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1 **Minor species in Venus' night side troposphere as**
2 **observed by VIRTIS-H/Venus Express**

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14 **Key Points:**

- 15 • The latitudinal variability of carbon monoxide on Venus is confirmed, as well as
16 its anti-correlation with carbonyl sulfide.
- 17 • Zonal variations of carbon monoxide near the equator may be correlated with zon-
18 ally shifted surface elevation.
- 19 • No variations of water vapor and sulfur dioxide were found; a spectral resolving
20 power nearing 10000 is needed to further investigate.

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Abstract

21 The 2.3 μm spectral window has been used to constrain the composition of the lower at-
22 mosphere (in the 30-40 km altitude range) on the night side of Venus for more than thirty
23 years. Here, we present a follow-up study of Marcq et al. (2008), but using the full VIRTIS-
24 H/Venus Express data archive as well as an updated radiative transfer forward model.
25 We are able to confirm a latitudinal increase of CO of about 30% between 0° and 60°N ,
26 as well as an anti-correlated vertical shift of OCS profile by about -1 km in the same
27 latitude range. Both variations are about twice smaller in the southern hemisphere. Cor-
28 relations of low latitude CO and OCS variations with zonally shifted surface elevation
29 is tentatively found. These results are consistent with CO and OCS variations result-
30 ing from the competition between local thermochemistry and a Hadley-cell-like general
31 circulation, albeit influenced by the orography. Finally, no evidence for spatial variations
32 of water vapor (combined H_2O and HDO) or sulfur dioxide could be evidenced in this
33 data set; better constraining possible variations of these species would require future mis-
34 sions to include infrared spectrometers operating at a spectral resolving power higher
35 than $\sim 10^4$, such as VenSpec-H onboard EnVision.
36

Plain Language Summary

37 Remotely measuring the composition of the Venusian atmosphere below the clouds
38 is challenging, yet yields invaluable insights about the atmospheric chemistry, circula-
39 tion and interaction with the surface and interior of the planet. The VIRTIS-H instru-
40 ment on board ESA's *Venus Express* orbiter (2006-2014) provides a rich data set in this
41 regard, thanks to its ability to observe and analyse, on the night side of the planet, the
42 infrared radiation emitted by the deep atmospheric layers. The results of our analyses
43 confirm the previously observed trends for the variations of two trace gases (carbon monox-
44 ide and carbonyl sulfide) with latitude, explained by the combined effects of chemical
45 reactions and transport by the atmospheric circulation. Variations of carbon monoxide
46 may also be linked to the variations of ground elevation, confirming the link between sur-
47 face topography and atmospheric circulation. However, we were unable to separate the
48 signature of heavy water vapor from ordinary water vapor or to detect any variations
49 in sulfur dioxide, both of which require more powerful infrared instruments such as those
50 planned on future Venus orbiters such as ESA's EnVision.
51

1 Introduction

Since its serendipitous discovery by Allen and Crawford (1984), the 2.3 μm night side emission of Venus has proven an invaluable tool to investigate the tropospheric chemistry and the lower cloud deck. Its thermal origin from atmospheric layers located between 30 and 40 km in altitude, below the clouds, was already suspected by Allen (1987), and its first radiative transfer modelings (Crisp et al., 1989; Bézard et al., 1990) have shown that numerous trace gases could be measured in this region: H_2O , HDO , HF , SO_2 , OCS and CO . For CO and OCS , the variation of their peak contribution functions within the spectral window was extended enough so that not only their mean abundance, but also their vertical gradient could be retrieved (Pollack et al., 1993).

Ground based observations at a high spectral resolution could even constrain the global average value of sulfur dioxide in the 30-40 km altitude range (Bézard et al., 1993; Marcq et al., 2021) as well as of the isotopic ratio of deuterium in water vapor (de Bergh et al., 1991, 1995), and found a value about 100 times higher than on Earth, with strong implication on the past history of water (Grinspoon, 1993). Conversely, early space borne observations made use of relatively low spectral resolution instruments such as NIMS/Galileo (Carlson et al., 1991; Collard et al., 1993) during its Venus fly-by. Such observations could only constrain the broadest spectral signature of CO (2-0 rovibrational band), but at a spatial resolution unachievable from Earth. In consequence, they were able to detect the latitudinal increase of CO from the lower to the higher latitudes, a feature that was confirmed by VIRTIS-M (Tsang et al., 2008, 2009; Haus et al., 2015; Tsang & McGouldrick, 2017) at a similar spectral resolution in the same spectral window, but on board the orbiter *Venus Express* (Drossart et al., 2007; Titov et al., 2006). They were thus able to map tropospheric CO abundance over the whole night side, with some hints of zonal variations, and provided a physical interpretation of these variations based on the competition between large scale Hadley cell-like vertical circulation and the existing vertical gradient of CO required by thermochemical equilibrium. In between, moderate ($500 < \lambda/\Delta\lambda < 5000$) spectral resolution observations were performed from both Earth-based telescopes (Marcq et al., 2005, 2006; Cotton et al., 2012; Arney et al., 2014) and from VIRTIS-H on board *Venus Express* (Marcq et al., 2008). These observations give access to the spatial variability of more trace species (CO , but also OCS and H_2O ; even SO_2 to a limited extent) than low ($\lambda/\Delta\lambda < 500$) spectral resolution observations, at a cost of a lower spatial resolution and/or coverage. They confirmed the spatial variability of

85 CO on planetary scales, and also found anti-correlated variations of OCS, but no evi-
 86 dence for any variability for H₂O or SO₂.

87 Here we present an update to the first VIRTIS-H 2.3 μm study performed by Marcq
 88 et al. (2008), with two major differences: (1) whereas Marcq et al. (2008) used a selec-
 89 tion data from the first 298 orbits of *Venus Express*, we use the whole extent of VIRTIS-
 90 H data acquired during the science mission as detailed in Sec. 2; and (2) the forward ra-
 91 diative transfer model and retrieval algorithms have undergone significant updates as de-
 92 tailed in Sec. 3. Measurements of tropospheric minor species (CO, OCS, water vapor,
 93 SO₂) and their variability according to latitude, longitude and/or local solar time are
 94 presented in Sec. 4, and discussed in the context of coupled chemistry and dynamics in
 95 Sec. 5. Implications for the design of future instrumentation can be found in the last Sec. 6.

96 **2 Observational data set**

97 **2.1 Global coverage**

98 In order to extend the study from Marcq et al. (2008), we downloaded from ESA’s
 99 Planetary Science Archive (PSA) the whole VIRTIS-H night side data set calibrated in
 100 physical units along the provided observational geometry information. We selected all
 101 files whose local solar time range intersects the 7pm-5am interval, leaving an hour of lo-
 102 cal solar time margin in order to avoid cross-terminator scattering of sunlight by the up-
 103 per clouds and hazes – the spectral radiance of day side Venus near 2.3 μm being about
 104 three orders of magnitude larger than the spectral radiance of the night side thermal emis-
 105 sion (Titov et al., 2018).

106 Successful observations were defined as yielding a relative accuracy better than 10%
 107 for carbon monoxide, which is the easiest species to retrieve; a criterion solely based on
 108 signal-to-noise ratio would fail to reject some observations of poor quality due remain-
 109 ing uncorrected biases (e.g. faulty straylight correction). They are shown in Fig. 1. Sev-
 110 eral remarks and comments can be inferred. First of all, latitudinal coverage is very good
 111 between ±60° in latitude, but is very sparse beyond these values. This is caused by the
 112 thicker lower clouds at higher latitudes (in both north and south pole collars and vor-
 113 tices) which drastically decreases the signal-to-noise ratio and therefore does not allow
 114 for fitting and retrieving minor species.

