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Terrestrial OH nightglow measurements during the Rosetta fly-by

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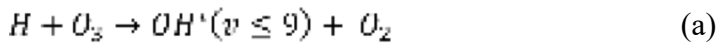
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Abstract

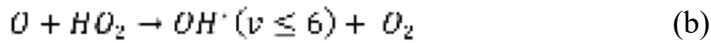
We present a study of the terrestrial hydroxyl nightglow emissions observed with the Visible and Infrared Thermal Imaging Spectrometer (VIRTIS) on board the Rosetta mission. During these observations, the OH $\Delta v=1$ and 2 sequences were measured simultaneously. This allowed investigating the relative population of the $v=1$ to 9 vibrational levels by using both sequences. In particular, the relative population of the vibrational level $v=1$ is determined for the first time from observations. The vibrational population decreases with increasing vibrational quantum number. A good agreement is found with a recent model calculation assuming multi-quantum relaxation for OH(v) quenching by O₂ and single-quantum relaxation for OH(v) by N₂.

1. Introduction

The Meinel bands of hydroxyl are a good proxy to investigate atmospheric properties for the terrestrial planets. These emissions have been extensively used to study the photochemical and dynamical properties of the atmosphere in the Earth's upper mesosphere. Vibrationally excited OH is produced through the Bates-Nicolet mechanism following the chemical reaction [Bates and Nicolet, 1950]:



which is effective in the Earth, Mars and Venus atmospheres. The reaction involving HO₂ short-lived molecules:



was found to be negligible in the excitation of the Earth's OH Meinel bands with v up to 6 [Meriwether, 1989], although its role is still debated, as discussed in Xu et al.[2012].

On Earth, the OH airglow layer is located near the mesopause [Lowe et al., 1996; She and Lowe, 1998], with a maximum at 87 km and a full width at half maximum (FWHM) of about 8 km [Baker and Stair, 1988].

Several studies based on ground- and space-based observations demonstrated that the OH Meinel emission profile is strongly sensitive to the atmospheric temperature and density profiles. Dynamic structures, like tides [Xu et al., 2010; Zhang et al., 1998; Shepherd et al., 1998; Ward, 1999; Zhang and Shepherd, 1999; Russell et al., 2005; Liu et al., 2008] or planetary waves [Snively et al., 2010; Gao et al., 2010] are found to affect the peak altitude by modulating the OH emission profile. Quenching by atomic oxygen contributes also to the vertical shift observed in the peak altitudes

53 from different Meinel bands [von Savigny and Lednyts'kyi, 2013]. Temperature inversions and
54 minor species mixing effects are also responsible for changes in the OH airglow vertical profile
55 [Melo et al., 1999; von Savigny et al., 2012]. Moreover, it has been observed that the emission
56 originating from higher vibrational levels typically occurs at higher altitudes [von Savigny et al.,
57 2012]. Ground-based spectroscopy of the OH airglow has also been used to infer the mesospheric
58 temperature at the airglow altitudes [Zhao et al., 2005; She and Lowe, 1998].

59 Here we present an analysis of the OH($\Delta v=1,2$) sequences in the Earth's atmosphere, observed
60 with the Visible and Infrared Thermal Imaging Spectrometer (VIRTIS) on board the Rosetta
61 spacecraft, during the Earth fly-by in November 2009, in order to obtain the population distribution
62 of the vibrational levels.

63 The observing geometry has limited the lowest accessible altitude to 87 km, with a vertical
64 resolution on the order of 10 km/pixel at best. Hence we limit our investigation to the altitude range
65 87-105 km, without exploring the vertical dependence of the emission. Moreover, the spectral
66 resolution is not adequate to resolve single ro-vibrational transitions, and the vibrational manifolds
67 are only partly resolved. For this reason, we assume that a Gaussian shape reproduces the emission
68 layer.

69 Our analysis will be compared with previous ground-based and space observations and with
70 model calculations. The method we employ in this analysis presents the advantage of being less
71 sensitive to spurious signals and instrumental artifacts because it uses the $\Delta v=1$ and $\Delta v=2$ spectra
72 which were simultaneously observed. In addition, spectra observed from space are not affected by
73 telluric absorptions, hence the full OH($\Delta v=1,2$) sequences of emissions can be retrieved without
74 correction for atmospheric extinction.

