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Retrieval of Martian atmospheric CO vertical profiles from NOMAD observations during the 1st year of TGO operations

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Key Points:

- Global map of CO profiles from NOMAD SO observations during the first year of TGO operations is presented for the first time.
- During the onset of the 2018 global dust storm, the CO VMRs are found to be depleted by 28% at 50 km compared to the average CO VMR values.
- High CO abundance at tropospheric altitudes is observed over NH during decay of the global dust storm when the atmospheric dust loading is high.

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Abstract

We present CO density profiles up to about 100 km in the Martian atmosphere obtained for the first time from retrievals of solar occultation measurements by the Nadir and Occultation for Mars Discovery (NOMAD) onboard ExoMars Trace Gas Orbiter (TGO). CO is an important trace gas on Mars, as it is controlled by CO₂ photolysis, chemical reaction with the OH radicals, and the global dynamics. However, the measurements of CO vertical profiles have been elusive until the arrival of TGO. We show how the NOMAD CO variations describe very well the Mars general circulation. We observe a depletion of CO in the upper troposphere and mesosphere during the peak period, L_S = 190°–200°, more pronounced over the northern latitudes, confirming a similar result recently reported by ACS onboard TGO. However, in the lower troposphere around 20 km, and at least at high latitudes of the S. hemisphere, NOMAD CO mixing ratios increase over 1500 ppmv during the GDS (Global Dust Storm) onset. This might be related to the downwelling branch of the Hadley circulation. A subsequent increase in tropospheric CO is observed during the decay phase of the GDS around L_S = 210° – 250° when the dust loading is still high. This could be associated with a reduction in the amount of OH radicals in the lower atmosphere due to lack of solar insolation. Once the GDS is over, CO steadily decreases globally during the southern summer season. A couple of distinct CO patterns associated with the Summer solstice and equinox circulation are reported and discussed.

Plain Language Summary

CO is an extremely interesting trace species in the Martian atmosphere. It has been used for both dynamical and photochemical studies of the atmosphere. But its vertical distribution has not been systematically measured until the arrival of the Exomars Trace Gas Orbiter (TGO). We use observations of the NOMAD (Nadir and Occultation for Mars Discovery) spectrometer onboard TGO to retrieve full profiles of mixing ratios of CO up to 100 km with a good vertical resolution. The retrievals cover two Martian seasons during which a global dust storm event occurred. We have found the behavior of CO during this event to be governed by local chemistry as well as by the long range transport. During the dust storm, CO mixing ratios are depleted all over the globe while over the southern high latitudes we discover an increase in CO due to transport from low latitudes during the end of the southern winter. The dynamical effect of global transport

62 is found in the vertical distribution of CO during the southern summer. Another impor-
63 tant result, where the local chemistry might be at play is the increase of CO in the low
64 altitudes over low and mid latitudes during the decay phase of the GDS.

65 1 Introduction

66 CO is a trace gas of large interest in the Martian atmosphere as its distribution is
67 ruled by photochemistry and dynamics. It is produced in the upper atmosphere of Mars
68 by photolysis of CO₂ and destroyed in the lower atmosphere by the hydroxyl radicals
69 (OH) (McElroy & Donahue, 1972; Parkinson & Hunten, 1972). Due to the long photo-
70 chemical lifetime of CO in the Martian atmosphere (González-Galindo et al., 2005) it
71 can be used as a dynamical tracer. As a non-condensable gas, CO shows strong varia-
72 tions in the polar regions during sublimation and condensation of the polar caps and a
73 slow seasonal variation in the low latitude region (Forget et al., 2008). The CO density
74 can also be used to constrain dynamical parameters of models such as eddy diffusion co-
75 efficients in the homosphere (Rodrigo et al., 1990; Yoshida et al., 2022).

76 CO on Mars was first detected from Earth by Kaplan et al. (1969) using high res-
77 olution infrared Fourier spectroscopy. Later, ground based observations used infrared (Billebaud
78 et al., 1991), microwave (Kakar et al., 1977; R. Clancy et al., 1983), millimeter (Lellouch
79 et al., 1991), and submillimeter (Encrenaz et al., 1991) spectroscopy. In more recent ground
80 based efforts, a positive north-to-south gradient of CO column averaged mixing ratio was
81 found in latitudinal mapping during summer (Krasnopolsky, 2015) and an opposite trend
82 was found during winter (Krasnopolsky, 2003). These ground based observations showed
83 an average CO volume mixing ratio (VMR) around 800 ppmv, with significant seasonal
84 and latitudinal variability.

85 Other instruments dedicated to the study of CO in the Mars' atmosphere are PFS
86 (Planetary Fourier Spectroscopy) and OMEGA (Observatoire pour la Minéralogie, l'Eau,
87 les Glaces et l'Activité) onboard MEX (Mars EXpress) (Encrenaz et al., 2006), CRISM
88 (Compact reconnaissance imaging spectrometer for Mars) onboard MRO (Smith et al.,
89 2009), and LNO (Limb Nadir Occultation) of NOMAD (The Nadir and Occultation for
90 MArS Discovery instrument) onboard TGO (Trace Gas Orbiter) (Smith et al., 2021). All
91 these instruments have significant coverage in both latitudes and seasons and have been
92 operating for several years. Recently, PFS measurements for 7 Martian years were used

93 to study the seasonality of CO from the nadir observations (Bouche et al., 2021). An-
94 other significant result found from the PFS observations is the low values of CO VMR
95 (Sindoni et al., 2011) during the perihelion season from high southern latitudes to 50°N.
96 In another study from the PFS observations, high constant values (1200 ppm) of the
97 CO column is found during $L_S = 330^\circ\text{-}360^\circ$ (Billebaud et al., 2009). OMEGA measure-
98 ments reveal the seasonal behavior of CO over the Hellas basin (Encrenaz et al., 2006)
99 where CO mixing ratios increase by a factor of 2 during winter. These results from PFS
100 and OMEGA indicate the seasonal and topographical effect on column abundance of CO.
101 A more detailed study of CO column with seasonal and spatial variations was performed
102 with the measurements from CRISM (Smith et al., 2009). These studies confirm deple-
103 tion of CO due to the sublimation of polar ice, a behavior predicted by Ar distribution
104 observed by the Gamma Ray Spectrometer (GRS) onboard Mars Odyssey spacecraft (Sprague
105 et al., 2004) and the Mars Global Climate Model developed at the Laboratoire de Météorologie
106 Dynamique (LMD-MGCM) (Forget et al., 2008) which is now known as Mars Planetary
107 Climate Model (Mars PCM) (Forget et al., 2022). CRISM measurements also show a
108 weak correlation of CO with surface pressure in the low latitude region (Smith et al., 2018).

109 From the above mentioned ground-based and space borne measurements, we know
110 a great deal about the variation of the column averaged CO and its qualitative relation
111 with the seasonal pressure cycle. Very recently, column averaged mixing ratio of CO is
112 reported by Smith et al. (2021) from the LNO measurement of NOMAD. They have es-
113 tablished a global average of 800 ppm. This revealed that CO seasonal variation is greater
114 in the polar regions than at the equator. The column measurements are mostly repre-
115 sentative of the surface CO VMR. However, in order to characterize its dynamical and
116 chemical sources and sinks, and underlying atmospheric variability, the density measure-
117 ments are necessary. For example, a recent study of the variation of Martian CO obtained
118 from the MIR (mid-infrared) channel of ACS (Atmospheric Chemistry Suite) onboard
119 TGO, found that CO is depleted during the MY 34 global dust storm (GDS) (Olsen et
120 al., 2021). The seasonal coverage used in Olsen et al. (2021) was limited. An extended
121 set of measurements from ACS but with a different spectral channel (NIR, near-infrared)
122 is included in this special issue (A. Fedorova et al. (2022)) which includes comparisons
123 with the measurements of MIR and TIRVIM.

124 Until the arrival of TGO, the empirical description of Martian CO density profiles,
125 with a good resolution in the vertical and in a systematic manner, was essentially ab-

126 sent. Here, we present vertical profiles of CO retrieved from the measurements recorded
127 by NOMAD solar occultation (SO) for the first year of TGO observations from April 2018
128 to March 2019, for the first time. This covers the last two seasons of MY 34 with fine
129 latitudinal coverage. The characteristics of the chosen dataset corresponding to these
130 observations are described in section 2 along with a brief description of the NOMAD SO
131 channel. Section 3 is dedicated to the preparation or cleaning steps of the data analy-
132 sis before they are used by our retrieval scheme. The retrieval scheme is described in de-
133 tail with an example of a scan retrieval in section 4. Results of the CO retrieval from
134 the above mentioned dataset are presented in section 5, where we describe the seasonal,
135 latitudinal, and diurnal variation in the CO distribution. The next section also includes
136 a proper comparison of the results with the model predictions.

137 2 The NOMAD Instrument and Dataset

138 NOMAD is a suite of three spectrometers onboard ExoMars Trace Gas Orbiter.
139 Two spectrometers operate in the infra-red (IR), the SO and the LNO. A third spectrom-
140 eter, UVIS (The Ultraviolet and Visible Spectrometer), operates in the ultraviolet and
141 visible (Vandaele et al., 2018, 2015). SO performs solar occultation and LNO performs
142 both limb scanning and nadir observation. The UV spectrometer is capable of scanning
143 the Mars atmosphere both in solar occultation and nadir viewing mode (Patel et al., 2017).
144 In this work, we will analyze data from the SO channel. This channel covers a spectral
145 range 2.2 – 4.3 μm and it has a nominal resolving power of 20,000 (Neefs et al., 2015).
146 NOMAD SO uses an echelle grating in Littrow configuration which means the incident,
147 reflected and Blaze angles are equal for this configuration (Neefs et al., 2015). The other
148 components of the SO channels are the detector, parabolic mirrors, and Acousto Opti-
149 cal Tunable Filter (AOTF). The NOMAD SO detector has 320 columns for the spectral
150 dimension and 256 for the spatial dimension. The AOTF acts as a bandpass filter con-
151 trolled by radio frequency input. Ideally, it selects one wavelength range for a radio fre-
152 quency input and sends it to the grating through parabolic mirrors, and diffracted light
153 falls on the detector after being guided by the parabolic mirrors. The wavenumber range
154 limit of the AOTF is adjusted to ideally capture only one diffraction order at a time. In
155 practice, adjacent diffraction orders do overlap and in the case of NOMAD SO see sec-
156 tion 3, this adds non-negligible components which need to be taken into account in the
157 forward model (Liuzzi et al., 2019; Villanueva et al., 2022). A quantitative description

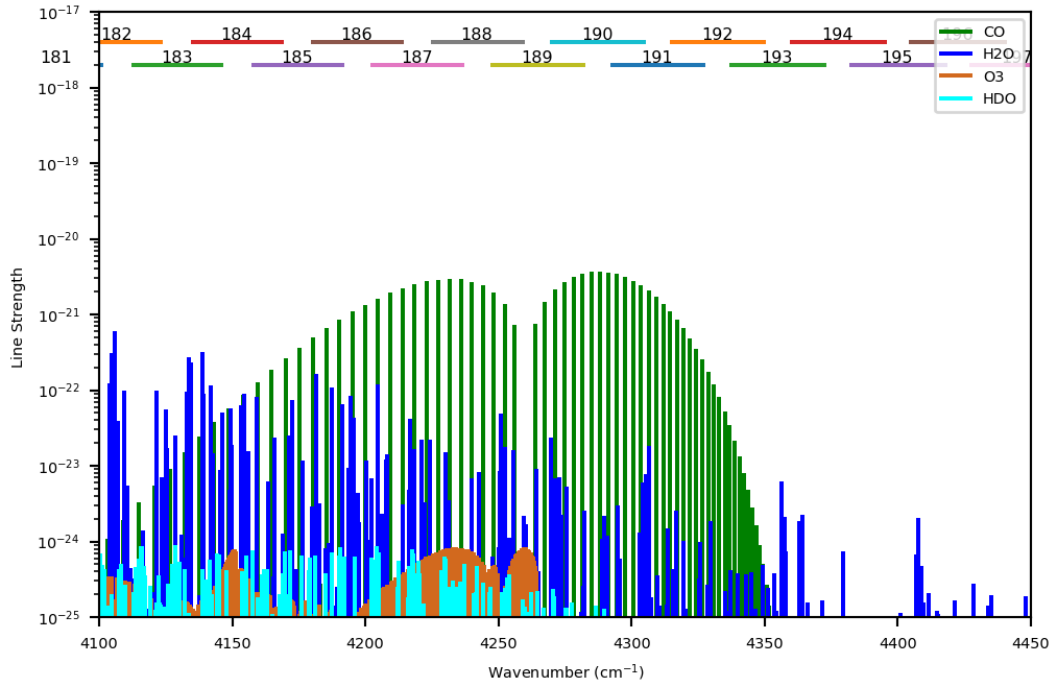


Figure 1. Spectral location of NOMAD SO diffraction orders for CO, and other atmospheric species with indication of strength of the strongest ro-vibrational absorption lines. The positions and line strengths are from HITRAN 2016 (Gordon et al., 2017).