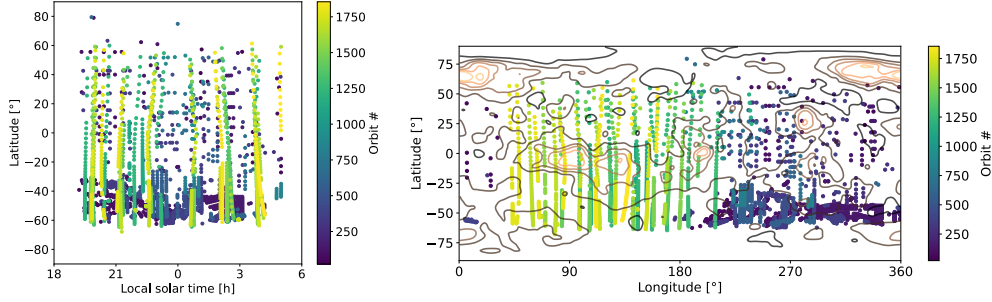


Figure 1. Location of the 3143 successful observations in local solar time/latitude (left) and longitude/latitude (right) coordinates. Color codes for the orbit number. Brown contours in the rightmost map stands for topographic elevation at -1 , 0 , 1 , 2 , 3 and 4 km following the Magellan-based digital elevation model used by Lebonnois et al. (2010).

115 Secondly, both local solar times and planetocentric longitudes are well sampled in
 116 our data set, albeit with a major difference: there is no correlation between the obser-
 117 vation date and their local solar time, whereas there is a strong correlation between the
 118 observation date and their longitude. The latter is caused by the slowly precessing quasi-
 119 polar orbit of *Venus Express* combined with the relatively short lifetime of VIRTIS-H
 120 (operating from 2006-05-14, first available orbit # 23, until 2011-06-21, last available or-
 121 bit # 1888) which prevented re-visit of previously observed longitudes. This should be
 122 kept in mind when discussing our results, since any trend seen with respect to observa-
 123 tion date could also be ascribed to longitudinal variations (and conversely). Such an is-
 124 ssue was already pointed out in the analysis of other data sets from other *Venus Express*
 125 instruments such as VMC or SPICAV (Khatuntsev et al., 2013; Bertaux et al., 2016; Marcq
 126 et al., 2020).

127 Finally, it can also be noted that we have more sampled points in the southern hemi-
 128 sphere than in the northern hemisphere, due to the high northern latitude of the peri-
 129 centre of *Venus Express*, yet the extensive coverage of this study constitutes a real im-
 130 provement over our previous study (Marcq et al., 2008).

131 2.2 Data processing

132 VIRTIS-H was the infrared high spectral resolution channel of the VIRTIS instru-
 133 ment (Coradini et al., 1998). Its optical design consisted in an echelle grating spectrom-
 134 eter, with 7 diffraction orders covering the $2\text{-}5\ \mu\text{m}$ range focused on the detector plane.

135 The spectral resolving power varied from 1500 to 2500, according to the diffraction or-
 136 der and the 432 discrete channels of each order, as shown by Marcq et al. (2008) (their
 137 Fig. 1). The spectral range of 3 among these diffraction orders intersected the 2.3 μm
 138 spectral window and are used hereafter: orders 5 (2.19-2.73 μm), 6 (2.00-2.50 μm) and
 139 7 (1.85-2.31 μm).

140 The first step of processing for these 3 diffraction orders is to correct for an even-
 141 odd effect by removing the spurious oscillating component through simple 2-pixel wide
 142 window averaging. Then, second degree polynomial adjustment is applied to the pro-
 143 vided wavelength assignment function (pixel number to physical wavelength), based on
 144 the distinctive spectral features seen in observations as well as in the forward model (Sec. 3.1).

145 **2.2.1 Zero-level adjustment**

146 As shown in (Marcq et al., 2008), a correction must be applied in order to set the
 147 proper zero-level for each relevant diffraction order (most likely caused by imperfect stray-
 148 light correction). We can take advantage here of the fact that orders 5 and 7 both in-
 149 clude part of the 2.3 μm window and part of the spectral interval where CO_2 continuum
 150 opacity leads to a negligible thermal emission. Thus, the average of order 5 spectra for
 151 wavelengths larger than 2.65 μm provides a good estimation of the true zero level for or-
 152 der 5, and so does the average of order 7 spectra for wavelengths ranging between 2.12
 153 and 2.17 μm (the imperfect straylight correction at wavelengths shortwards of 2.12 μm
 154 is not good enough for their inclusion). Order 6 does encompass the whole 2.3 μm win-
 155 dow, but does not include significant contributions outside of these windows. Therefore,
 156 we have to use here a linear interpolation (with respect to wavelength) between the short-
 157 end zero level found with order 7 and the long-end zero-level found with order 5. Also,
 158 since order 7 only includes the CO_2 -dominated part of the 2.3 μm window, it is only used
 159 for deriving the zero level for order 6 as stated above. We refer the reader to the Fig. 2
 160 in Marcq et al. (2008) for a graphical illustration.

161 Finally, we perform a binning between 30 consecutive spectra, yielding a typical
 162 footprint of several hundreds of kilometers, as a trade-off between the signal-to-noise ra-
 163 tio value, and accuracy of the observed geographical location (mainly latitude due to the
 164 aforementioned polar orbit of *Venus Express*). The minimal binned Signal-to-Noise ra-
 165 tio (SNR) required for a valid spectral inversion (previously defined as the successful fit-

166 ting of carbon monoxide abundance with a relative accuracy better than 10%, see Sec. 3.3
 167 for details about the spectral inversion) ranges from 20 to 50, which corresponds to a
 168 SNR between 4 and 10 for an individual unbinned spectrum.

169 3 Methods

170 3.1 Radiative transfer model

171 The line-by-line forward radiative transfer model we use is the same as used by Marcq
 172 et al. (2021), which in turn is an update of the model used in our first analysis of VIRTIS-
 173 H spectra (Marcq et al., 2008) and described in previous work (Marcq et al., 2005, 2006).
 174 More precisely, our previous CO₂ line database from HITEMP has been updated accord-
 175 ing to the ab initio list from Huang et al. (2014) – keeping the same sub-Lorentzian parametriza-
 176 tion as in Pollack et al. (1993)) – and our former two stream custom radiative transfer
 177 equation solver has been replaced by DISORT (Stamnes et al., 1988) in the eight-stream
 178 approximation. This allows us to include the effect of emission angle on the thermal spec-
 179 tra (i.e. limb darkening) at a reasonable computation cost – tests using more than 8 streams
 180 did not yield significant improvements in our computations, considering the simple an-
 181 alytic scattering phase functions that we use and the near-nadir observation geometry
 182 in our data set, with emission angles never exceeding 30°.

