N, longitude=14° at the highest altitude). The black and blue curves show the vertical profiles of water vapor volume mixing ratio retrieved from the spectra in the diffraction orders 168 and 134, respectively. The red curves are their weighted averages. The horizontal bars represent 1-sigma errors due to the instrumental noise.



Figure 5. Seasonal variation of the water vapor vertical profiles at $Ls=162-260^{\circ}$ retrieved from the NOMAD data in (a) the northern hemisphere and (b) the southern hemisphere, (c, d) those predicted by the GEM-Mars for non-dust storm conditions, and (e, f) the differences between the NOMAD retrievals and the GCM predictions. The retrievals and GEM predictions are binned in $1^{\circ} Ls \times 1$ km altitude grid (averaged in latitude and longitude). The top panels of (a) and (b) show the latitudes and local solar time of the measurements.



Figure 6. Latitudinal variation of the water vapor vertical profiles retrieved from NOMAD data 759 (the top panels of (a)-(e)), predicted by the GEM-Mars for non-dust storm conditions (the bottom 760 panels of (a)-(e)) in the seasonal range between $Ls = 180-195^{\circ}$ (Fig. (a), before the global dust 761 storm), $Ls = 195-202^{\circ}$ (Fig. (b), during the growth phase of the storm), $Ls = 210-220^{\circ}$ (Fig. (c), 762 during the mature phase of the storm), $Ls = 220-240^{\circ}$ (Fig. (d), during the decay phase of the 763 storm), and $Ls = 240-260^{\circ}$ (Fig. (e), during the decay phase of the storm). The retrievals and 764 GEM predictions are binned in 5° latitude \times 1 km altitude grid (averaged in season and 765 longitude). 766



Figure 7. Seasonal variation of the water vapor vertical profiles at $Ls = 298-345^{\circ}$ retrieved from the NOMAD data in (a) the northern hemisphere and (b) the southern hemisphere, (c, d) those predicted by the GEM-Mars for non-dust storm conditions, and (e, f) the differences between the NOMAD retrievals and the GCM predictions. The retrievals and GEM predictions are binned in 1° $Ls \times 1$ km altitude grid (averaged in latitude and longitude). The top panels of (a) and (b) show the latitudes and local solar time of the measurements.



Figure 8. Latitudinal variation of the water vapor vertical profiles retrieved from NOMAD data (the top panels), predicted by the GEM-Mars for non-dust storm conditions (the bottom panels) in the seasonal range between $Ls = 300-320^{\circ}$ (a), $Ls = 320-330^{\circ}$ (b, at the time of the regional dust storm), $Ls = 330-345^{\circ}$ (c). The retrievals and GEM predictions are binned in 5° latitude × 1 km altitude grid (averaged in season and longitude).

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