158 of this contamination for the CO orders used in this work is presented below in section
 159 3. Once the diffraction order is selected, the incoming radiation is dispersed through the
 160 grating and incident on the detector's pixels. The intensity distribution of the diffrac-
 161 tion order depends on the AOTF transfer function and the blaze function (Liuzzi et al.,
 162 2019). The blaze function characterizes the diffraction efficiency of grating depending
 163 on the wavelength. These two functions, i.e., the AOTF transfer function and the grat-
 164 ing blaze function need to be very well characterized in order to simulate the NOMAD
 165 measured radiance correctly. For the complete optical description of the SO channel, one
 166 can refer to Neefs et al. (2015).

167 The NOMAD diffraction orders suitable for CO retrievals are shown in Figure 1,
 168 together with line positions from HITRAN 2016 (Gordon et al., 2017). In the NOMAD
 169 SO spectra, the CO rotational-vibrational lines are well separated and there is no sig-
 170 nificant absorption from other atmospheric species such as H₂O, CO₂ in the orders be-
 171 tween 186 and 191. The H₂O lines are shown for comparison purposes only; they are much
 172 weaker than the CO lines and are not observed in the NOMAD data. We choose order

173 190 for the retrieval because some of the strongest CO absorption lines lie in this order.
174 Also, the number of atmospheric scans performed by NOMAD SO is much larger than
175 in any other diffraction order during the first year of operations.

176 **3 Data Cleaning and Pre-processing**

177 In this work, we use NOMAD Level 1 calibrated transmittances. These are down-
178 loaded as formatted data and provide values of atmospheric transmittances, measure-
179 ment noise, and calibration parameters such as the parameters for the pixel to wavenum-
180 ber conversion, the center of the blaze function, etc. We first clean these transmittances
181 from diverse systematics such as spectral bending and shift (see section 3.2). before in-
182 gesting it into the actual inversion scheme. We call this internal step pre-processing and
183 it is described below in section 3.2. Both pre-processing and retrieval, make use of a line-
184 by-line forward model, central to our analysis, which is described next.

185 **3.1 The KOPRA Forward Model**

186 We use the Karlsruhe Optimized and Precise Radiative transfer Algorithm (KO-
187 PRA) described in Stiller (2000), to simulate the NOMAD SO measurements, both dur-
188 ing the “data cleaning” phase and at the core of the inversion processor. KOPRA has
189 been widely used for the retrievals of Earth atmospheric gases and aerosols (Schreier et
190 al., 2018; Höpfner & Emde, 2005). KOPRA solves the radiative transfer (RT) equation
191 in a line-by-line approach to calculate monochromatic radiance. In our adaptation to Mars
192 solar occultation conditions, the atmosphere is divided into layers of 1 km width from
193 the ground to 60 km, layers of 2 km from 60 km to 90 km, 2.5 km from 90 km to 130
194 km, and 5 km from 130 km to 220 km. The RT calculations are done along the line of
195 sight of the observations. The model calculates the line absorptions from the spectro-
196 scopic information provided by the HITRAN 2016 database (Gordon et al., 2017).

197 The instrumental line shape (ILS) of the NOMAD SO is one of the essential pa-
198 rameter for atmospheric retrievals as it reflects the manner in which the spectra are recorded
199 across the detector (Liuzzi et al., 2019; Aoki et al., 2021; I. R. Thomas et al., 2022). The
200 NOMAD SO instrumental response, including the AOTF and the ILS have been the sub-
201 ject of a recent revision, and we use a parameterization from a consolidated approach
202 in collaboration with various teams within the NOMAD consortium (Villanueva et al.,

203 2022). The ILS is parameterized as double Gaussian function. Its peaks, widths and the
204 distance between the Gaussian functions for every order have been determined combin-
205 ing several analytical and retrieval methods. The variability of the ILS within each or-
206 der and across orders is a NOMAD-SO specific difficulty which we consider as well char-
207 acterized, as can be seen from inspection of the retrieval’s residuals. However, for the
208 AOTF transfer function, its parameterization is further fine-tuned around the values sug-
209 gested by Villanueva et al. (2022). This fine-tuning is described in Appendix A, and per-
210 mitted a significant improvement in the retrievals. We convolved the KOPRA spectra
211 with the revised ILS, the fine-tuned AOTF, and the blaze function (Brines et al., 2022).
212 This convolution is done within the forward model.

213 3.2 Pre-processing

214 An example of NOMAD calibrated transmission data from one particular scan in
215 diffraction order 190 is shown in Figure 2, panel a. Recorded against pixel number, trans-
216 mittances are transformed into wavenumber dependent function during the nominal cal-
217 ibration, using a second order polynomial relation between wavenumber and pixel num-
218 ber (Liuzzi et al., 2019). The different colors correspond to different tangent heights, and
219 the vertical lines in red indicate the position of the center of the CO rotational-vibrational
220 lines for the main order 190. The transmittances clearly suffer from spectral shift and
221 bending of the continuum across the detector (Figure 2, panel a) which occurs due to
222 thermally induced mechanical stress on the detector Liuzzi et al. (2019). This bending
223 effect is almost ubiquitous, present in all NOMAD orders at all altitudes and scans (Vandaele
224 et al., 2018). It changes from each altitude to the next in a quasi-random manner, and
225 systematically responds to a (variable) 4th order polynomial. Notice that this is very dif-
226 ferent to the atmospheric continuum due to aerosols absorption, which can be consid-
227 ered as spectrally constant due to the narrow span of the NOMAD orders (Vandaele et
228 al., 2018) and which is usually important at low altitudes.

229 Before using these data in a proper inversion of densities they need to be corrected
230 for these two effects. For this purpose, the NOMAD transmittances are divided into small
231 micro-windows (MW) containing at least one absorption line (see Figure 2). The spans
232 of these MWs are shown in Figure 2 and are $4273.50 - 4275.80 \text{ cm}^{-1}$, $4277.20 - 4282.200$
233 cm^{-1} , $4283.2 - 4288.80 \text{ cm}^{-1}$, $4290.00 - 4295.20 \text{ cm}^{-1}$, and, $4296.20 - 4301.00 \text{ cm}^{-1}$.

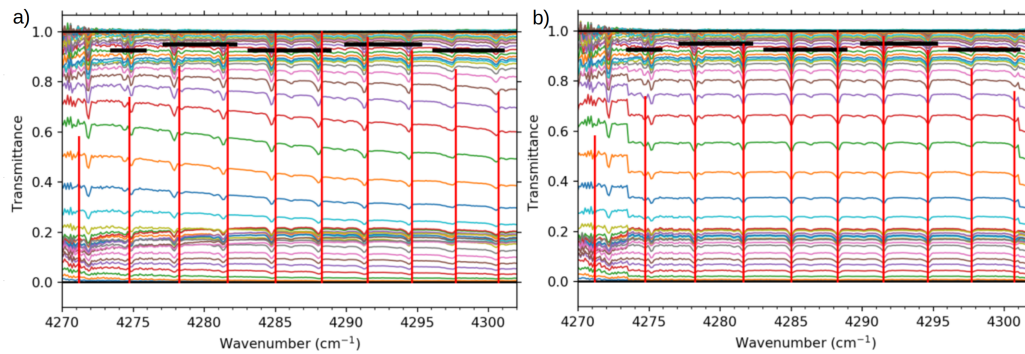


Figure 2. Example of our cleaning method applied to diffraction order 190 in one specific scan (20180503_151030_SO_A_E). Panel a, all the original spectra (calibrated transmittances) taken in this scan, showing clear bending effects and spectral shifts. Panel b, spectra after cleaning. Notice the cleaning is only applied to a subset of the full diffraction order. Vertical red solid lines indicate the expected position of the CO spectral lines (HITRAN 2016). The five short horizontal lines in black near the top of the panels indicate the microwindows used for this cleaning. see text for details.

234 Starting with the bending effect, in all NOMAD orders this is usually very small
 235 in the central part of the order. In each order it is corrected using a polynomial fitting
 236 to the background continuum or baseline, as simulated with KOPRA at each tangent
 237 altitude. This correction is used as a multiplicative factor because the ultimate reason
 238 for this effect comes from small changes in the AOTF response during each SO scan. This
 239 explains the spectrally low variation (4th polynomial) in the spectra. During these changes
 240 the AOTF varies very little in the center of the order, where it is essentially one. There-
 241 fore the effect on the measured radiances should be multiplicative, and consequently, also
 242 on the transmittances. The use of a line-by-line calculation at each altitude to correct
 243 for the bending guarantees that the effect from gas line-overlapping is taken into account
 244 and is separated from the actual bending effect. Still, this correction may be a source
 245 of uncertainty for the retrieval of aerosol properties, and a simple strategy to minimize
 246 this is to select the center of the order to derive them at each altitude. This is the topic
 247 of another work (Stolzenbach et al., 2022) by our team, also submitted to this special
 248 issue.

249 Regarding the spectral shift, present in all spectra, i.e., at all altitudes, and vari-
 250 able from scan to scan, this is determined from comparisons of data and model simu-

251 lations at all altitudes with significant line absorption (to minimize errors during its de-
252 termination). These spectral shifts, as it also occurs in other diffraction orders, vary across
253 the order in a linear manner. A linear function is fitted on the values of the shifts ob-
254 tained for each MW, and this is interpolated across the wavenumber range of the spec-
255 tra for a full-order correction. Microwindows near the edge of the order are not consid-
256 ered due to higher noise there. Details of these corrections are presented in a compan-
257 ion paper in this special issue (López Valverde et al., 2022).

258 Our inversion scheme uses radiances instead of transmittances. Thus the cleaned
259 Level-1a transmittances are converted into solar radiances using a space view simulation
260 with KOPRA, at 220 km tangent altitude, using the AOTF appropriate for this diffrac-
261 tion order and the grating properties described by the blaze function (Liuzzi et al., 2019).
262 The space view radiance is calculated using a reference solar radiance supplied externally.
263 This reference radiance has no other function and has no impact on the inversion.

264 One additional caution, related to the MWs selection, deserves some discussion.
265 As mentioned before, one characteristic of real AOTF spectrometers is that the addi-
266 tion of adjacent diffraction orders to the main order is not negligible. These contribu-
267 tions, when added to the forward model, guided the selection of the MWs in this study
268 (Villanueva et al. (2022) also see Appendix A). Figure 3 shows one example of this con-
269 tamination by adjacent orders in the case of order 190. The vertical lines in the plot in-
270 dicate the positions of CO absorption lines from different adjacent orders. The main or-
271 der is indicated with the solid green color and the adjacent orders on both sides with the
272 same color but with a solid line on the left and with a dotted line on the right. Three
273 central absorption lines of the spectra within 4285 cm^{-1} and 4292 cm^{-1} are not at all
274 contaminated with the absorption from the adjacent orders. As can be seen from Fig-
275 ure 3 most of the contaminations are from orders 187, 188, 189 on the left and 191 on
276 the right. A maximum of four orders contamination to either sides is shown in Figure
277 3 which is sufficient to explain the absorption line in the diffraction order 190. This con-
278 tamination, in addition to line strength considerations, was taken into account during
279 the selection of the MWs used in this work. For the inversion of CO (next section) we
280 finally selected two MWs near the center of the order, shown in Figure 3.