183 The atmosphere is divided into 90, 1-km thick plane-parallel layers from the sur-
 184 face up to 90 km. The temperature vs. pressure profile is kept constant according to the
 185 standard VIRA profile (Seiff, 1983), since this profile does not vary significantly below
 186 the clouds (in the 30-40 km altitude range most contributing to the thermal emission)
 187 within the investigated latitude and/or local solar time ranges (see Sec. 2.1). Clouds are
 188 modeled according to Crisp (1986), using a four modes size distribution (1,2,2',3) as de-
 189 scribed in Crisp et al. (1989) and Henyey-Greenstein scattering phase functions. Line
 190 opacities from CO₂, CO, H₂O, HDO, OCS, SO₂ and HF are extracted from (Jacquinet-
 191 Husson et al., 2011) except for CO₂ (Huang et al., 2014) as previously stated. Finally,
 192 we also have updated the (constant over the whole 2.3 μm window) value of the CO₂ con-
 193 tinuum opacity, from our previous value of 3.5 to $2.7 \cdot 10^{-8} \text{ cm}^{-1} \cdot \text{amagat}^{-2}$, which al-
 194 lowed for a better fit of the CO₂-dominated spectral range from 2.2 to 2.3 μm.

195 **3.2 Fitted parameters**

196 **3.2.1 Cloud opacity**

197 In order to account for brightness variations that affect the whole 2.3 μm spectral
 198 window (see Fig. 3), we chose to fit the lower cloud opacity through a multiplicative fac-
 199 tor applied to the particles' number densities in the 48-57 km altitude range. This is con-
 200 sistent with the observed morphology of the night side thermal emission contrasts (Carlson
 201 et al., 1991; Peralta et al., 2019) coming from lower clouds opacity variations. These night
 202 side IR contrasts differ noticeably from the contrasts observed on the day side in the UV
 203 range (Markiewicz et al., 2007; Yamazaki et al., 2018), and originating from the upper
 204 clouds.

205 Arney et al. (2014) accounted for the cloud opacity variations in the same way, and
 206 also fitted the sulfuric acid concentration in the droplets. This was made possible by the
 207 fact they analyzed simultaneous and colocated 1.74 μm and 2.3 μm observations, there-
 208 fore constraining a much larger wavelength range compared to the VIRTIS-H observa-
 209 tions which can only encompass the 2.3 μm window alone. Therefore, we assumed a con-
 210 stant value for H_2SO_4 concentration in the droplets at 75% (Hansen & Hovenier, 1974).

211 **3.2.2 Minor gaseous species**

212 Our radiative transfer code implements vertical profiles for gaseous species by spec-
 213 ifying a limited number of control points defined in terms of pressure P and volume mix-
 214 ing ratios q , and interpolating linearly $\log q$ with respect to $\log P$ between these control
 215 points, similarly to Pollack et al. (1993). The vertical location of these control points is
 216 unchanged compared to our previous studies (Marcq et al., 2005, 2006, 2008), and is read-
 217 ily visible in Fig. 2.

218 Carbon monoxide (CO), water vapor (H_2O and HDO) and sulfur dioxide (SO_2) ver-
 219 tical profiles are assumed to vary using a simple multiplicative constant applied to their
 220 volume mixing ratios for each control point. Hydrogen fluoride (HF) is included, but not
 221 fitted due to the narrowness of its spectral lines, which are unresolved at VIRTIS-H spec-
 222 tral resolution.

223 **3.2.2.1 Carbonyl sulfide** On the other hand, carbonyl sulfide (OCS) exhibits a
 224 strong vertical gradient in the 30-40 km altitude range where its contribution function

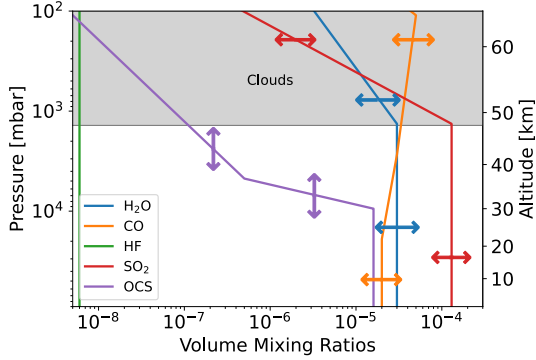


Figure 2. Nominal vertical profiles of gaseous minor species used by the forward radiative transfer model. The arrows show how these profiles are varied to fit the observed VIRTIS-H spectra.

225 peaks, so that we should in principle fit both its average value and vertical gradient (Pollack
 226 et al., 1993). Our attempts to improve their determination of the vertical gradient by
 227 fitting the variations of both OCS vertical gradient and VMR simultaneously were un-
 228 successful (degeneracy issues with OCS mixing ratio), probably due to the insufficient
 229 spectral resolution of both orders 5 and 6.

230 We could have adopted the same mixing ratio multiplication as for other minor species,
 231 as done by Arney et al. (2014), but it yielded comparatively poorer fits. It appears in-
 232 deed that we had a better fit by vertically translating the OCS reference profile (con-
 233 sidering we had to restrict ourselves to a single fitted parameter). More specifically, we
 234 vertically move the control points by applying a constant multiplicative factor to the pres-
 235 sure of the control points rather than to the mixing ratios as we did for other minor species
 236 – multiplying the pressure by $(1 + \alpha)$ is locally equivalent to a vertical translation by
 237 $-\alpha H$ where H is the local atmospheric scale height as defined by the dimensionless hy-
 238 drostatic equilibrium equation: $dP/P = -dz/H$.

239 Such a parametrization is physically sensible, since previous studies (Tsang et al.,
 240 2008; Marcq & Lebonnois, 2013) have interpreted tropospheric variations of minor species
 241 (including OCS) as resulting from a competition between local chemical processes and
 242 updrafts/downdrafts associated with the meridional-vertical circulation. Alternatively,
 243 we could adopt the same parametrization for other minor species but this was deemed
 244 too complex for those other species since their relative vertical gradient – defined as $\Delta(\log q)/\Delta(\log P)$
 245 – is not as large as that of OCS.

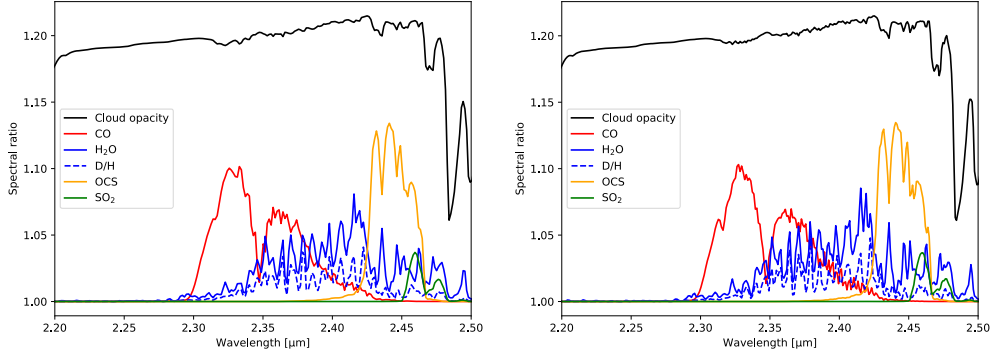


Figure 3. Synthetic spectral ratios convolved by instrumental PSF for orders 5 (left) and 6 (right) corresponding to a 10% relative difference in the atmospheric parameters described in Sec. 3.2. The small difference between both orders is due to their different PSF (see Fig. 4).