75 In section 2 the analysis of the VIRTIS/Rosetta spectral data is described, focusing on the
76 terrestrial OH nightglow emission in the infrared. An empirical radiative model of the spectral
77 range where OH($\Delta v=1,2$) emissions occur is described in section 3 with the aim to allow a retrieval

of the vibrational populations. In section 4, we discuss the results and compare them with analyses of experimental ground-based observations and theoretical works.

2. Data selection

Rosetta is the first large-class European mission devoted to the close investigation of small bodies in the Solar System [Schulz, 2009]. It was launched on 2nd March 2004 towards the primary target of the mission, comet 67P/Churyumov-Gerasimenko (67P/C-G). The main scientific objectives of the Rosetta mission dealt with the study of the comet 67P/C-G, with special emphasis on its nucleus and coma. Gravity assist maneuvers with the Earth and Mars [Coradini *et al.*, 2010; Migliorini *et al.*, 2013] were performed during the ten year-long cruise phase, as well as fly-bys with the main belt asteroids (2867) Steins [Tosi *et al.*, 2010; Leyrat *et al.*, 2011] and (21) Lutetia [Coradini *et al.*, 2011]. The investigation of the planetary airglow emissions is one of the goals foreseen for the mission during the Earth and Mars fly-bys.

The VIRTIS instrument [Coradini *et al.*, 2007] includes two spectral channels: (1) the VIRTIS-M mapping spectrometer, with imaging capabilities and a medium spectral resolution and (2) the VIRTIS-H echelle spectrometer, with a higher spectral resolution compared to VIRTIS-M, but without imaging capabilities. VIRTIS-M covers the 0.3–5.1 μm range in 864 spectral bands by means of two co-aligned channels: the Vis/NIR channel, operating from 0.3 to 1.0 μm with a spectral sampling of 2 nm; and the IR channel, from 1.0 to 5.1 μm with a spectral sampling of 9.5 nm; the spectral resolution is of the order of 3 nm and 20 nm in the visible and infrared, respectively. Each data cube in the visible and/or infrared has a dimension of $432 \times N_s \times N_l$, where 432 is the number of spectral channels, N_l (number of lines) depends on the length of the observation, while N_s (number of samples) is the number of spatial pixels composing a line, usually equal to 256. The field of view of each square-shaped pixel is 0.25 mrad wide, hence a 256 pixel $\times$ 256 pixel image, obtained by using a scanning mirror, covers a 64 mrad $\times$ 64 mrad field (which

104 corresponds to $220 \text{ arcmin} \times 220 \text{ arcmin}$). The full FOV is acquired in time by repeating successive
105 acquisitions while the internal steerable mirror performs a scan or maintain it at fixed position while
106 the spacecraft is drifting (pushbroom mode). The Earth data discussed here were acquired in the
107 scan mode, allowing one to acquire a sequence of limbs consecutively.

108 The two VIRTIS-M focal planes are equipped with order-sorting filters to reduce
109 contaminations due to higher spectral orders coming from the diffraction gratings. The two filters
110 placed on the visible channel's detector have a junction placed at $0.640\text{-}0.651 \text{ }\mu\text{m}$; the five on the
111 infrared channel detector have four junctions corresponding to $1.415\text{-}1.576 \text{ }\mu\text{m}$, $2.388\text{-}2.548 \text{ }\mu\text{m}$,
112 $3.671\text{-}3.765 \text{ }\mu\text{m}$, and $4.284\text{-}4.397 \text{ }\mu\text{m}$ wavelength [Coradini *et al.*, 2007]. In general the spectral
113 radiance measured through the junctions has been corrected by the calibration pipeline. However,
114 despite this correction some residual signals remain present on the first two junctions of the IR
115 channel.

116 Since VIRTIS-M spectral range extends up to $5.1 \text{ }\mu\text{m}$, thermal environment plays a major role in
117 the instrumental performance. During the fly-by, the VIRTIS radiator FOV was partially filled with
118 the Earth, resulting in an optical bench temperature of about 147 K , significantly larger than the
119 typical operative temperature of 135 K . This excess in temperature results in a wavelength shift of
120 $\sim 11 \text{ nm}$ in the spectral calibration [Migliorini *et al.*, 2013], which was corrected in the calibration
121 pipeline [Filacchione *et al.*, 2006].