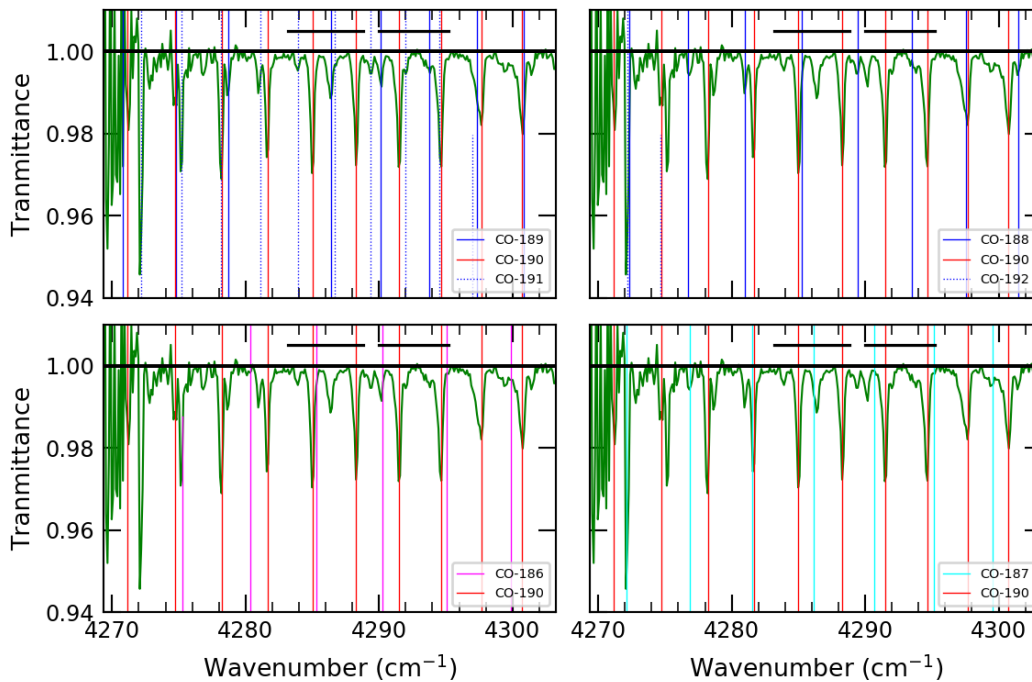


Figure 3. Example of contamination from the four adjacent orders at each side of the principal diffraction order 190. Colored vertical lines represent the position of the CO absorption present in the different orders as indicated in the legends. Four panels are used for clarity, showing different adjacent orders in each of them versus the central order 190. The selected MWs for the inversion are shown in thick black horizontal lines. See text for details.

281 4 Retrieval Scheme

282 After the cleaning of the Level-1a transmission data described above, we performed
 283 the CO retrieval using an inversion processor called Retrieval Control Programming (RCP)
 284 described in von Clarmann et al. (2003). We summarize here some of its characteristics.
 285 Our global-fit retrieval scheme and application are common to other NOMAD retrieval
 286 targets at the IAA team, including H₂O (Brines et al., 2022), temperature and CO₂ den-
 287 sities (Lopez-Valverde et al, 2022), and aerosols (Stolzenbach et al., 2022), all in this spe-
 288 cial issue.

289 RCP uses KOPRA as its forward model and follows a usual iterative approach to
 290 solve the inverse solution to the RT equation, based upon a Levenberg-Marquardt least
 291 square minimization algorithm. RCP can use diverse solution methods, like an optimal
 292 estimation approach, but in this work, we use a more flexible first order Tikhonov reg-
 293 ularization. In each iteration, it runs KOPRA and the simulated spectra are compared
 294 to the measured ones at each altitude simultaneously until convergence is reached when
 295 the variation in the target quantities are smaller than a fraction of the noise error (Jurado Navarro
 296 et al., 2016). RCP also provides averaging kernel (AK) and noise error for the target.
 297 The AK is expressed as $\mathbf{A} = \mathbf{G}\mathbf{K}$ where \mathbf{G} is the gain matrix and \mathbf{K} is Jacobian ma-
 298 trix (Rodgers, 2000). The gain matrix is defined in Rodgers (2000) as follows,

$$\mathbf{G} = (\mathbf{K}^T \mathbf{S}^{-1} \mathbf{K} + \mathbf{R})^{-1} \mathbf{K}^T \mathbf{S}_y^{-1}. \quad (1)$$

\mathbf{K} is the Jacobian matrix, \mathbf{S}_y is the covariance matrix for the measurement noise and
 \mathbf{R} is the regularization matrix. The value of the AK is always below unity. The noise
 error is mapped onto the retrieval grid as defined in the equation below.

$$\mathbf{S}_x = \mathbf{G}^T \mathbf{S}_y \mathbf{G}. \quad (2)$$

The χ^2 is defined as:

$$\chi^2 = (\mathbf{y} - \mathbf{F}(\mathbf{x}))^T \mathbf{S}_y^{-1} (\mathbf{y} - \mathbf{F}(\mathbf{x})) + (\mathbf{x} - \mathbf{x}_a)^T \mathbf{R} (\mathbf{x} - \mathbf{x}_a), \quad (3)$$

299 where \mathbf{y} is the measurement, $\mathbf{F}(\mathbf{x})$ is the forward model \mathbf{x}_a is the *a priori* state vector
 300 and \mathbf{x} is the true state vector which includes our target CO abundance profile. The re-
 301 trieval of CO with our retrieval scheme is found to be robust against large variations in
 302 the *a priori* values of CO (see Appendix B).

303 The *a priori* CO profiles extended from the planet's surface up to 200 km and are
304 obtained from the Mars PCM (Forget et al., 1999; Lefèvre et al., 2021) using the dust
305 scenarios for MY 34 and MY 35 (Montabone et al., 2015, 2020). The model uses dust
306 and water cycles described in (Navarro et al., 2014). The model outputs are at defined
307 grids and are interpolated to the time and location of the NOMAD SO observation. The
308 atmosphere needed by the forward model is built using the pressure and temperature
309 retrieved from SO observation of diffraction order 149 (López Valverde et al., 2022).

310 In a typical CO VMR profile retrieval, RCP performs the simultaneous inversion
311 of 3 quantities or targets, the CO VMR, a spectral shift, and a baseline accommodat-
312 ing factor named scale-h. The shift is used as a target with the intention to detect any
313 residual spectral shift remaining after the correction already performed at the pre-processing
314 level. Scale-h is a spectrally constant transmittance level within each MW, adjusted to
315 account for the impact of aerosol absorption on the continuum and any small remain-
316 ing calibration biases in each MW. As mentioned above, the continuum absorption within
317 the narrow NOMAD diffraction orders is spectrally flat (Vandaele et al., 2018) but with
318 this Scale-h parameter we allow for small variations between MWs.

319 Before the actual application to real measurements, an extensive study of synthetic
320 retrievals, generated using KOPRA, was performed. These were very useful to test the
321 performance of the inversion, its independence on *a priori* assumptions, and to fine-tune
322 the regularization. Appendix B presents some details and results from the synthetic re-
323 trievals.

324 4.1 Typical Retrieval Performance

325 To illustrate the performance of an inversion scheme, it is of paramount importance
326 to examine the residuals or best fit obtained, and to characterize the error propagation
327 and vertical resolution by an analysis of the AKs.

328 An example of the best fit spectrum achieved with our inversion scheme is shown
329 in the upper panel of Figure 4, for one particular scan and a tangent altitude of 74 km.
330 Figure 4 also shows the spectrum generated with the *a priori* CO profile, for compar-
331 ison. The lower panel depicts the residual transmittances (at the end of the iteration and
332 convergence) together with the model-data difference when using the *a priori*. The best

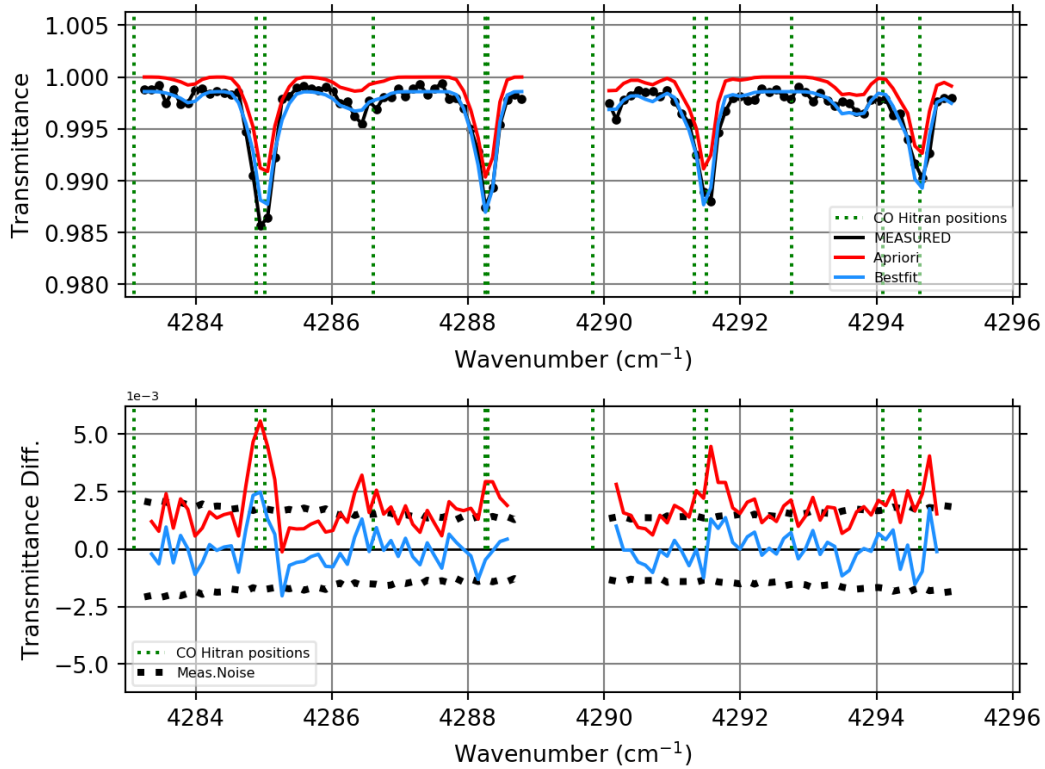


Figure 4. Best fit spectrum to the NOMAD cleaned transmittances at a tangent altitude of about 74 km from scan 20180801 055956 SO A E 190 (Lat 66°N, Lon 161°W, Ls 222°). The top panel shows the measured spectrum in solid black line with dots, the best fit spectrum at the end of the inversion is blue, and the spectrum obtained with the *a priori* atmospheric profile in red. The spectrum is split in 2 segments which correspond to the 2 MWs used in the inversion. The lower panel shows the residuals (Model fit - NOMAD) in blue, together with the same quantity but calculated with the model spectrum for the *a priori* atmosphere, in red. The dotted lines represent the NOMAD SO measurement errors used in the inversion.

333 fit residual transmittance spectrum lies within the limit of the measurement noise at all
 334 wavenumbers.

335 Figure 5 shows a set of typical diagnostics for the scan in Figure 4. From left to
 336 right, the four panels show the retrieved CO profile, the rows of the AK, the percent-
 337 age error at each altitude, and the vertical resolution profile, respectively. The averag-
 338 ing kernel shows the mapping of the measurements onto the retrieval grid. The verti-
 339 cal resolution is the usual interpretation of the full width at half maximum of the AK
 340 at each retrieval altitude. The CO VMRs retrieved in this example are greater than the

341 *a priori* (shown in red), specially above 60 km. The errors are typically within 10%–
342 15% below 80 km. The vertical resolution is within 5 km in the region below 80 km and
343 increases with altitude, as the measurement noise and the retrieval error also do. The
344 shaded region below 28 km, in the first panel, indicates the altitude where the slant op-
345 tical depth is larger than 1.5. We do not retrieve CO in the altitudes with high dust opac-
346 ities to avoid biases from the severe contamination in (reduction of) the spectral absorp-
347 tion lines. For such spectra, the averaging kernel values obtained in the retrieval diag-
348 nostics are zero, corresponding to no information.