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3.3 Fitting algorithm

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Figure 3 shows the various spectral ranges affected by altering the vertical profiles of the aforementioned minor species, dividing synthetic spectra with a 10% variation in the above described atmospheric parameters (i.e. lower cloud opacity, multiplicative factor for gaseous species profiles) by our reference nominal synthetic spectrum. Our fitting strategy takes advantage of these different spectral ranges sequentially as follows:

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1. Pre-computation of look-up tables of high-resolution spectra (0.5 cm^{-1}) encompassing the variation range found in observations: {cloud opacity, CO} ; {cloud opacity, CO, H₂O, HDO} ; {cloud opacity, CO, OCS, SO₂}.
2. Convolution according to the instrumental point spread function (PSF) for orders 5 and 6. A simple Gaussian convolution function would not yield satisfactory results, so we had to resort to custom, non-analytical PSF derived from ground based instrumental characterizations (Fig. 4).
3. Fit of lower cloud opacity **and** CO mixing ratio through multilinear interpolation over the convolved look-up table. The used spectral interval can be found in Table 1.
4. Fit of H₂O **or** HDO mixing ratio through multilinear interpolation over the convolved look-up table. Cloud opacity and CO mixing ratios values found previously are kept the same. The used spectral interval can be found in Table 1.
5. Fit of OCS shift altitude and SO₂ mixing ratio through multilinear interpolation over the convolved look-up table. The cloud opacity and CO values found previ-

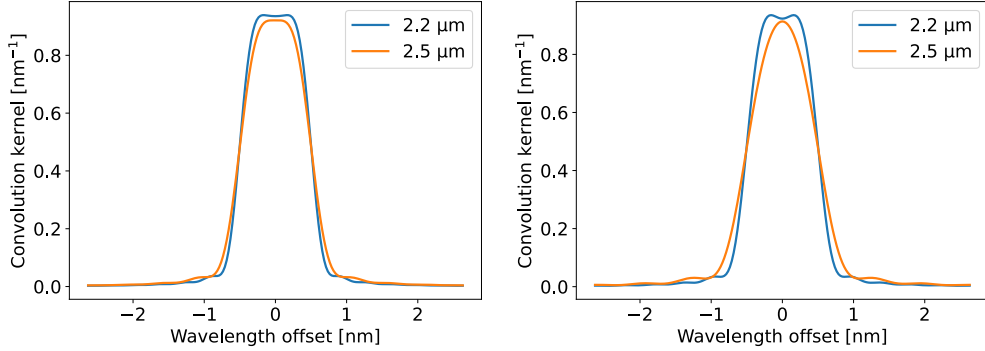


Figure 4. Normalized PSF shapes for order 5 (left) and 6 (right) for extremal wavelengths within the 2.3 μm window.

Fitted parameters	Wavelength ranges [μm]	Order(s) used
Cloud opacity, CO	2.25 – 2.28; 2.3 – 2.365; 2.4 – 2.41; 2.48 – 2.50	6
H ₂ O, HDO	2.345 – 2.415	5, 6
OCS, SO ₂	2.38 – 2.52	5, 6

Table 1. Wavelength ranges and VIRTIS-H diffraction orders used by the fitting algorithm.

267 ously is kept the same (since no variations of water vapor are found compared to
 268 the nominal profile, there is no need to take its variations into account). The used
 269 spectral interval can be found in Table 1.

270 The values given in Table 1 deserve some comments. First, fitting the cloud opac-
 271 ity requires a large wavelength coverage since it affects the whole spectral window, hence
 272 the large dispersion of used wavelengths intervals used. Second, for CO and cloud opac-
 273 ity, the need for wavelengths shorter than 2.3 μm prevented us from using order 5 (which
 274 cannot be used for quantitative retrievals at wavelengths shorter than 2.3 μm), so that
 275 we used only order 6 to retrieve them. The use of a single order does not hamper the
 276 accuracy of these fits, since CO absorption rovibrational band and cloud opacity are the
 277 two most salient features of the 2.3 μm windows and can be fitted even at a very low spec-
 278 tral resolution such as VIRTIS-M (Tsang et al., 2008; Tsang & McGouldrick, 2017).

279 As for the two remaining fitting steps, both orders 5 and 6 were considered simul-
 280 taneously when computing the χ^2 cost function to be minimized. If no single value of
 281 the parameters could fit the spectra obtained with both orders simultaneously, it led to

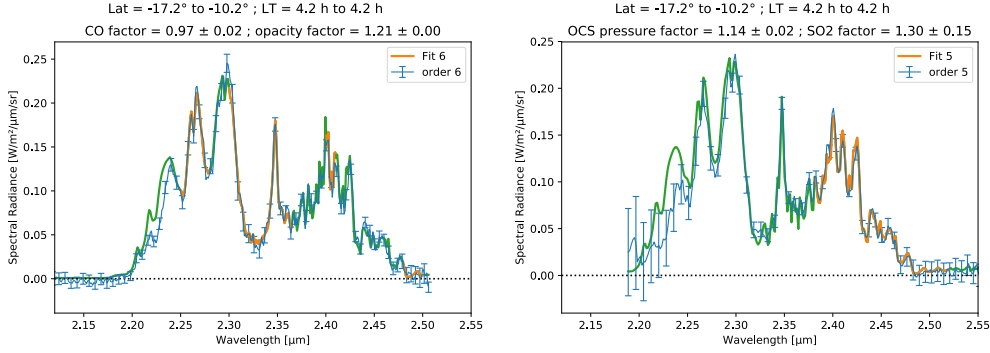


Figure 5. Best fits obtained during orbit # 277 for CO & cloud opacity with order 6 (left), and OCS & SO₂ with order 5 (right). Fitted intervals from Table 1 are shown in orange for the best fits, non-fitted intervals are shown in green for the best fits, and observed binned spectra along their 1 σ error bars are shown in blue.

282 a poor accuracy of the fitted parameters, which can be filtered out as faulty in a second
 283 step. The χ^2 minimizer used the Levenberg-Marquardt iterative algorithm implemen-
 284 tation from Newville et al. (2022). Examples of such fits on both orders are shown in Fig. 5.
 285 The agreement with observational data is good except in the 2.2-2.3 μm interval dom-
 286 inated by the poorly constrained CO₂-CO₂ absorption.

287 Emission angle is not included as a dimension in the computed look-up tables, since
 288 its influence on the whole spectral windows was found to be very close to the analyti-
 289 cal parametrization that we already used in Marcq et al. (2021), namely $I(\mu) = [0.26 + 0.74\mu] I(0)$
 290 where μ stands for the cosine of the emission angle. Finally, constant scaling coefficients
 291 for orders 5 and 6 were let as free parameters in the fitting routine, to account for slight
 292 residual differences in their respective absolute calibrations.

293 4 Results

294 4.1 Carbon monoxide (CO)

295 4.1.1 Latitudinal profile

296 The most robust feature of carbon monoxide distribution below the clouds of Venus
 297 is a permanent latitudinal gradient, first evidenced by NIMS/Galileo (Collard et al., 1993)
 298 and confirmed by all spectroscopic observations since then – see Marcq et al. (2018) for
 299 a detailed review of primary references. Fig. 6 displays our retrievals of carbon monox-

300 ide with respect to latitude, which confirms these previous observations with a minimum
 301 of 27 ± 2 ppmv (at 36 km) for latitudes below $\pm 20^\circ$ up to a maximum of 35 ± 2 ppmv
 302 near a latitude of $+50^\circ$, with statistical standard deviation and 1σ random accuracy be-
 303 ing similar.