122 In this paper we focus our analysis on data acquired in the IR spectral range, during the Earth
123 gravity assist that occurred in November 2009. Prior to this fly-by, two more gravity assists with
124 our planet had been performed by Rosetta on 4 March 2005 and between 13 and 14 November
125 2007. They will not be discussed here because the observations were not suitable to study
126 nightglow emissions. During the first fly-by, no limb observation was performed, while during the
127 second fly-by the limb mode observations concentrated on the dayside.

128 In the approach phase of the November 2009 Rosetta fly-by with the Earth, a sequence of full-
129 disk images of the night side of the planet was acquired by VIRTIS-M from a distance spanning the

130 range 228,000-240,000 km. These data were not suitable for the O₂ and OH nightglow investigation
131 because of the low spatial resolution, and hence are not considered in this study. When the
132 spacecraft was less than 55,000 km away from the Earth, limb scans of the night side were carried
133 out, starting from 150 km above the surface. These data satisfy our requirements to study the
134 nightglow emission; the resulting vertical resolution (13 - 15 km/pixel) is similar to the thickness of
135 the emission layer and the duration of each scan (2 min) is short enough to minimize possible time
136 variability of the emissions.

137 The VIRTIS airglow data (image name I1_00216713355) consist of a collection of two limb
138 scans in the same image, acquired a few minutes apart. The total duration of the IR channel scan is
139 12 minutes, corresponding to a sequence of 73 lines with an integration time of 7 sec each. The two
140 limbs cover the latitude region from 38° to 47° in the northern hemisphere, and are centered at
141 1:30–2:00 AM solar local time. A more detailed description of the Earth's observations with
142 VIRTIS was given in *Migliorini et al.* [2013] and *Hurley et al.* [2014].

143 In order to remove high frequency spatial noise, the cube-image was cleaned using a median
144 filter combined with a smoothing procedure, applied in the spatial direction while the temporal and
145 spectral dimensions were kept unchanged.

146 Since it was verified that the emission is roughly located at about 90 km, we averaged spectra
147 collected between 87 and 105 km in order to increase the signal-to-noise ratio. A total of 300
148 individual spectra were summed together for each limb scan. The analysis was applied to the two
149 VIRTIS observations separately, because the contribution of the background is different from one
150 scan to the other. The radiance was converted into Rayleigh units (1 R corresponds to 10⁶
151 photons/cm² s⁻¹ in 4π steradians). The resulting spectrum is shown in Figure 1 for one of the two
152 limb scans.

153 [HERE FIGURE 1]