349 After few experiments, we make use of a minimum value (0.03) of the AK. Retrieved
350 CO densities with AKs below this minimum are not trusted. All the results presented
351 in this study are qualified by this condition. Figure 5, panel b shows the diagonal of the
352 AKs for a typical NOMAD SO scan and we observe that below about 80 km and down
353 to 60 km these peaks are basically constant and they show small trend, with decreas-
354 ing values below 60 km. This slow decrease in the AK could be indicative of line satu-
355 ration. The lines used in order 190 are among the strongest in the IR spectrum. We made
356 a study of the line depths in these limb geometry data in a small number of solar occul-
357 tation scans. This study show us that the saturation effect in the line absorption start
358 below about 80 km and becomes stronger. However, our forward model is a state-of-the-
359 art line-by-line radiative transfer code that reproduces well the saturation effects in the
360 line absorption. In the case of saturated absorption lines, the information is mostly de-
361 rived from the wings of the absorption lines, because the line depth at the line centre
362 does not vary with the increasing CO density. How much information can be extracted,
363 i.e., how large is the sensitivity to the CO density, is provided by the AKs.

364 4.2 Effect of Climatology on Retrieved CO

365 We studied the effect of the *a priori* temperature, by using the PCM as a refer-
366 ence and adding ad-hoc perturbations on this profile. Here we discuss one example, for
367 scan 20180423 204351 1p0a SO A I 190. Figure 6, shows two retrievals, from the same
368 SO scan using two sets of pressures and temperatures. The profiles in black correspond
369 the *a priori* obtained from the PCM. The red lines correspond to the retrieval using as
370 *a priori* the *T/P* profile obtained from the inversion of this NOMAD SO scan (in diffrac-
371 tion order 149, results obtained by López Valverde et al. (2022)). The retrieved CO VMR
372 and absolute number densities in the two cases are shown in panels c and d respectively.

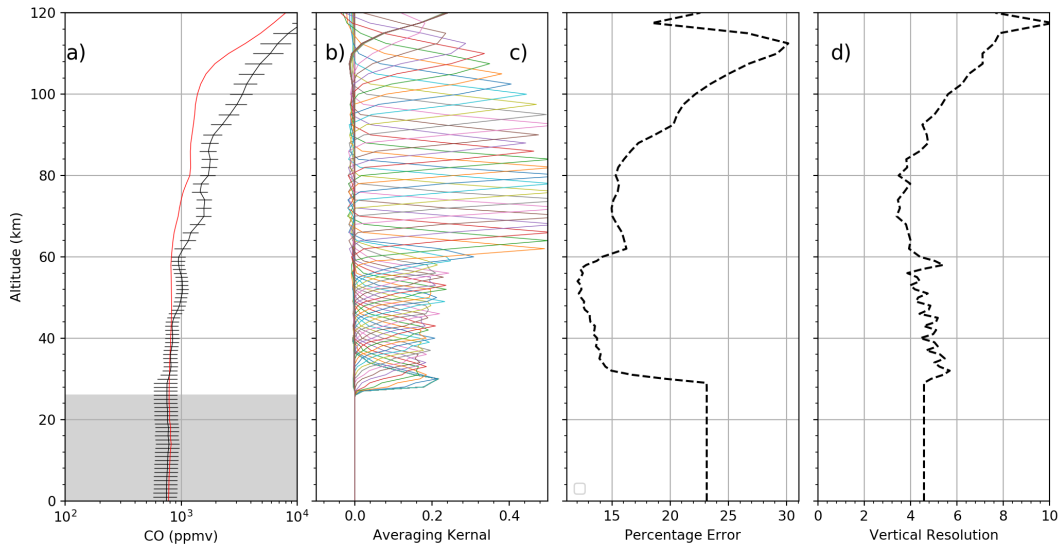


Figure 5. Retrieved CO profile from the same scan as in Figure 4 with error bars (panel a). The red line is the *a priori* profiles, the black line is the retrieved CO. The shaded area below 28 km represents the region of very high dust loading, where the SO spectra are not inverted. Panel b: selected rows of the Averaging Kernel matrix; the sharp change in the peak of the averaging kernel at 60 km is due to the change in the retrieval grid (see section 3.1 for the retrieval grid definition). Panel c: percentage error in the CO profile. Panel d: Vertical resolution, as given by the widths of the averaging kernel rows.

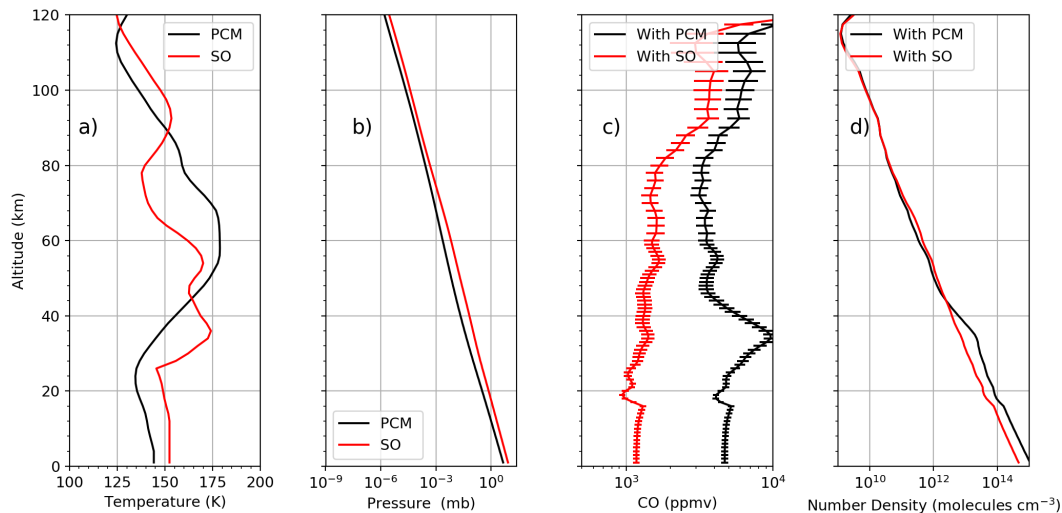


Figure 6. The effect of the reference thermal structure on the retrieved CO. Panel a: Temperature profiles from PCM (black line) and from NOMAD retrievals in order 149 (red line). Panel b: Pressure profiles from PCM (black) and from NOMAD (red). Panel c: retrieved CO VMR using the PCM inputs (black line) versus the NOMAD retrievals (red lines). Panel d: Same as Panel c but for the CO local number density (cm^{-3})

373 The obtained NOMAD pressures are higher than the PCM ones at each altitude, which
 374 in turn, results in a lower VMR than when using the PCM. The absolute CO density
 375 should be more independent of the densities assumed than the CO VMR, and this in-
 376 deed observed above 50 km. But this is not the case of its dependence on the temper-
 377 atures. And we observed in panel (d) that the CO absolute densities are varying with
 378 the assumed temperatures, with a crossing point around 45 km tangent height, which
 379 is present in both temperature and retrieved CO. Since this temperature effect is very
 380 important, in this work, we only used scans for which retrieved NOMAD temperatures
 381 are available (from a parallel effort in our team, see companion paper on temperature
 382 retrievals led by M. A. Lopez-Valverde in this special issue). In other words, we only con-
 383 sidered the subset of NOMAD SO scans where orders 190 and 149 were measured simul-
 384 taneously, and the final set consists of those where the temperature converged, which is
 385 a very high percentage of the available scans (López Valverde et al., 2022). This is done
 386 regardless of the difference between the *a priori* and the retrieved thermal structure.

5 Results and Discussion

The results of all our retrievals are shown in Figure 7. Due to the large variation in the CO relative abundance with altitude, we show the CO VMR values in log scale. The dataset extends from $L_S = 160^\circ$ to $L_S = 354^\circ$ (Figure 7, panels a and b) covering important seasons such as the onset of the global dust storm ($L_S = 190^\circ$) (Guzewich et al., 2019), the GDS decay phase ($L_S > 235^\circ$), the end of the southern spring ($L_S = 245^\circ - 263^\circ$), and the whole of the southern summer. In this section, following López Valverde et al. (2022), we refer to the atmospheric region from the surface up to 50 km as troposphere, and to that above as mesosphere.

5.1 Seasonal Variation of CO

CO is a long lived gas species affected by the general circulation in the Martian atmosphere, and therefore an excellent tracer of the global dynamics on Mars (Lefèvre & Krasnopolsky, 2017). Figure 7 permits some insight into the seasonal variation of the CO VMR; panels c and d show the CO VMR distribution over the northern and southern hemispheres respectively. In general, and in agreement with PCM simulations, we observe a CO VMR increasing with altitude in the upper troposphere and through the mesosphere, as a result of the downward mixing from the CO source region at high altitude, following CO_2 photodissociation (Krasnopolsky, 1995; González-Galindo et al., 2009; Modak et al., 2020). Particularly large values and gradients are observed above about 60 km, but with significant variations. This variability can be better visualized by inspecting a given CO VMR iso-contour in Figure 7, like the value of 2500 ppmv (in orange color). This value is arbitrary but serves as a marker between two altitude regions with different physics regarding CO; the upper one driven by photochemical production and the lower one dominated by CO losses. CO VMR values as large as 2500 ppmv occur very high in the atmosphere at mid and low latitudes, following CO_2 photodissociation, but can be seen at much lower altitudes towards the poles due to global dynamics, as discussed below in section 5.2. This iso-contour is therefore particularly useful to illustrate the CO variability, both with season and latitude. Panels a and b show the track of the solar occultation, where this mix of changes with season and latitude can be seen. Latitude has a large impact, different in the north and south hemispheres since the coverage of high latitudes is different in each hemisphere. Over the southern high latitudes the occultation spent less time which produces sudden changes in the iso-contour. As

419 we discuss below in section 5.2, it is possible to decouple the latitudinal and seasonal trends
420 to study only one of these, for example latitudinal variation for a small seasonal period.

421 There are two essential and different seasons where the dynamical behavior of the
422 Martian atmosphere is distinct, the equinoxes and the solstices. So, we analyze here the
423 CO distribution in these two periods, and also its variability in between. During the north-
424 ern autumn equinox period ($L_S = 160^\circ - 180^\circ$) when two Hadley cells extend from
425 the equator to high latitudes (Forget et al., 1999) and the solar insolation is similar in
426 both hemispheres, we expect similar CO mixing ratios at similar latitudes in the north
427 and south. And this is apparently the case in Figure 7, although unfortunately, only a
428 few NOMAD scans are available during that period. For example, over the northern high
429 latitudes ($> 60^\circ\text{N}$) high values (~ 1500 ppm) of CO VMRs are observed down to 20
430 km. Similar values of CO are also observed over the southern latitude 60°S .

431 The appearance of the MY 34 GDS in 2018 perturbed the global circulation and
432 we expect the CO distribution to depart from what should be its regular seasonal cy-
433 cle. We observe in Figure 7 that during the onset of the GDS in the period $L_S = 190^\circ$
434 to $L_S = 205^\circ$, the high altitude VMRs decrease (when looking at a fixed altitude, spe-
435 cially in the mesosphere). This shows the well known tropospheric warming by solar heat-
436 ing of aerosols and the associated upwards expansion in atmospheric density, moving our
437 iso-contour of 2500 ppm up to 100 km in the NH (northern hemisphere) during the GDS
438 peak ($L_S = 205^\circ$). Over the southern polar latitudes, however, the aerosols amount were
439 lower, and that CO VMR iso-contour can be seen around 75 km.

440 After the onset of the GDS, the 2500 ppmv iso-contour shows a CO VMR quickly
441 increasing at all mesospheric altitudes in the NH until $L_S \sim 225^\circ$. During the GDS,
442 the high dust loading warms (and expand) the atmosphere which, in turn, reinforces the
443 hemispheric Hadley cells. This increased circulation, has been shown to transport trace
444 gases up to mesospheric altitudes (Neary et al., 2020; Vandaele et al., 2019; Shaposhnikov
445 et al., 2019; R. T. Clancy et al., 2010). Since $L_S \sim 225^\circ$ until about $L_S \sim 282^\circ$ the
446 iso-contour stays approximately constant at 70 km tangent height. Surely the first part
447 of this period is dominated by the decay phase of the GDS while the second may be more
448 typical of a seasonal behavior, i.e., typical of a non-GDS year. The time evolution of the
449 mesospheric CO VMR over SH (southern hemisphere) at mid-latitudes does not show
450 those two different periods after the GDS. We observe a steadily small decrease with time

451 up to about $L_S = 300^\circ$. Our iso-contour is located at higher altitudes than in the NH,
452 which shows an inter-hemispheric asymmetry, specially evident around the southern sum-
453 mer solstice, $L_S = 270^\circ$. At the end of the southern summer, after $L_S = 330^\circ$, the CO
454 VMRs increase all over the globe, in both hemispheres. This increase is particularly no-
455 ticeable at tropospheric altitudes.