304 These mixing ratio values are about 3 ppm higher than in our first analysis, which
 305 can be ascribed to either a secular increase, longitudinal variations (see Sec. 4.1.2) and/or
 306 to our updates in our radiative transfer model (see Sec. 3.1), more specifically it may be
 307 caused by our update from a 2-stream radiative transfer solver to an 8-stream model.
 308 Further investigations are needed to explain this discrepancy. In any case, this under-
 309 lines the fact that spectroscopic measurements are more suited to constrain variations
 310 rather than absolute mixing ratios, since the latter depend more on the assumptions made
 311 by the radiative transfer model (Bézard et al., 2009).

312 Interestingly, we see some evidence for north-south asymmetry, with a latitudinal
 313 gradient less steep in the southern hemisphere compared to the northern one, as well as
 314 a slight offset of the minimum in the $-20^\circ/+10^\circ$ latitude range. These details are very
 315 reminiscent of the 2009 observations from Arney et al. (2014), and will be discussed later
 316 in Sec. 5. We also have a larger dispersion of measurements in the southern hemisphere,
 317 but it is impossible to tell whether it is genuine or whether it results from our more ex-
 318 tensive sampling and/or smaller footprints in the southern hemisphere, first mentioned
 319 in Sec. 2.1.

320 *4.1.2 Other variabilities*

321 Thanks to the extensiveness of our data set, we can investigate the variability of
 322 carbon monoxide with respect to other variables, restricting to latitudes below $\pm 20^\circ$ in
 323 order to exclude any contamination by the well-documented latitudinal variability (Sec. 4.1.1).
 324 No significant variability with respect to local solar time was found, in contrast with the
 325 findings of Tsang and McGouldrick (2017) using VIRTIS-M, who found elevated values
 326 for CO between 10pm and 3am near the equator. However, using VIRTIS-H we could
 327 not observe the highest latitudes, where they also reported a ~ 2 ppmv dusk enhance-
 328 ment in CO, due to insufficient signal-to-noise.

329 Some low-latitude CO variability with longitude might be present in Fig. 7, with
 330 maximal values between 0 and 100° about 8 ppmv higher than the minimal values near

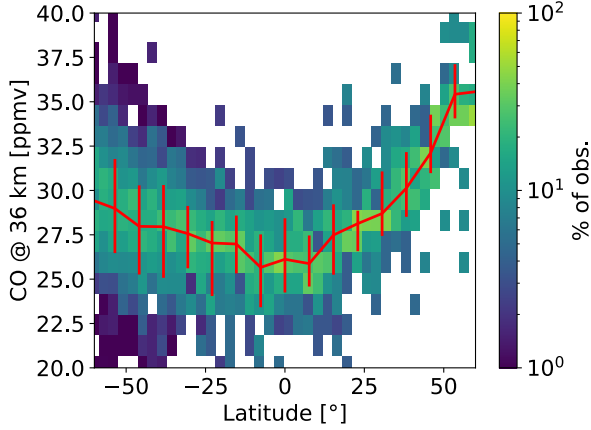


Figure 6. Carbon monoxide mixing ratios at 36 km with respect to latitude. The color code reflects the percentage of retrievals in the corresponding CO mixing ratio bin relative to all retrievals in the same latitudinal bin. The red curve stands for the moving median, and red bars represent the $\pm 1\sigma$ statistical dispersion around the median.

331 a longitude of 300° . An alternate interpretation would be that we observe a secular in-
 332 crease of CO between 2006 and 2011, since we are unable to disentangle both variables
 333 (see Sec. 2.1). Although this interpretation seems less likely, such an increase would help
 334 us to reconcile our comparatively high estimates of CO, since the CO mixing ratio val-
 335 ues we report for first 250 orbits – which were the only ones processed at the time of Marcq
 336 et al. (2008) – are indeed similar to those reported by Marcq et al. (2008). This would
 337 imply that their estimates were not fully representative of the longitudinal/temporal av-
 338 erage, due to insufficient longitudinal/temporal coverage.

339 4.2 Carbonyl sulfide (OCS)

340 4.2.1 Latitudinal variations

341 Carbonyl sulfide has been found to exhibit some degree of anticorrelation with car-
 342 bon monoxide from various ground based observations (Marcq et al., 2005, 2006; Arney
 343 et al., 2014) at a spectral resolution similar to ours. The latitudinal variations of OCS
 344 are shown in Fig. 8. A downward shift of about 1 ± 0.3 km of the OCS reference pro-
 345 file is found at 50°N compared to the equatorial value. Since a downward vertical shift
 346 of the OCS reference profile results in a decrease in OCS mixing ratio for a given alti-
 347 tude in the 30–40 km range, we are therefore able to confirm the latitudinal anti-correlation

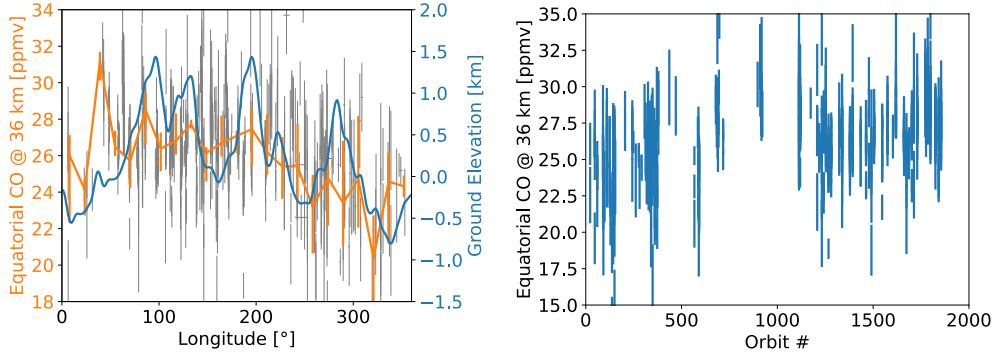


Figure 7. Carbon monoxide mixing ratios at 36 km with respect to surface longitude (left) and VEx orbit number (right), in both cases restricted to latitudes below $\pm 25^\circ$. Error bars stand for 1σ random uncertainties. The meridional (latitudes below $\pm 25^\circ$) average of topographic elevation according to Magellan-based digital elevation model used by Lebonnois et al. (2010) is also shown on the left.

348 between CO and OCS. Even the detailed features seen for CO latitudinal variations (Sec. 4.1.1)
 349 are found for OCS too: north-south asymmetry, maximal shift values in the $-20^\circ / +$
 350 10° latitude range, giving further credence to the CO/OCS anti-correlation.

351 Also, the anti-correlation is stronger compared to the one previously evidenced by
 352 e.g. Marcq et al. (2008)[their Fig. 6] where OCS was fitted through a simple multiplica-
 353 tive parameter. This strongly implies that this vertical shift does a better job to param-
 354 eterize OCS variability with a single parameter.

355 **4.2.2 Other variabilities**

356 Considering the latitudinal anti-correlation between CO and OCS, one would ex-
 357 pect the same anticorrelation to prevail between CO and OCS when plotted against lon-
 358 gitude, solar time, date, etc. In contrast, Fig. 9 shows a correlation pattern with lon-
 359 gitudinal elevation, and/or a possible increase with increasing date. Both would make CO
 360 and OCS shift altitude positively correlated with respect to secular variations. Hypothe-
 361 ses regarding these behaviors will be discussed and detailed in Sec. 5.