154

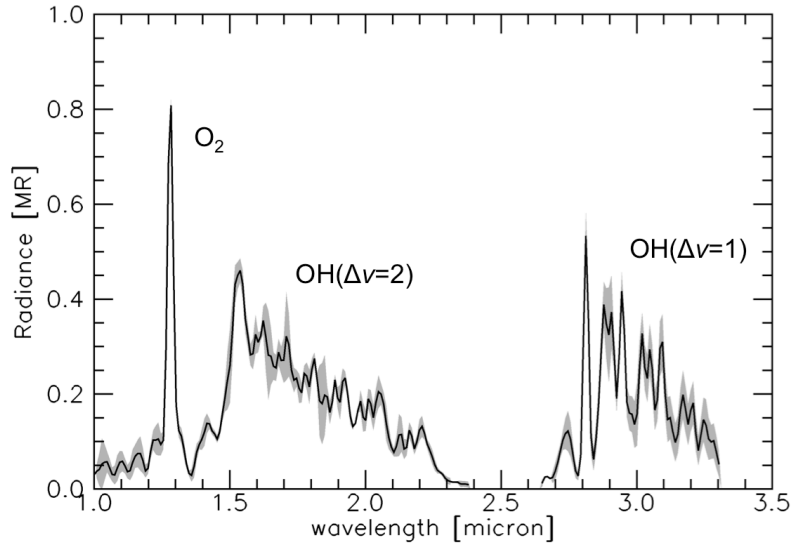


Figure1. VIRTIS mean nightglow spectral radiance, in the altitude range 87-105 km. This dataset was processed with a median filter applied to the spatial component, in order to eliminate instrumental artifacts and with a background correction obtained by subtracting the spectrum measured between 120 and 125 km. Finally a wavelength calibration adjustment has been performed by applying a shift of ~ 11 nm towards shorter wavelengths (see also Migliorini et al. [2013]). The shaded area indicates the experimental error, calculated from the Noise Equivalent Spectral Radiance (NESR). The O_2 and $OH(\Delta v=1,2)$ emissions are indicated.

In the mean spectrum, the O_2 emission centred at $1.27 \mu\text{m}$ is clearly observed, as well as the $\Delta v=2$ OH Meinel bands in the $1.4\text{-}2.4 \mu\text{m}$ region, and $\Delta v=1$ bands in the $2.7\text{-}3.3 \mu\text{m}$ region, as indicated in Figure 1. The spectral region beyond $3.5 \mu\text{m}$ is dominated by the thermal emission and will not be discussed here.

Due to the limited spectral and spatial resolution, the variation of the peak emission's altitude for the different transitions studied by several authors [Kaufmann et al., 2008; McDade, 1991; von Savigny et al., 2012] cannot be verified in VIRTIS data.

3. Spectral model

174 We use the PGOPHER code (<http://pgopher.chm.bris.ac.uk>) to generate the line intensities of
175 the rotational transitions for each vibrational band. The population distribution of the rotational
176 levels is assumed to be Maxwellian. A rotational temperature of 200 K has been used in order to
177 calculate a synthetic spectrum of the OH($\Delta v=1$ and 2) emissions. This temperature is close to the
178 average value at the mesopause [Xu *et al.*, 2012]. The variation of the rotational temperature with
179 the vibrational levels has been discussed in Cosby and Slanger [2007] and Noll *et al.* [2014]. The
180 difference between the rotational temperature in the $v=1$ manifold with respect to the $v=10$ manifold
181 may be as large as 20 K. We simulated the rotational manifold for three different temperatures (200
182 K, 250 K and 300 K) and compared the simulations, convolved to the VIRTIS spectral resolution of
183 20 nm, with the observed spectra. It appeared that a rotational temperature of 200 K produced the
184 best agreement. We note, however, that a rotational temperature difference of the order of 15-20 K
185 only affects the structure inside each band, whose effect is negligible at the VIRTIS resolution. The
186 emission spectra of the OH Meinel bands can thus be simulated by multiplying the calculated
187 rotational manifolds for all vibrational quantum numbers ($v=1-9$) with the (unknown) populations
188 of the upper levels and the relative Einstein coefficients for each vibrational transition. The Einstein
189 coefficients were taken from Xu *et al.* [2012], calculated for a temperature of 200 K. The variation
190 of the Einstein coefficients with temperature is very small (see e.g. Xu *et al.* [2012]) and thus our
191 simulation is largely independent from the rotational temperature assumed. We then performed a
192 least squares fit, which yielded the relative populations of the vibrational levels ($v=1$ to 9), taking
193 into account both $\Delta v=1$ and $\Delta v=2$ bands. Figure 2 a-b shows the comparison between the VIRTIS
194 spectra and the corresponding best fit for the available data.

195 [HERE FIGURE 2]