456 The distribution of CO VMRs below 50 km reflects a completely different seasonal
457 variability because it is not directly affected by CO₂ photolysis. During the onset of the
458 GDS, CO mixing ratios are depleted at these altitudes. A similar depletion is also re-
459 ported by Olsen et al. (2021) from ACS MIR observations. This depletion returned to
460 possibly normal equinoctial abundances after the GDS, until in the southern summer
461 another depletion of CO VMR occurred, around $L_S = 270^\circ - 300^\circ$ (uncertain start in
462 our records due to the lack of TGO observations around $L_S = 265^\circ - 280^\circ$). This oc-
463 curred in both hemispheres. Although these two periods share a drop in CO VMR, they
464 may not respond to exactly the same processes. The well known increase in CO₂ den-
465 sity in the southern solstice, during the sublimation of the southern polar cap, (Hess et
466 al., 1979; Forget et al., 2008) is surely behind the low CO observed after $L_S = 280^\circ$.
467 However, another mechanism may be at play during very dusty conditions. During the
468 GDS and its decay phase high water vapor abundance has been reported (A. Fedorova
469 et al., 2021; Villanueva et al., 2021; Alday et al., 2021; Belyaev et al., 2021; Brines et al.,
470 2022). This suggests a depletion of CO due to enhanced H₂O which increases the hy-
471 droxyl radicals (OH).

472 At the lowermost altitudes explored, around 20-30 km, the retrieved CO shows large
473 VMR after the GDS in the NH. This increase seems to disappear through the decay phase,
474 and is not observed around $L_S = 270^\circ$. In the SH, however the SO measurements dur-
475 ing the onset of the GDS coincides with higher latitudes and a high CO abundance is
476 also seen. This may also disappear as the season advances, but this is not confirmed as
477 our data coverage is incomplete at these low altitudes. Let us bring up here that at the
478 lowermost altitudes the CO sensitivity of our inversion is lower, and climatological ef-
479 fects (*a priori*) may play a role.

480 At the end of the southern summer, during $L_S = 335^\circ - 354^\circ$, the CO VMR
481 increases at high latitudes in both hemispheres. This enhancement is due to the down-
482 welling branch of the equinoctial hemispheric Hadley cells (Forget et al., 1999; Neary &

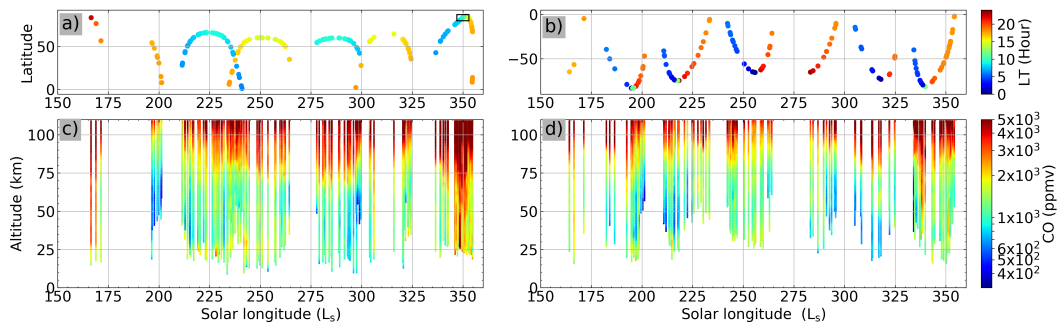


Figure 7. All CO profiles retrieved for this study. Panels a and b show the seasonal and latitudinal distribution of the dataset used, with the colors on scale indicating the localtime. The bottom panels show L_S -altitude distribution of the CO profiles corresponding to the location shown in the top panels. Colors are selected to facilitate inspection of iso-contours, like the orange iso-contour of 2500 ppm, which varies through the seasons (see text for details). A box has been included in panel (a) to indicate the position of the data selected for the study of local time effects (see text)

Daerden, 2018). This enhancement in CO is consistent with a higher column abundance observed in a nadir geometry by the NOMAD LNO channel (Smith et al., 2021).

5.2 Latitudinal Variation

In Figures 8 and 9 we show the latitudinal variation of the CO profiles retrieved from NOMAD-SO and simulated with the PCM, respectively. The CO mixing ratios simulated by the Mars PCM are shown in order to illustrate both the departure from the *a priori* and the predicted dynamical effects on the distribution. According to the PCM simulation, two Hadley cells exist, rising air from the equator and moving to high latitudes in both hemispheres during the two equinoxes, in early northern spring and fall. The ascending branch of the Hadley cells shifts according to the season, as the heating in the subsolar point moves over the planetary surface (Read et al., 2015). During solstices, one of the Hadley branches becomes an intense cross-hemispheric cell, particularly strong during the southern summer solstice ($L_S = 270^\circ - 300^\circ$) (Forget et al., 1999).

Each panel of the Figures 8 and 9 corresponds to a fixed season indicated at the top of each panel. These eight periods include the onset of the dust storm ($L_S = 190^\circ - 205^\circ$), the decaying phase of the GDS ($L_S = 210^\circ - 225^\circ$), the initial phase of the

499 perihelion season ($L_S = 240^\circ - 250^\circ$), the late phase of perihelion ($L_S = 250^\circ -$
500 -265°), and some periods of the southern summer ($L_S = 285^\circ - 300^\circ$, $L_S = 300^\circ -$
501 -315° , $L_S = 316^\circ - 330^\circ$ and $L_S = 331^\circ - 345^\circ$).

502 During the period $L_S = 190^\circ - 205^\circ$ (Figures 8 and 9, panel a), the Martian
503 dynamics is dominated by two Hadley cells increasing the CO mixing ratios at higher
504 latitudes in both northern and southern hemispheres. In particular, the high mixing ra-
505 tios found around 25 km over the southern polar latitudes seem to be the result of such
506 a transport. A similar increase in CO VMR amount over the high northern latitudes has
507 also been found by Olsen et al. (2021) during the equinox period.

508 As the season advances, the CO distributions in panels c, d, e, and f, show vari-
509 ations that can be related to slowly varying solar driven CO_2 photolysis at high altitudes
510 (González-Galindo et al., 2009) and to a Hadley circulation pattern with an ascending
511 flux which is evolving through these periods. The latitudinal gradient in the CO VMR
512 above ~ 70 km vary with the seasons, as can be seen from the changes between pan-
513 els. Around perihelion, the changes between adjacent panels c and d of Figures 8 and
514 9 are too short in time for a significant change in solar isolation. One possibility is that
515 these are due to large scale transport. We cannot exclude also small scale variations, in-
516 cluding measurement noise and thermal effects in the background atmosphere, for which
517 CO VMR is very sensitive. At tropospheric altitudes, there are also changes which seem
518 to be linked to water vapor. We know water vapor presents significant variations in the
519 lower atmosphere, and in particular in these seasons is lower than the amount observed
520 during the GDS (Aoki et al., 2019; A. A. Fedorova et al., 2020; Brines et al., 2022). Un-
521 fortunately, we do not have homogeneous nor symmetric coverage in latitudes between
522 both hemispheres during this key period that includes the perihelion and the southern
523 summer solstice. CO seems to be an excellent tracer to characterize this season but more
524 insightful analysis requires the use of GCM simulations or data assimilation.

525 During $L_S = 285^\circ - 300^\circ$ and $L_S = 300^\circ - 315^\circ$ (panels e and f in Figure 8),
526 a fully developed global inter-hemispheric circulation can be seen in the CO distribution.
527 The latitudinal coverage is patchy and we should rather claim that the distribution ob-
528 served is compatible with such a global cross-hemispheric pattern, more neatly shown
529 with model simulations (Daerden et al., 2019; Olsen et al., 2021). With this caution in
530 mind, the downwelling branch in the NH seems to be at around 60°N , and seems to reach

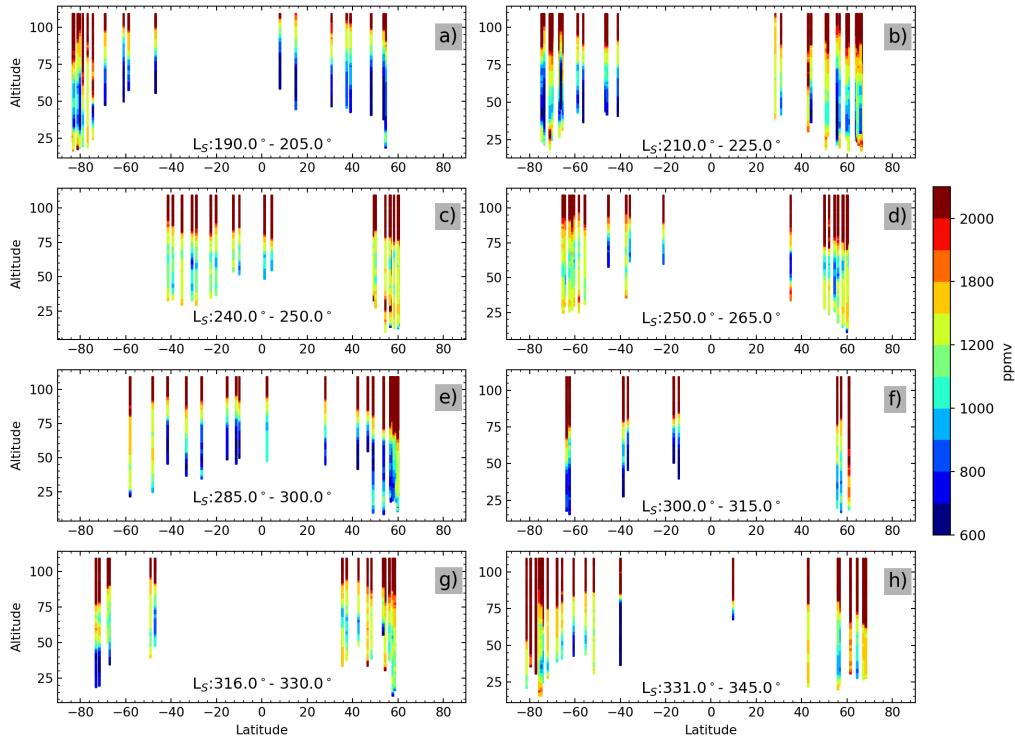


Figure 8. Latitudinal distribution of the retrieved CO profiles after grouping the results in 8 L_s intervals, as indicated in each panel. The color scale indicates the CO VMR values in ppmv.

531 down to lower tropospheric altitudes, 20-25 km, layers which seem to be enriched in CO
 532 compared to lower latitudes. We notice that during this season the distribution from the
 533 PCM shows a lower enrichment in CO, which indicates a weaker downwelling in the model.
 534 Over 60°N , downwelling of the PCM CO seems to reach only around altitude ~ 55 km
 535 increasing the VMR values > 1000 ppm while in the next panel (Figure 9, panel f) the
 536 downwelling seems to increase and reach the altitudes around 40 km and VMR values
 537 around ~ 900 ppm.