362 **4.3 Water vapor (H₂O, HDO)**

363 Our first attempt was to fit both H₂O and HDO mixing ratio separately, thereby
 364 measuring also the D/H isotopic ratio of water vapor, known to be much higher than

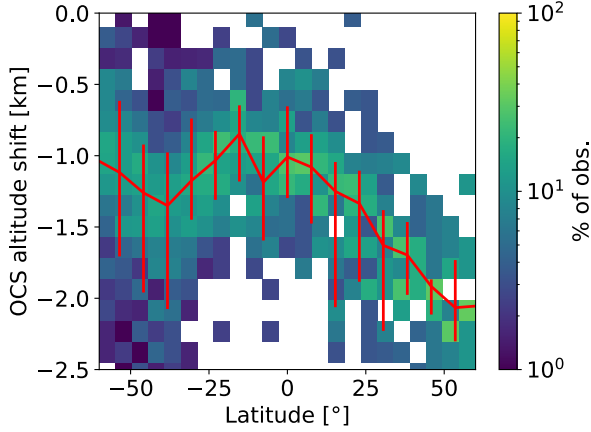


Figure 8. Vertical shift of OCS reference profile with respect to latitude. Color codes for the percentage of retrievals in the corresponding OCS vertical shift bin relative to all retrievals in the same latitudinal bin. The red curve and error bars represent the $\pm 1\sigma$ statistical dispersion around the median.

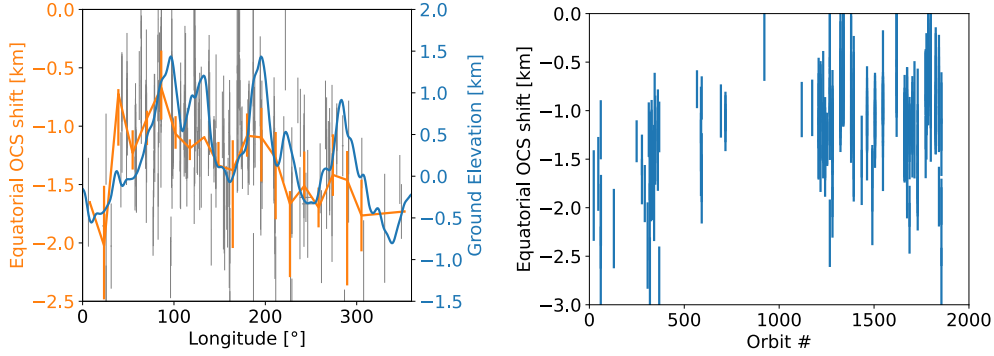


Figure 9. Carbonyl sulfide altitude shift with respect to surface longitude (left) and VEx orbit number (right), in both cases restricted to latitudes below $\pm 25^\circ$. Error bars stand for 1σ random uncertainties. The meridional (latitudes below $\pm 25^\circ$) average of topographic elevation according to Magellan-based digital elevation model used by Lebonnois et al. (2010) is also shown on the left.

365 the terrestrial value (de Bergh et al., 1991) at about 130 ± 40 SMOW (standard mean
 366 ocean water). Unfortunately, VIRTIS-H spectral resolution is not sufficient to achieve
 367 this, and a strong degeneracy was observed between H_2O and D/H retrievals, spanning
 368 a large interval (D/H ratio between 125 ± 50 , H_2O mixing ratio between 30 ± 10 ppmv
 369 at 35 km). To further proceed, we (1) assumed a constant D/H ratio of 130 and fit to-
 370 tal water vapor ($\text{H}_2\text{O} + \text{HDO}$) mixing ratio; or (2) assumed a constant water vapor mix-
 371 ing ratio at 30 ppmv (Marcq et al., 2018) and fit D/H ratio. Both results are shown in

372 Fig. 10 according to latitude. The latitudinal average is 27 ± 3.5 ppmv for water vapor
 373 mixing ratio, and 95 ± 25 for D/H ratio, in agreement with previous measurements, as
 374 well as with measurements obtained at lower altitude (Bézard et al., 2011; Fedorova et
 375 al., 2015) confirming that water vapor is well mixed in the troposphere.

376 1σ error bars yielded by the fitting algorithm are very close to the observed stan-
 377 dard deviation, which indicates that no significant trend in space or time can be inferred
 378 from our data set. Some hint of latitudinal variability might exist for water vapor mix-
 379 ing ratio (with a very weak equatorial maximum as seen in Fig. 10) although it would
 380 need further observations to be confirmed since it is below significance level with VIRTIS-
 381 H accuracy.

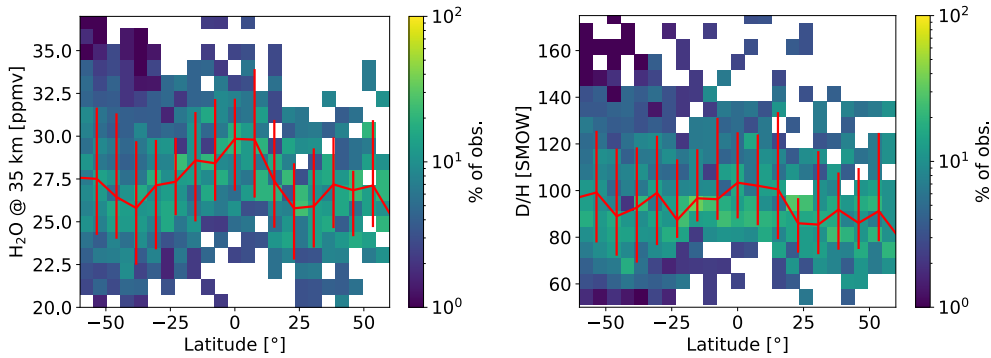


Figure 10. H₂O volume mixing ratio when assuming a constant D/H ratio of 130 (left) and D/H ratio assuming a constant water vapor mixing ratio of 30 ppmv (right) with respect to latitude. Color codes for the percentage of retrievals in the corresponding H₂O (left) and D/H (right) bin relative to all retrievals in the same latitudinal bin. The red curve and error bars represent the $\pm 1\sigma$ statistical dispersion around the median.

382 4.4 Sulfur dioxide (SO₂)

383 Our sulfur dioxide retrievals with respect to observed latitude are shown in Fig. 11.
 384 The average mixing ratio we find is 190 ± 40 ppmv near 35 km, somewhat higher than
 385 the commonly accepted value 130 ± 50 ppmv (Arney et al., 2014; Marcq et al., 2018),
 386 although in agreement with the latest iSHELL observations (Marcq et al., 2021), which
 387 made use of a very similar radiative transfer model and retrieval algorithm. However,
 388 the same caveats as in our relatively high mixing ratios retrieved for CO also apply here,
 389 so that variations are better constrained than absolute mixing ratios. Unfortunately, the

390 relatively poor accuracy ($\sim 20\%$) of these SO_2 retrievals does not allow us to confirm
 391 the latitudinal trend evidenced by Marcq et al. (2021), which highlights the necessity of
 392 a much higher spectral resolution to fully explore SO_2 variability below the clouds. Sim-
 393 ilarly to what we reported in Sec. 4.3 for water vapor, 1σ error bars yielded by the fit-
 394 ting algorithm for SO_2 are very close to the observed standard deviation, which indicates
 395 that no significant trend in space or time can be inferred from our data set.