196

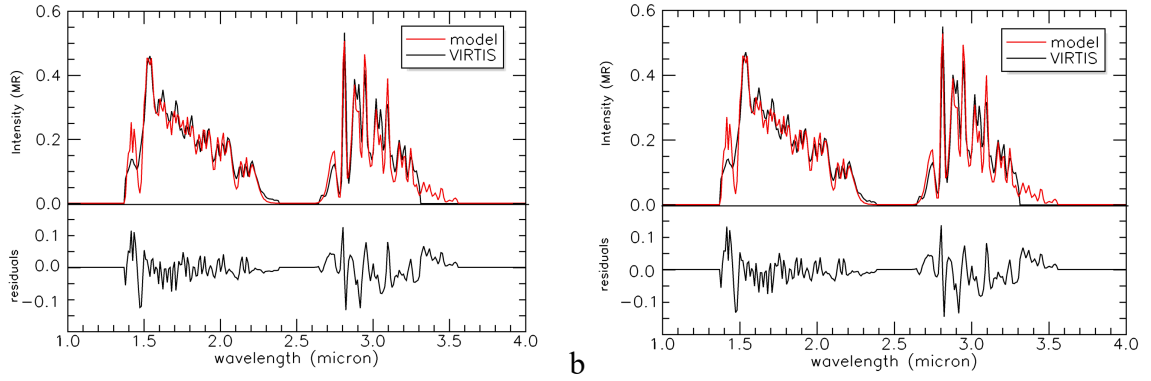


Figure 2. *a - Comparison between the VIRTIS spectrum (in black) and the spectral model (in red) for the first limb scan. The radiance outside the wavelengths regions covered by the OH emissions is set to 0, to limit the fit to the spectral region involving OH emissions. In the bottom panel of the figure, the residuals are shown. b- the same for the second limb scan.*

Similarly, a fit is obtained for the second VIRTIS spectrum, acquired within a few minutes from the first one. Both fits are in good agreement and provide consistent results for the populations of levels from $v=1$ to 9, except for the region between 1.4 and 1.5 μm , where a VIRTIS order-sorting filter junction is located. The model overestimates the intensity of the (2-0) transition at 1.46 μm in both spectra. The same discrepancy was observed by comparing the TIMED/SABER observations with models, as reported by *Xu et al.* [2012]. The TIMED/SABER observations had been corrected for the atmospheric attenuation, mostly due by water vapor. The good agreement between fit and VIRTIS observations of the (2-1) transition (around 2.82 μm) seems to exclude a specific quenching of the $v=2$ level. A possible explanation may be an error in the Einstein coefficient.

The average values of each population obtained for the two fits are provided in Table 1. To calculate the error in the vibrational populations, a set of 40 spectra was statistically generated by adding a random error to the original spectra. Each spectrum so obtained was fitted in the same way as the VIRTIS spectra, and the standard deviation of the results was taken as errors in the

determination of the vibrational population. Values of the percentage into each v level for the two spectra, together with the calculated statistical error, are shown in Table 1.

Table 1. *The fitted OH vibrational population from the two VIRTIS spectra. The population values are given in terms of percentage, while the errors are absolute.*

Level	Population – 1 st fit	Error	Population – 2 nd fit	Error
1	37.8	1.0	37.58	1.03
2	21.02	1.15	20.33	1.19
3	17.95	0.8	18.36	0.83
4	7.27	0.46	7.40	0.48
5	4.88	0.30	4.92	0.31
6	3.59	0.26	3.66	0.27
7	2.91	0.21	2.99	0.22
8	2.77	0.20	2.84	0.20
9	1.83	0.24	1.91	0.25

4. Discussion

The terrestrial nightglow emissions at the mid-latitudes of the northern hemisphere have been investigated by using VIRTIS observations during a Rosetta Earth fly-by. During the season (November) of these observations, the OH nightglow is quite intense at the altitudes, local time and latitudes observed by VIRTIS, as reported also by the Wind Imaging Interferometer (WINDII) [Liu *et al.*, 2008].

The vibrationally excited hydroxyl radical can decay through spontaneous emission of a photon, giving rise to the observed emission, or can be quenched through collisions with other ambient molecules (N₂, O₂) or atoms (O). The quenching process can occur stepwise involving transitions from a specific vibrational level to a lower vibrational level, or all at once to the vibrational ground level. This so-called sudden death mechanism, which can occur by vibrational quenching or by chemical reaction, results in a smaller population of the low vibrational levels. While the radiative

236 processes depend merely on the Einstein coefficients, which are rather independent of temperature,
237 the quenching processes may depend on the reaction rates, which may have strong temperature
238 dependence and on the availability of quenchers. This implies that the vibrational population of the
239 OH radical also may depends on the time and latitude of observation.