538 At low altitudes, the TGO mapping is incomplete but low CO amounts are clearly
 539 observed at mid-low latitudes ($50\text{S}-50\text{N}$) from the beginning of the perihelion season (panel
 540 d in Figure 8). This low abundance is maintained or slightly increased during the south-
 541 ern summer (panels e-f). Later on, at the end of southern summer / northern fall, ob-
 542 servations by TGO at higher latitudes were possible again (panels g and h in Figure 8).
 543 The CO abundance seems to increase there, possibly as a result of dynamics, following
 544 downwelling in both hemispheres. Also, the amount of water vapor is decreasing, spe-
 545 cially in the NH (A. Fedorova et al., 2021), which may add to the CO increase. In the

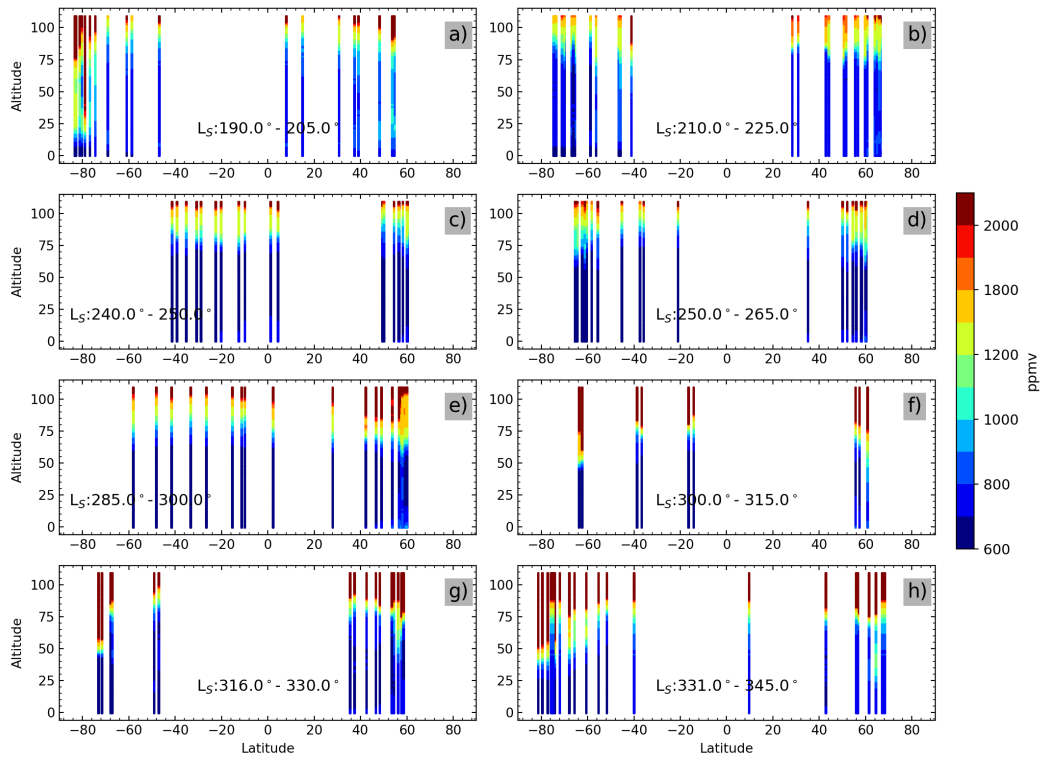


Figure 9. Latitudinal distribution as in Figure 8 but for the PCM CO, using the same set of 8 L_s intervals, as indicated in each panel. The color scale indicates the CO VMR values in ppmv.

546 period shown in panels g and h we observe the largest CO over the North polar region.
547 Such variations of the low altitude carbon monoxide cannot be seen in the PCM sim-
548 ulations (Figure 9). The PCM CO VMR between 20 and 40 km show small variations
549 compared to that observed in the NOMAD SO observations.

550 5.3 Diurnal Variations

551 Given the mixtures of latitudes, seasons, and local times, it is not straightforward
552 to separate them to highlight possible diurnal variations. The L_S spread of the morn-
553 ning (or the evening) observations over NH (see Figure 7 a) are twice as long as the ex-
554 tent of these observations over SH (see Figure 7 b). Also, note that for a given L_S range,
555 NH observations are taken in the morning while SH observations are taken in the evening
556 and vice-versa. We looked for locations where both latitude and L_S have small variations,
557 but only found cases at very high latitudes. Here we examine the retrieved profiles within
558 the box indicated in Figure 7 panel a, which extends in L_S from 347° to 353° and in lat-
559 itudes from 80°N to 86°N . The results are shown in Figure 10. Analysis of diurnal cy-
560 cles at such high latitudes requires cautions, specially during equinox conditions, when
561 the axial tilt is perpendicular to the sun. Trace mixing ratios in polar regions are very
562 affected by global dynamics, hence some variations may not be necessarily diurnal. In
563 addition, there can be an effect due to the L_S variation within our box, because at very
564 high latitudes and close to equinox, an L_S range of only 6° (about a week long) repre-
565 sents significant changes in the illumination conditions.

566 The CO VMR in Figure 10 shows a decrease with local time as well as with L_S be-
567 low about 60 km. However, we cannot confirm this relation based only on this observa-
568 tion. As mentioned above the period of time described by this figure lies at the end of
569 the northern polar night, approaching equinox. Therefore, most likely this variation is
570 rather related to either downwelling within a polar vortex or to variations in the ther-
571 mal structure or a combination of both. The polar vortex is particularly strong during
572 Martian winter conditions but such an isolated region can also be formed near equinoxes
573 (Mitchell et al., 2015; Toigo et al., 2017; Streeter et al., 2021). The decrease in the CO
574 VMR with L_S may also indicate breaking of the polar vortex.

575 In Figure 10 we observe two peculiar profiles whose VMR values are quite large be-
576 low about 45 km. These correspond to the latitudes 80.5°N and 80.9°N and, again, there

577 are a few plausible explanations of this result, although none of them is completely sat-
578 isfactory. One possibility is descending CO-rich air within a displaced polar vortex. How-
579 ever, the two locations might require the vortex to be split, as they are not closely placed,
580 and this is not a common situation in Mars (Mitchell et al., 2015). A second possibil-
581 ity is associated to extremely cold patches, near the CO₂ condensation temperature. In
582 such an instance, there should follow an increase in the relative abundance of CO. For
583 this purpose we have examined the individual temperature and density profiles obtained
584 from the NOMAD data by our team down to about 26 km (López Valverde et al., 2022).
585 They are shown in the right panel of Figure 10. The retrieved temperature below about
586 20-25 km should not be trusted due to low signal (they are merged to the PCM *a pri-*
587 *ori*), but in the altitudes where the CO VMR is high the temperatures are not partic-
588 ularly extreme, a result which certainly does not support this cold-patch hypothesis. Still,
589 this does not completely rule out the possibility of a stronger CO₂ condensation in those
590 two profiles, because such process might happen at lower altitudes, near the Martian sur-
591 face, which cannot be seen in our data. We should turn to a third possibility, which is
592 some unknown bias in the inversion. This is not a fully satisfactory explanation either,
593 since we do not observe any peculiarity in the residuals, nor in the retrieval performance
594 (convergence, AKs) of these two profiles. In our opinion, these peculiar CO VMR pro-
595 files demand more observations at high latitudes, analyzing NOMAD data collected dur-
596 ing more Mars Years, which is an ongoing work in our team.

597 5.4 Comparison with PCM Simulation

598 Figures 11 and 12 show comparisons between the CO distribution from the Mars
599 PCM and our retrieved values. For each NOMAD SO scan, a CO VMR profile was ex-
600 tracted from the model at the exact latitude, longitude, local time, and L_S of the occul-
601 tation path at 50 km altitude, i.e., approximately at the scan's midpoint.

602 Figure 11 shows the same trend in the vertical distribution of CO in both sets, with
603 a relative abundance increasing with altitude. The scans are shown in colors following
604 the solar longitude, but without distinction among latitudes or hemispheres. This per-
605 mits a quick visual realization of the global increase in CO VMR with season at high al-
606 titudes, between L_S = 170° (blue profiles) and L_S = 350° (dark red). However, in the
607 NOMAD VMR, the vertical positive gradient is particularly strong above 60 km, while
608 in the PCM some profiles present steep gradients also at tropospheric altitudes. The re-

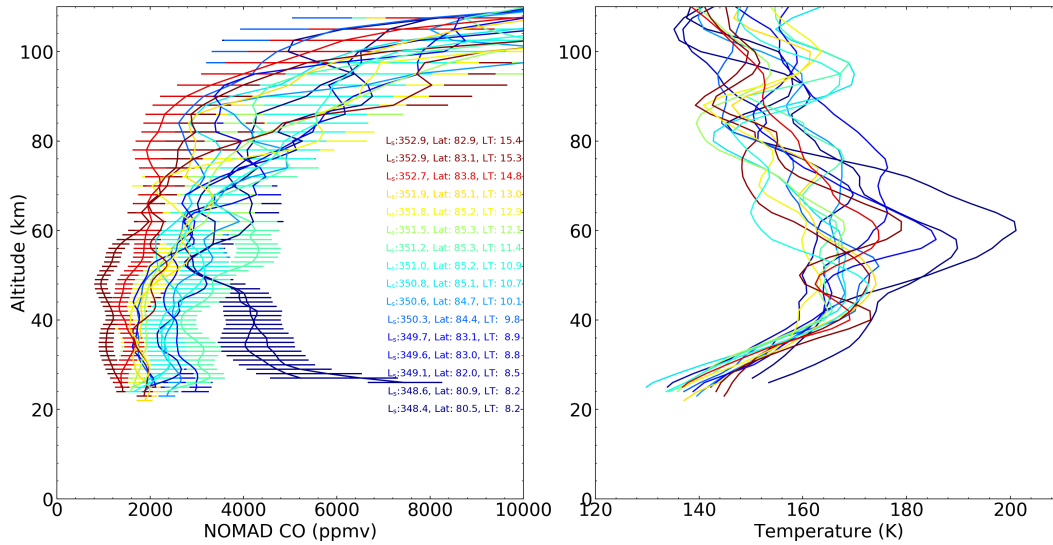


Figure 10. Left panel: variation of CO profiles with local time within a small latitudinal and seasonal box of ranges $\text{Lat} = 80^\circ\text{-}85^\circ$ and $L_S = 347^\circ - 353^\circ$. The color of the curves indicate the local time of the observations. Location and local time are indicated by the in-box text with the same color. Right panel: retrieved temperatures from NOMAD data associated to each of the CO profiles in the left panel. Temperatures below 20-25 km are not trusted due to low signal. See text for details.

609 retrieved profiles show small scale variations consistent with the measurement noise but
 610 also larger oscillations not observed in the PCM data. Similar larger oscillations are also
 611 found in the retrieved temperature profiles (López Valverde et al., 2022) compared to
 612 the PCM values. We think this may indicate larger dynamical activity than in the model
 613 in the form of thermal tides and gravity waves.

614 Further insights into the NOMAD-PCM comparison can be obtained with Figure
 615 12. This shows that the PCM global distribution and our retrievals agree well. Partic-
 616 ularly, the seasonal behavior of the CO VMR above ~ 60 km is similar in both distri-
 617 butions. However, some differences can be seen and are highlighted next. First, the PCM
 618 CO abundances are globally lower than the NOMAD retrievals at all altitudes, especially
 619 in the mesosphere (above 60 km). There are exceptions to this rule, especially at low al-
 620 titudes, which are evident in Figure 11. Secondly, the two equinox periods show signif-
 621 icant differences. During the northern autumn equinox period, $L_S = 160^\circ - 180^\circ$, mix-
 622 ing ratios lower than the PCM are observed at high latitudes. A similar result was ob-
 623 tained by Olsen et al. (2021) from their model-data comparisons. The agreement between

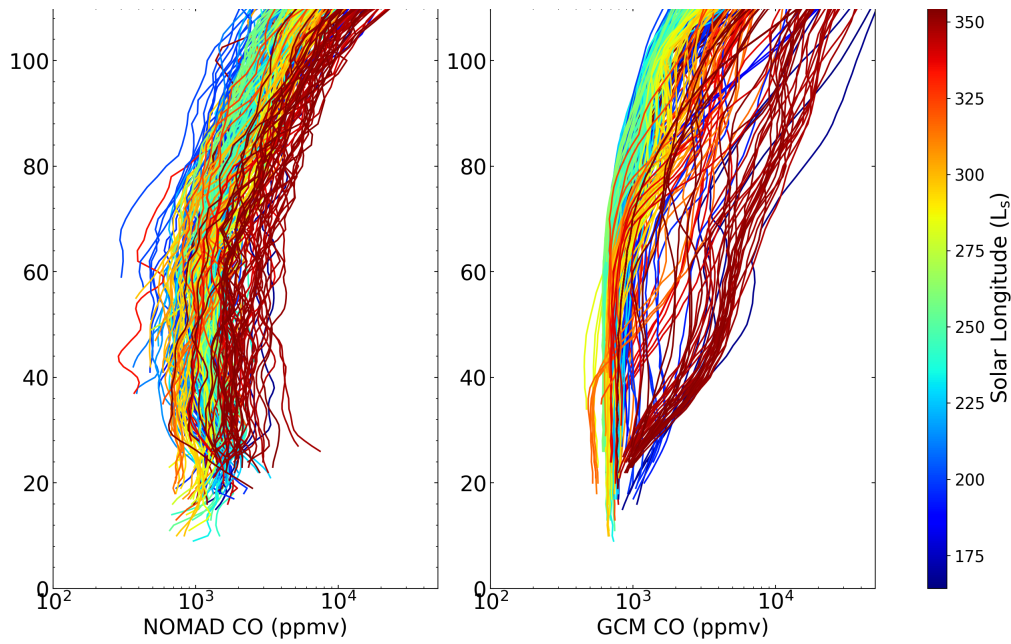


Figure 11. Comparison of the whole set of retrieved NOMAD CO VMR profiles (left panel) with the PCM results (right-hand side panel). The color scale indicates the solar longitude of the scans.