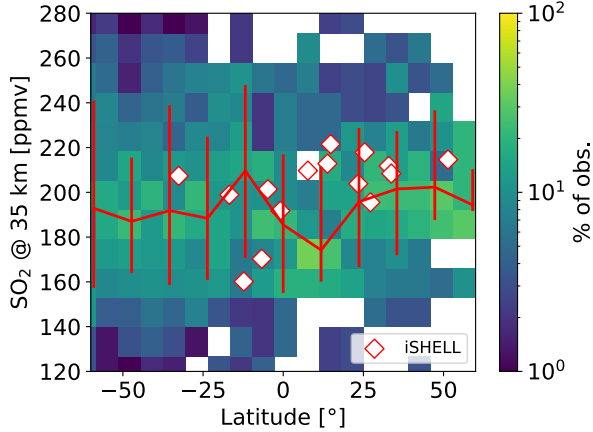


Figure 11. SO_2 volume mixing ratio with respect to latitude. Color codes for the percentage of retrievals in the corresponding SO_2 bin relative to all retrievals in the same latitudinal bin. The red curve and error bars represent the $\pm 1\sigma$ statistical dispersion around the median. Diamonds show the iSHELL retrievals from Marcq et al. (2021).

396 5 Interpretation

397 5.1 General framework

398 The classical interpretation of the latitudinal gradient observed for CO and OCS (Tsang
 399 et al., 2009; Tsang & McGouldrick, 2017) can be summarized as follows: for minor species
 400 which exhibit a significant vertical gradient in the probed layers (such as CO, and even
 401 more so OCS), their mixing ratio variations at planetary scale result mainly from a com-
 402 petition between vertical advection (driven by general circulation) and local chemical pro-
 403 cess which counterbalances this dynamical offsets. Within this framework, the latitudi-
 404 nal variations we have measured for CO and OCS (Sec. 4.1.1 and 4.2.1) are consistent
 405 with upwelling at lower latitudes and conversely downwelling at higher latitudes, so a
 406 Hadley-cell type circulation as expected from general circulation models (Lebonnois et

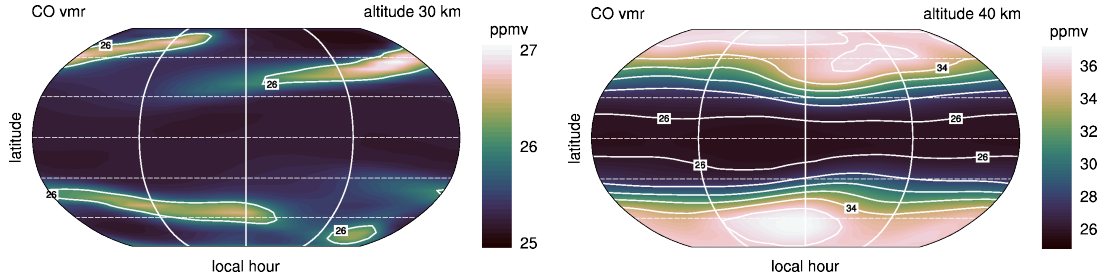


Figure 12. CO volume mixing ratio as modeled by Stolzenbach et al. (2023) at an altitude of 30 km (left) and 40 km (right). Dashed white lines indicate parallels (30° apart in latitude), and solid white lines indicate 6AM, noon and 6PM local solar times.

al., 2010; Stolzenbach et al., 2023). More specifically, the most recent 3D Venus planetary climate model including chemical processes (Stolzenbach et al., 2023) finds that, below the clouds in the 30-40 km altitude range, CO is increasing by 8 to 40% between 0° and $\pm 60^\circ$ in latitude, comparable with our observed variations of $\sim 30\%$ (Fig. 12). Their zonally averaged vertical profile of OCS at higher latitude is also shifted downwards between $\pm 60^\circ$ and 0° by a similar amount to our observations (~ 1.3 km compared to our ~ 1 km). Such an interpretation supports the choice we made to parameterize OCS variations, namely by vertical shifts of its reference profile. They also find a small enhancement in water vapor at lower latitudes (30 ppmv compared to a latitudinal average of 27-28 ppmv), compatible with our observations and their uncertainties. For SO_2 , their model is currently unable to reproduce an abundance of ~ 100 ppmv below the clouds, so that it cannot yet be used to explore SO_2 chemistry below the clouds.

This interpretation of OCS variations as vertical shifts of a nominal profile could be extended to other species, so that it would be interesting to convert our CO mixing ratios variations in terms of vertical translations of the reference CO profile. This yields $\Delta z = \frac{\Delta q_{\text{CO}}}{\partial q_{\text{CO}}/\partial z} \approx \frac{10 \text{ ppmv}}{1 \text{ ppmv/km}} \approx 10 \text{ km}$, where 1 ppmv/km is the value of the vertical CO gradient in the 30-40 km altitude range, and 10 ppmv the variations of CO between higher and lower latitudes that we observe. This equivalent 10 km vertical shift for CO is about 10 times larger than the vertical shift we observe for OCS. This highlights the fact that this altitude shift should not be understood as an actual vertical displacement (which would be common to all species), but as a more abstract quantity depending on the vertical advection, chemical relaxation time scale, local vertical gradient of the con-

429 sidered species, etc. Indeed, Marcq and Lebonnois (2013) found very different relaxation
430 times for CO and OCS.

431 Also, the region probed in our observations is close to the deep mixed layer of which
432 the height may reach ~ 30 km and above on some descending probe, with vertical wind
433 of several m/s that may have measured. From the *Pioneer Venus* probes (Seiff et al.,
434 1985), it seems that there is a strong variability of the depth of this layer with latitude
435 and local time. *Venera* and *Pioneer Venus* probes also measured a low vertical gradi-
436 ent of the zonal wind between 20 and 30-35 km (Schubert et al., 1980), suggesting of a
437 mixed layer with potentially non-negligible vertical wind. Only Baker et al. (2000) stud-
438 ied the small-scale turbulence in this layer, with vertical wind of speeds several m/s (Ainsworth
439 & Herman, 1975). This would be consistent with the observed variability of minor species
440 with a significant vertical gradient in their VMR profile, namely CO and OCS.

441 Finally, one can observe in our latitudinal variation pattern for both CO and OCS
442 a significant asymmetry between the northern and southern hemisphere, already suspected
443 by previous observers (Tsang et al., 2009; Arney et al., 2014). However, this asymme-
444 try could be also due to the asymmetrical coverage of Venus Express, with the south-
445 ern hemisphere being observed from further away but at much lower orbital speeds, and
446 therefore co-adding over about 10 times smaller footprints – this provides a reasonable
447 explanation for the larger statistical dispersion of measurements observed in the south-
448 ern hemisphere compared to the northern hemisphere which does not resolve small scale
449 variability. Nevertheless, if this asymmetry is genuine and in absence of significant sea-
450 sonal cycles, the most likely cause for this asymmetry would be the effect of topography
451 upon the vertical circulation, spectacularly evidenced up to cloud top by wave propa-
452 gation (Piccialli et al., 2014; Fukuhara et al., 2017). Intriguingly, although the meridional-
453 vertical circulation below the clouds modeled by Stolzenbach et al. (2023) is asymmet-
454 ric with respect to the equator, their modeling of the latitudinal variations of minor species
455 such as CO and OCS do not display a noticeable North-South asymmetry. It should also
456 be noted that, unlike Mars, Venus zonally-averaged topography does not exhibit a sig-
457 nificant North-South dichotomy; the only major source of North-South asymmetry be-
458 ing *Aphrodite Terra* centered around 20°S . We further investigate the idea of topographic
459 influence upon observed minor species variability in the following section.