240 We compare our results with ground based measurements reported in *Krassovsky et al.* [1962],
241 *Ferguson and Parkinson* [1963], *Harrison and Kendall* [1973], *Turnbull and Lowe* [1983], *Oliva*
242 *and Origlia* [1992], and with model calculations by *Llewellyn et al.* [1978], *von Clarmann et al*
243 [2010] and *Xu et al* [2012].

244 The heterogeneity of the previous measurements limits the comparison to few levels, which are
245 present in all the considered datasets. For this reason, the comparison is made by normalizing all
246 relative populations to that of the $v=4$ level, which has been reported by all authors.

247 *Oliva and Origlia* [1992] provide a list of high resolved rotational lines of hydroxyl, from which
248 we derived the vibrational populations, by summing the identified lines for each level. As stated
249 also by the authors, the line list is sometimes incomplete or limited by experimental issues, such as
250 the atmospheric attenuation, so that the calculated populations have to be considered as lower
251 limits.

252 The model proposed by *von Clarmann et al.* [2010] produces excited OH in all vibrational levels
253 ($v=1-10$), by the hydrogen-ozone reaction. They used the non-LTE GRANADA model [*Funke et*
254 *al.*, 2012] to calculate the relative OH populations for vibrational levels 0 to 9, for six different
255 atmospheric conditions, assuming a stepwise quenching by O₂, N₂ and O. The model case
256 describing a ‘mid-latitudes night’ atmosphere is the only one close to the observing conditions of
257 the VIRTIS data discussed here. Hence, in Table 2 we report von Clarmann’s results only for this
258 model conditions. Note that the rate coefficients for OH production and for the quenching processes
259 were much different from those used by *Llewellyn et al.* [1978].

260 *Xu et al.* [2012] compare the TIMED/SABER observations at 1.6 and 2.0 μm with a nightglow
261 emission model, which cover 4 OH transitions out of the $\Delta v=2$. They concluded that reaction (a) is
262 the dominant source for the OH nightglow emission for the vibrational levels with $v \geq 4$. The
263 assumption of multi-quantum relaxation by O_2 and single-quantum relaxation by N_2 produced the
264 best agreement with the SABER data, while the sudden death model did not reproduce the
265 observations.

266 Except for our high $v=3$ population, all the relative populations from *Krassovsky et al.* [1962],
267 *Harrison and Kendall* [1973] and *Ferguson and Parkinson* [1963] are in agreement with those
268 derived here. We examined the possibility that the order-sorting filter at 1.415-1.576 μm could
269 cause some residual instrumental effect (see e.g. *Tosi et al.*, 2012), but repeating the fits without this
270 spectral interval did not produce appreciable changes in the results. The studies of *Krassovsky et al.*
271 [1962] and *Harrison and Kendall* [1973] were based on observations of the $\Delta v=3, 4, 5, 6$ and the
272 $\Delta v=2, 3, 4, 5$ sequences, but no information about the $v=1$ level could be obtained. Our analysis
273 yields a population for the $v=1$ level which is about 5 times higher than the $v=4$ level. None of the
274 previous analyses of experimental observations allowed the determination of the population of the
275 $v=1$ level, while only two of them report the population of $v=2$. Of these two, the value of *Turnbull*
276 *and Lowe* [1983] is very similar to ours, while those reported by *Oliva and Origlia* [1992] seem
277 unrealistic. The population of $v=1$ level has been calculated by using different model simulations.
278 *Llewellyn et al.* [1978] report a value 4.16 for the $v=1$ population with respect to the $v=4$ population,
279 while *Xu et al.* [2012] report a value of 4.47, which is in reasonable agreement with the one
280 obtained from the VIRTIS data. The value by *von Clarmann et al.* [2010] is significantly lower
281 (3.27). The populations of levels 1 to 3 are underestimated in *von Clarmann et al.* [2010], while
282 those with $v > 4$ are higher than the populations derived from the VIRTIS/Rosetta data. These
283 discrepancies and the difference with the populations calculated by *Xu et al.* [2012] possibly stem
284 from the use of a single quantum relaxation in collisions with O by *von Clarmann et al.*, whereas

285 other models assume sudden-death chemical loss, where the excited OH molecule is directly
286 deactivated to the $v=0$ ground level. Another difference lies in the treatment of collisions with O₂:
287 *von Clarmann et al.* assume single quantum relaxation, while *Xu et al.* adopt a multi-quantum
288 scheme.