624 model and data seems better at lower-mid latitudes and in both hemispheres, although
 625 not many profiles are available in this period to support this idea. In the other equinox
 626 period, during $L_S = 345^\circ - 354^\circ$, the retrieved CO is lower than the PCM in the NH
 627 by at least a factor of 2 while the opposite is observed in the SH.

628 Among other differences, the PCM maps do not show a depletion of CO during the
 629 onset and decay phases of the GDS as strong as in the NOMAD results. However, in this
 630 period, comparatively a better quantitative agreement is obtained between the *a priori*
 631 and the retrieved CO. The modelled water vapor and temperatures are also in better agree-
 632 ment with the retrieved water vapor (Brines et al., 2022) and the retrieved temperature
 633 (López Valverde et al., 2022). A last significant difference is observed around $L_S = 315^\circ$
 634 at altitudes between 60 and 80 km. This corresponds to southern high latitudes, where
 635 both distributions show a large CO abundance at mesospheric altitudes, driven by an
 636 active global circulation, but it is larger in the PCM by a factor 3-5.

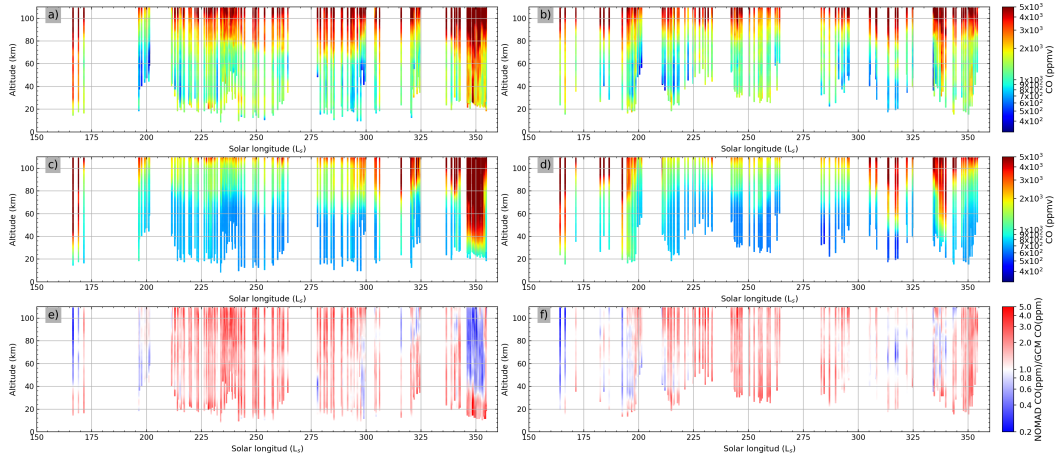


Figure 12. Comparison of PCM CO densities with the retrieved ones. The top panels a and b show the retrieved CO VMR. Panels c and d show the PCM CO mixing ratios and panels e and f show the ratios NOMAD/PCM. Left panels (a, c, and e) correspond to the NH and right panels (b, d, and f) to the SH.

6 Comparison with Previous Results

Figure 13 shows a comparison of our CO vertical profiles with the profiles retrieved from ACS observations (Olsen et al., 2021). The ACS and SO profiles observed within a box of 5° latitude and 5° L_S are chosen for the comparison. From the 32 profiles studied by Olsen et al. (2021), 17 were selected for this comparison. Since the ACS and SO observations are not exactly collocated and they are processed with two very different retrieval schemes, some differences in the CO densities are expected. Profiles in panels b, c, & d show the CO densities during the onset of the GDS. The results from the both instruments compare well although our retrieved values are lower than those from ACS below 80 km. The average of the differences, in the range between the lowest available altitude and 75 km, are 225 and 250 ppm in panel b, 306 ppm in panel c and 308 ppm in panel d. During the high dust loading period, however, the average difference is lower, within 150 - 200 ppm (for panels e to j). The highest average difference can be seen over the northern high latitudes during the equinox and reaches 850 - 1200 ppm (panel a).

A major difference among these profiles is to be noted above 80 km where an increase in VMR is usually observed, due to photochemical production. Our values start to increase above 80 km whereas the ACS values start increasing above 100 km. Let us

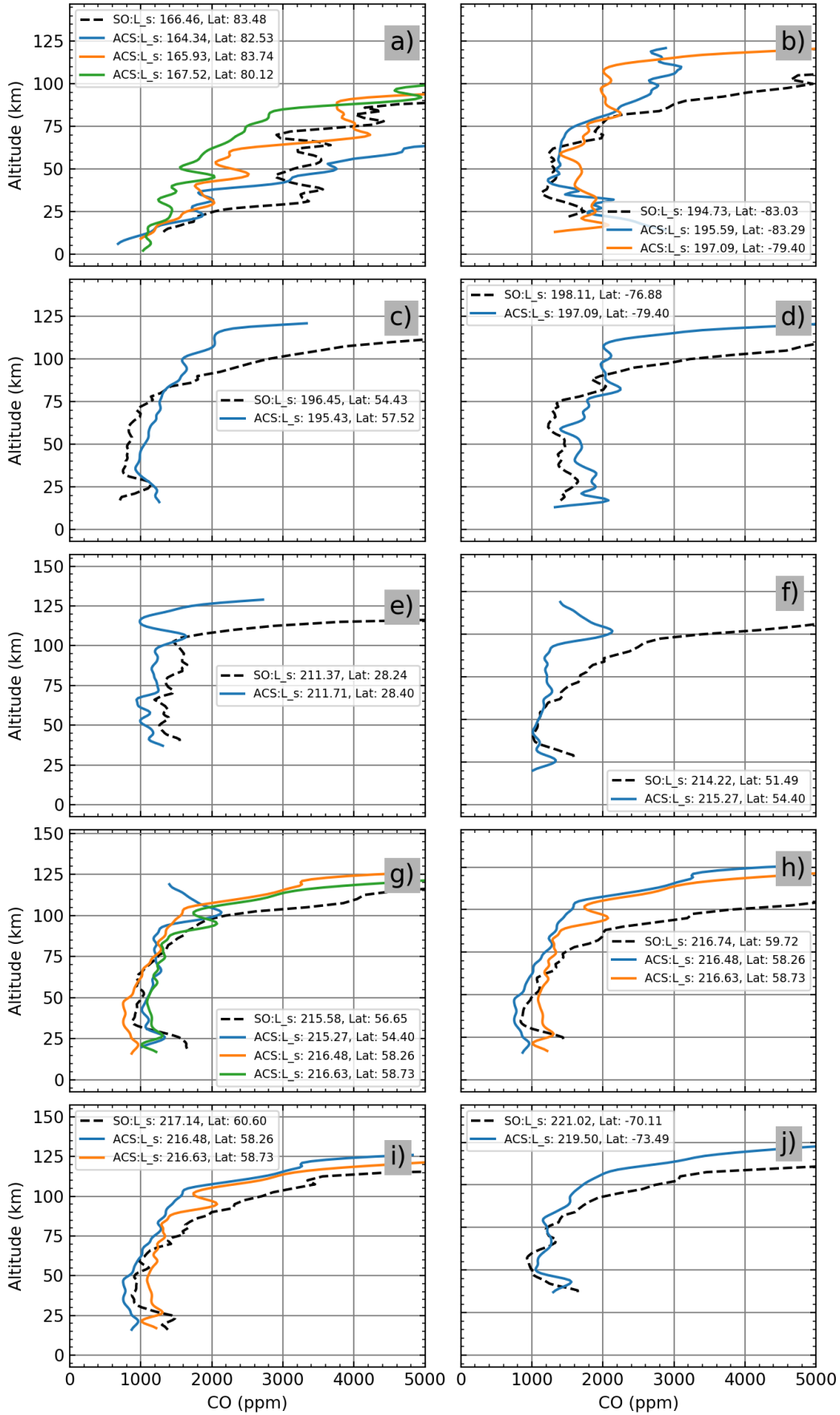


Figure 13. Comparison of the CO profiles in this study with the CO profiles obtained from ACS observations (Olsen et al., 2021). The common profiles within the Lat-LS box of dimension 30° are compared.

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655 recall that at these high altitudes the measurements are particularly noisy, even in the
656 temperature retrievals, and both ACS MIR and NOMAD SO present the largest uncer-
657 tainties. Apart from these differences, and as mentioned above, our CO VMR results com-
658 pare well with the ACS profiles, the differences between them arising most likely from
659 different thermal structures and differences in retrieval methods. This is a first compar-
660 ison intended to illustrate the need of a further, more in-depth, and coordinated com-
661 parison between both instruments.

662 Other instruments from the Mars orbit and ground-based observations have pro-
663 vided column abundances of CO. These columnar mixing ratios are obviously represen-
664 tative of the CO abundance near the planet’s surface. In our retrieved profiles, the low-
665 est altitude of sounding is variable depending on atmospheric dust loading, but is above
666 20 km in most solar occultation scans. The amount of CO in a column below our low-
667 ermost altitude is between 5 and 50 times the amount of CO above. In order to com-
668 pare with previous nadir measurements, instead of CO column mixing ratios, we have
669 calculated mean VMRs of our retrieved CO profiles. These are computed from the low-
670 ermost retrieved altitude up to 60 km, averaging these in the L_S bins of Figure 8 and
671 comprising all the latitudes between 45°N and 45°S . The global average value obtained
672 is 884 ppm. This compares well with LNO and PFS global averages, which are 800 ppm
673 (Smith et al., 2021) and 820 ppm (Bouche et al., 2021) respectively. The global average
674 values obtained from ACS NIR and MIR is 970 ppm (A. Fedorova et al., 2022) which
675 is 9% higher than our values. Therefore, in terms of global averages, our VMRs are com-
676 parable with the previous results. During the GDS, however, we obtained a lower av-
677 erage VMR, of 619 ppm, while the LNO and PFS nadir data do not show this depletion.
678 Both MIR and NIR also found an averaged depletion in this period. In another period
679 that we analyzed, during the southern summer around $L_S = 270^\circ - 300^\circ$, our values
680 are around 900 ppm, which is higher than the column measurements by LNO and CRISM
681 but agree well with NIR and MIR results. Our averaged binned values also seem to agree
682 with NIR and MIR at the end of the MY34, with average VMR around 800-850 ppm.

683 7 Summary and Conclusions

684 For the first time, we have retrieved Martian CO density profiles up to about 100 km
685 tangent altitudes from the NOMAD solar occultation channel for MY 34 between $L_S =$
686 $160^\circ - 354^\circ$ using collocated retrievals of temperatures and pressures. The two main

687 sources of uncertainty in our results come from the variability in the AOTF transfer func-
688 tion and from the temperatures. The retrieval errors are typically lower than 10% be-
689 low 80km which can grow up to 30% above this altitude. Below 80 km the vertical res-
690 olution is usually better than 5 km.

691 The distribution of CO abundances observed by NOMAD globally agrees with lat-
692 itudinal and seasonal variations obtained before with nadir observations, made by space-
693 craft instruments like CRISM (Smith et al., 2009), PFS (Sindoni et al., 2011) and NOMAD-
694 LNO nadir sounding (Smith et al., 2021). Our retrieved CO mixing ratios agree well with
695 the retrieved CO mixing ratios from ACS observations. The agreements are strikingly
696 well during the onset and decay period of the GDS. The agreement with ACS results dur-
697 ing the equinox period over northern high latitudes is also good regarding the sharp in-
698 crease in VMR values with altitude, which was obtained with both the instruments (Fig-
699 ure 13).