5.2 Correlations with topography

Following the work of Bertaux et al. (2016), we have looked for correlation between mean topography, and zonally-shifted CO/OCS mixing ratios at lower latitudes (below $\pm 25^\circ$, so that we are not sensitive to the above mentioned latitudinal gradients). The result is plotted in Fig. 13. Interestingly, we find for CO a maximal correlation for a shift of about 45° in longitude, with larger values for (zonally shifted) CO above higher elevations (namely *Aphrodite Terra*), comparable with the shift value found by Bertaux et al. (2016) for cloud top level zonal winds, UV albedo and H₂O abundance in the same low latitude region. This suggests that the propagation of mountain waves interacts with the general circulation in the 30-40 km altitude range, resulting in a weaker low latitude upwelling in (zonally shifted) proximity of higher surface elevations.

Direct evidence for an influence of topography on the atmospheric waves was provided by Akatsuki, with mountain waves observed at cloud top level, preferably during the late afternoon (Kouyama et al., 2017). There are also recent observations (Encrenaz et al., 2020) and modeling (Stolzenbach et al., 2023) efforts that agree on a local minimum for cloud top SO₂ close to a longitude of 0° . Our observations of CO longitudinal variability below the clouds further strengthens the case for a direct influence of topography upon vertical motions in the Venusian troposphere (whether large scale advection or wave propagation), with a zonal shift explained by the zonal superrotation. A more quantitative assessment and interpretation would require mesoscale dynamical models (Lefèvre et al., 2020) coupled with chemical processes (including heterogeneous processes within the lower haze) at work below the clouds, which is not available yet. Coupled chemical-dynamical models exist at a planetary scale (Lebonnois et al., 2010; Stolzenbach et al., 2023) and are able to reproduce at least qualitatively the observed large scale latitudinal gradient of CO below the clouds, which gives us hope that, eventually, smaller-scale processes can be simulated too.

Fig. 13 also shows a different shift value for OCS than for CO (10° compared to 45° for CO), which is consistent for a shorter chemical relaxation time for OCS found by Marcq and Lebonnois (2013), making it a less sensitive tracer of the variations in vertical circulation induced by topography. Conversely, its steep vertical gradient (allowed by its short relaxation time) makes it better suited to monitor variations in vertical transport.

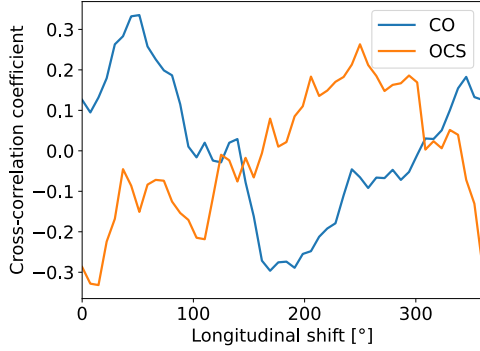


Figure 13. Cross-correlation (for latitudes below 25°) between zonally shifted surface elevation and CO mixing ratio / OCS altitude shift as a function of longitudinal shift.

6 Conclusion

The full VIRTIS-H night side $2.3\ \mu\text{m}$ data set allows us to confirm and refine the previous findings about trace species variability in the troposphere (Marcq et al., 2005, 2006, 2008; Tsang et al., 2009; Arney et al., 2014), namely: (1) a 15 to 30% increase in carbon monoxide (CO) with increasing latitude between the equator and $\pm 60^\circ$ in latitude; (2) an latitudinal anti-correlation between CO and carbonyl sulfide (OCS); (3) no evidence for significant latitudinal variability for water vapor (as well as its D/H isotopic ratio) and sulfur dioxide (SO_2); (4) confirmation of the north-south asymmetry in CO and OCS variations, pointing to an orographic influence on the deep large-scale atmospheric circulation. The globally averaged values are higher than previous estimates for some species (marginally for CO in the 30-40 ppmv range at 36 km, more so for SO_2 at 190 ± 35 ppmv), but this can be explained with our updated radiative transfer model – absolute mixing ratios are notoriously harder to constrain than their spatial or temporal variations, see e.g. (Bézar et al., 2011). We were also able to discover new features in this data set, the largest so far for $2.3\ \mu\text{m}$ night side spectroscopy: (1) OCS variations are better explained through vertical shifts of its abundance profile (very steep in the probed layers) rather than through a multiplicative factor, which is fully consistent with the aforementioned dynamical interpretation; (2) CO (and possibly OCS) exhibit a zonal variability at lower latitudes, correlated with the longitudinally shifted average surface elevation, yielding another hint about the influence of topography on the deep circulation.

513 These VIRTIS-H results highlight implicitly, through their limitations, the scien-
 514 tific needs for future spectroscopic measurements of the 2.3 μm window. First of all, a
 515 higher spectral resolving power ($\sim 10^4$ or higher) would allow for an enhanced accuracy
 516 on the retrievals, allowing to explore the variability of minors species other than CO and
 517 OCS, e.g. H₂O, HDO and SO₂ (Marcq et al., 2021). Also, the “dynamics vs. chemistry”
 518 interpretation of the observed spatial contrasts will require at some point a more quan-
 519 titative approach, based on the use of coupled chemical models (Stolzenbach et al., 2023)
 520 which already reproduce the qualitative trends, and mesoscale/large eddy simulations
 521 currently being developed (Lefèvre et al., 2018, 2020, 2022). This, in turn, emphasizes
 522 the need to observe the spatial contrasts with a resolution of ~ 30 km (ultimately lim-
 523 ited by the multiple scattering of the thermal emission within the cloud layers), much
 524 improved compared to VIRTIS-H (several hundreds of km).

525 Such improved performance is indeed expected for the payload of next generation
 526 Venus orbiters. In particular, the high resolution channel VenSpec-H of the VenSpec spec-
 527 trometer suite (Helbert et al., 2019) onboard ESA’s *EnVision* orbiter should be able to
 528 observe, among other spectral bands, the 2.3 μm spectral window at a spectral resolu-
 529 tion ~ 8000 and a spatial resolution ~ 100 km. Monitoring this spectral window for a
 530 nominal duration of six Venusian years will undoubtedly yield new insights about the
 531 coupling between dynamics and chemistry below the clouds of Venus, as well as provide
 532 spatial and temporal context for the complementary in situ measurements of trace species,
 533 e.g. those planned by the Venus Tunable Laser Spectrometer (VTLS) and Venus Mass
 534 Spectrometer (VMS) during NASA’s DAVINCI atmospheric probe descent (Garvin et
 535 al., 2022).

536 **Open Research Section**

537 VIRTIS-H data can be obtained from ESA’s Planetary Science Archive (PSA) : [https://](https://archives.esac.esa.int/psa/)
 538 archives.esac.esa.int/psa/. The VIRTIS/Venus Express dataset is also searchable
 539 from the Europlanet / VESPA portal (<https://vespa.obspm.fr>), cured by PADC at
 540 Paris Observatory.

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 547 4000128137).

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