289 *Bunn and Gush* [1972] also measured the $\Delta v=1, 2$ sequences using balloon-borne instruments.
290 They found relative populations to be $P(v=1)/P(v=2)=2.26$ and $P(v=2)/P(v=3)=1.76$. Our study
291 suggests ratios of 1.82(16) and 1.14(12), respectively. The populations calculated from the
292 intensities of sky emissions taken from *Oliva and Origlia* [1992] do not follow the trend of
293 decreasing populations with increasing levels. It appears that their $v=2$ and $v=7$ populations are
294 largely underestimated. In fact, these authors mention that some of the line intensities could be
295 underestimated by large factors as they lie in regions of low atmospheric transmission.

296 Observations with the same VIRTIS instrument on the European Venus Express mission
297 allowed discovering the OH infrared night airglow on the Venus nightside in 2007 [*Piccioni et al.*,
298 2008]. The OH airglow spectra on the two planets show some differences in the intensity
299 distribution of the different bands. For example, the relative intensities of the $\Delta v=1$ bands
300 originating from $v' > 2$ levels are higher in the terrestrial spectrum than in the Venus case. This
301 difference is presumably linked to the much larger abundance of CO₂ in the Venus atmosphere. On
302 Venus (and presumably on Mars), CO₂ is the dominant quencher, while O₂ and O play the major
303 role on Earth to deactivate vibrationally excited OH. *Soret et al.* [2012] demonstrated that for
304 Venus, compared with these terrestrial results, the paradigm of single vibrational quantum collision
305 deactivation by CO₂ provides a much better agreement with both the spectral structure and the
306 observed total brightness than the “sudden death” model.

307

Table 2: Comparison of relative vibrational populations with previous studies. Values are normalized at $v=4$, for which all the considered works provide values.

	This work	Krassovsky et al. [1962] ^a	Ferguson and Parkinson [1963] ^a	Harrison and Kendall [1973] ^a	Turnbull and Lowe [1983] ^a	Oliva and Origlia [1992] ^a	Llewellyn et al. [1978] ^b	vonClarmann et al. [2010] ^b	Xu et al. [2012] ^b
	38-47°N	55-62°N	55-62°N	56°N	43°N	29°S	45°N	Mid Lats	30-50°N
Level	Nov 2009	1957-1958	1957-1958	Dec 1971	Apr 1980	Apr 1991			
1	5.1±0.5						4.16	3.27	4.47
2	2.8±0.36				2.83	0.47	2.24	1.85	2.41
3	2.47±0.29	1.73	1.96	1.27	1.76	1.44	1.43	1.31	1.50
4	1.00±0.14	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00
5	0.67±0.09	0.67	0.80	0.57		0.77	0.78	0.77	0.68
6	0.49±0.07	0.40	0.47	0.44	0.25	0.26	0.60	0.59	0.47
7	0.40±0.06	0.29	0.38	0.27	0.17	0.01	0.52	0.49	0.35
8	0.38±0.05	0.23	0.24	0.29	0.16	0.26	0.52	0.33	0.30
9	0.25±0.05	0.18	0.22	0.17	0.16	0.22	0.51	0.19	0.22

^aGround-based observations

^bNumerical model

5. Conclusions

For the first time, the data analyzed in this study allow investigating the OH infrared nightglow emission from space and the vibrational population down to $v=1$. The $\Delta v=1$ and $\Delta v=2$ sequences have been observed simultaneously. We remind, however, that VIRTIS spatial and spectral resolution does not allow a complete retrieval of the vertical profile of each single transition, and hence our measurements are limited. No correction for the telluric absorption is needed and the populations deduced for the 1 to 9 vibrational levels might thus be considered as quite reliable.

The results of our analysis have been compared with ground-based observations and theoretical models. A rather good agreement is found with the model proposed by Xu et al. [2012], assuming multi-quantum relaxation by O₂ and single-quantum relaxation by N₂. The slightly smaller

323 populations for the levels $v=1-3$ in *Xu et al.* [2012] can possibly be explained by the omission of
324 reaction (b) in their calculation.

325

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331 previous works.

332

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