700 We find a strong depletion in CO VMRs during the onset of GDS which is more
701 pronounced over the NH. This was also reported by Olsen et al. (2021), who mentioned
702 a possible explanation, related to the amount of H₂O. During the dust storm season, the
703 atmospheric water vapor increases, and therefore the OH radicals, which causes this de-
704 pletion. We think this may indeed be one of the reasons, and below we describe the pe-
705 riod, southern summer, where we also observe links between the H₂O and CO abundances.

706 We observe an enhancement in CO at altitudes around 25 km over the southern
707 hemisphere during the onset of the GDS. We attribute this enhancement to the down-
708 welling branch of the southern Hadley cell (bringing down mesospheric air, enriched in
709 CO, as detailed in section 5.2). However, a different increase in the CO densities is found
710 over the northern hemisphere from low to mid latitudes during the decay phase of the
711 GDS. In contrast to the previous one, this enhancement could rather be dominated by
712 the lack of OH radicals following the decrease of incoming solar radiation.

713 The seasonal behavior of CO profiles reveals that CO VMRs below 60 km decrease
714 during the southern summer all over the globe. This decrease may respond to a com-
715 bination of effects. One of these can be the release of CO₂ and H₂O from the southern
716 polar cap during this season (Forget et al., 2008). Increase of CO VMR at these altitudes
717 is only found at the beginning of southern spring and the end of southern summer.

718 Regarding the latitudinal distribution, a couple of distinct periods can be seen in
719 the data. In particular, the period $L_S = 285^\circ - 300^\circ$ shows the impact of the global Hadley
720 circulation. The descending branch of the global Hadley cell lies over 60°N , while the
721 ascending branch is seen in the SH around 40°S . This describes well the known dynam-
722 ical global pattern on Mars during the southern summer solstice $L_S = 270^\circ - 300^\circ$
723 (Forget et al., 1999; Read et al., 2015; Neary et al., 2020).

724 Regarding the comparison with the Mars PCM (our *a priori*) the simulated CO
725 VMRs agree qualitatively with the retrieved CO. A good quantitative agreement between
726 the simulated and the observed CO VMRs is found during the onset of the GDS and dur-
727 ing the period around $L_S = 300^\circ$ when the modelled water vapor and temperature are
728 also in better agreement with the retrieved values (López Valverde et al., 2022; Brines
729 et al., 2022). Nonetheless, there are some significant differences. Among the differences
730 with the PCM simulations, our retrieved CO VMR seems to be lower than the model
731 prediction during the beginning of the northern spring over NH. In this season the re-
732 trieved CO VMRs are larger over the SH. In general, the retrieved VMRs are higher than
733 the simulated values during $L_S = 210^\circ - 290^\circ$. An important difference was found in
734 the comparison of the latitudinal distributions of CO (Figures 8 and 9), is the presence
735 of a weaker south-to-north transport in the model during the southern summer. The NO-
736 MAD SO profiles (Figure 8, panel e) show a larger enrichment of CO mixing ratios over
737 the northern high latitude surely due to a stronger global Hadley cell.

738 We plan to extend our retrieval approach to the whole NOMAD dataset in order
739 to study CO distribution for a couple of complete Martian years, to compare the CO mix-
740 ing ratios between years with and without global dust storm, and to gain further insight
741 into seasonal and latitudinal variations in a more complete coverage. This dataset can
742 also be very valuable for PCM modeling and validation purposes. Detailed comparisons
743 with ACS observations of CO are also foreseen.

744 **Appendix A AOTF Characterization**

745 The NOMAD SO measurements are acquired by selecting different diffraction or-
746 ders, and each datafile corresponds to one order. This is because the AOTF ideally se-
747 lects one order at a time for diffraction through the grating, and we call it the main diffrac-
748 tion order during that observation. The tuning of the AOTF for whichever main diffrac-

749 tion order is set through a radio-frequency input. A typical AOTF tuning relation with
750 wavenumber is given in Neefs et al. (2015). For example, for a radio frequency of 25MHz,
751 the diffraction order 190 is selected. The diffraction efficiency of the AOTF is charac-
752 terized by a function over the permitted bandpass and is called AOTF transfer function
753 (I. Thomas et al., 2016; Liuzzi et al., 2019). The precise functional of the AOTF is very
754 important to characterize the measurement, and it has been the subject of a recent re-
755 vision (Villanueva et al., 2022), which proposed an asymmetric sinc-squared function
756 characterized by four parameters. The functional and the parameters were determined
757 using what are called mini-scans, i.e., a specific sun observation campaign devoted to fine-
758 tune the channel calibration. As a result of that study, nominal values for those param-
759 eters were proposed for each of the NOMAD SO diffraction orders. However, the em-
760 pirical characterization of the parameters is not perfect, and the uncertainties are par-
761 ticularly large in the spectral region of the CO orders 186-191.

762 For this reason, CO is one of the hardest targets of the NOMAD SO channel, and
763 it requires a good fine-tuning of its AOTF. In order to gain some insight into this key
764 analysis, we need to analyze further some of the AOTF parameters.

765 The sinc square AOTF function basically consists of a series of decreasing sidelobes
766 around the main peak, this one laying in the main order while the sidelobes peak in the
767 adjacent orders. The magnitude and shape of the sidelobes is parameterized with the
768 help of an "asymmetry factor" (AF) and a "sidelobes ratio" (SR). As its name suggests,
769 a larger sidelobes ratio increases the relative importance of the sidelobes and decreases
770 the AOTF response in the central lobe, i.e., in the main order. On the other hand, a large
771 asymmetry factor decreases the left lobes and introduces asymmetry between the lobes
772 on both sides. For the detailed discussion on the AOTF parameters, we refer to Villanueva
773 et al. (2022). Our AOTF also allows for a possible non-zero baseline component, which
774 is parameterized as a shallow and wide Gaussian function, which in the spectral region
775 of the CO orders is about 70.71 cm^{-1} wide and has a peak (referred to as Gaussian off-
776 set, or GOS hereafter) of 0.2132, for a normalized AOTF in the central lobe.

777 An extensive analysis using retrievals of full CO profiles was performed in order
778 to fine-tune these parameters, following a perturbation study and looking for a minimiza-
779 tion of the retrievals' residuals. For the evaluation of the residuals, we use the goodness
780 of a fit as defined by a χ^2 value (Eq. 3 in Section 4). In addition, the accurate determi-
781 nation of the AOTF parameters should result in similar carbon monoxide densities for

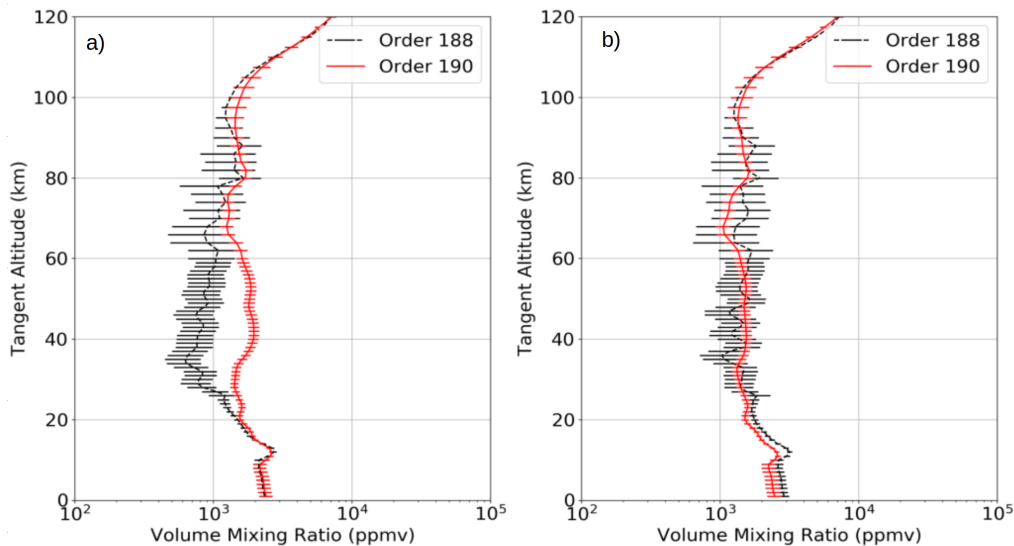


Figure A1. Agreement between the CO profiles from orders 188 and 190 before (panel a) and after (panel b) the fine-tuning of the AOTF parameters for scan *20180907_014655_1p0a_SO_A-I*. In these retrievals a dust filter of opacity 0.5 has been applied which corresponds to an altitude ~ 18 km. This implies no use of measurement below the altitude.

different CO orders due to a more realistic contribution to the CO absorption lines from different orders.

Figure A1 (panel a) shows an example of a CO profile retrieved for one particular scan (*20180907_014655_1p0a_SO_A-I* taken at around 58N and Ls 245) where two different diffraction orders were measured at the same time, orders 188 and 190. In this retrieval, the nominal AOTF parameters were used. The overall agreement between these two CO profiles in the altitude region between 20 and 80 km is not very satisfactory.

In order to make the fine-tuning simple, we started with one of the parameters, in particular the Gaussian offset, or GOS. We chose only the central part of the CO orders, dominated by a relatively small number of CO absorption lines. In the center of a given order, the contribution from that main order is largest compared to other orders' contributions, and the GOS parameter plays the most important role in determining the total contribution (Villanueva et al., 2022). We also selected one scan with low to moderate measurement noise and confined our retrievals to altitudes with low dust opacity (*20180907_014655_1p0a_SO_A-I*, mentioned above). In this scan, a slant optical depth lower than 0.5 means altitudes above 18 km, approximately. We chose a set of values around

798 the nominal value $GOS=0.2132$ proposed by Villanueva et al. (2022) for order 190. Fig-
799 ure A2 shows a comparison among the vertical CO profiles obtained with six different
800 GOS values. The CO VMR in the region between 30 and 80 km is very sensitive to this
801 parameter. The lowest χ^2 was found for values within 0.1 and 0.2. A similar exercise is
802 repeated for order 188. Values of 0.18 for order 188 and 0.2 for order 190 were finally
803 selected as our best Gaussian offsets.

804 As for the other parameters namely, the asymmetry factor (AF) and the sidelobe
805 ratio (SR), another extensive set of retrievals was performed, once the GOS values were
806 fixed. A table of the χ^2 values obtained from their best fit residuals at the end of the
807 inversions are presented in Figure A3. Similar analysis has been performed to fine-tuning
808 other NOMAD SO orders by our team (see companion papers in this special issue). The
809 closer the χ^2 value to unity the better the fitting of the retrieval (Jurado Navarro et al.,
810 2016). In Figure A3 the region of this parameter space with best χ^2 values, shown in dark
811 blue, occupies a broad diagonal. The values finally selected for this study lie in this di-
812 agonal correspond to $SR=2.41$ and $AF=2.31$.

813 Back to Figure A1, the CO VMR obtained with the fine-tuned parameters described
814 here are shown in panel b. They are much closer than with the nominal values from Villanueva
815 et al. (2022) and the differences lie within the errors of the retrieval. We recommend these
816 values and AOTF for any future retrievals of CO from these NOMAD SO orders.

817 Appendix B Synthetic Retrievals

818 Synthetic retrievals represent an excellent exercise to analyze the performance of
819 a given retrieval setup and assumptions, like regularization and dependence on a priori
820 climatologies. In this study, synthetic spectra were generated using our forward model
821 KOPRA, and are intended to supply spectra equivalent to the Level 1 calibrated trans-
822 mittances provided by the NOMAD PI team, but we used a given set of AOTF param-
823 eters, therefore avoiding uncertainties from the actual instrument's AOTF. For the same
824 reason, these spectra are free from bending effects and from spectral shifts due to inac-
825 curacies during calibration (see Section 3). The atmospheric pressure and temperature,
826 and the *a priori* CO were obtained from the Mars PCM model. The KOPRA simula-
827 tions correspond to the precise altitudes of a given scan in order 190. A random noise
828 component with a standard deviation given by the measurement noise in the calibrated
829 datafile was added to the KOPRA simulations.

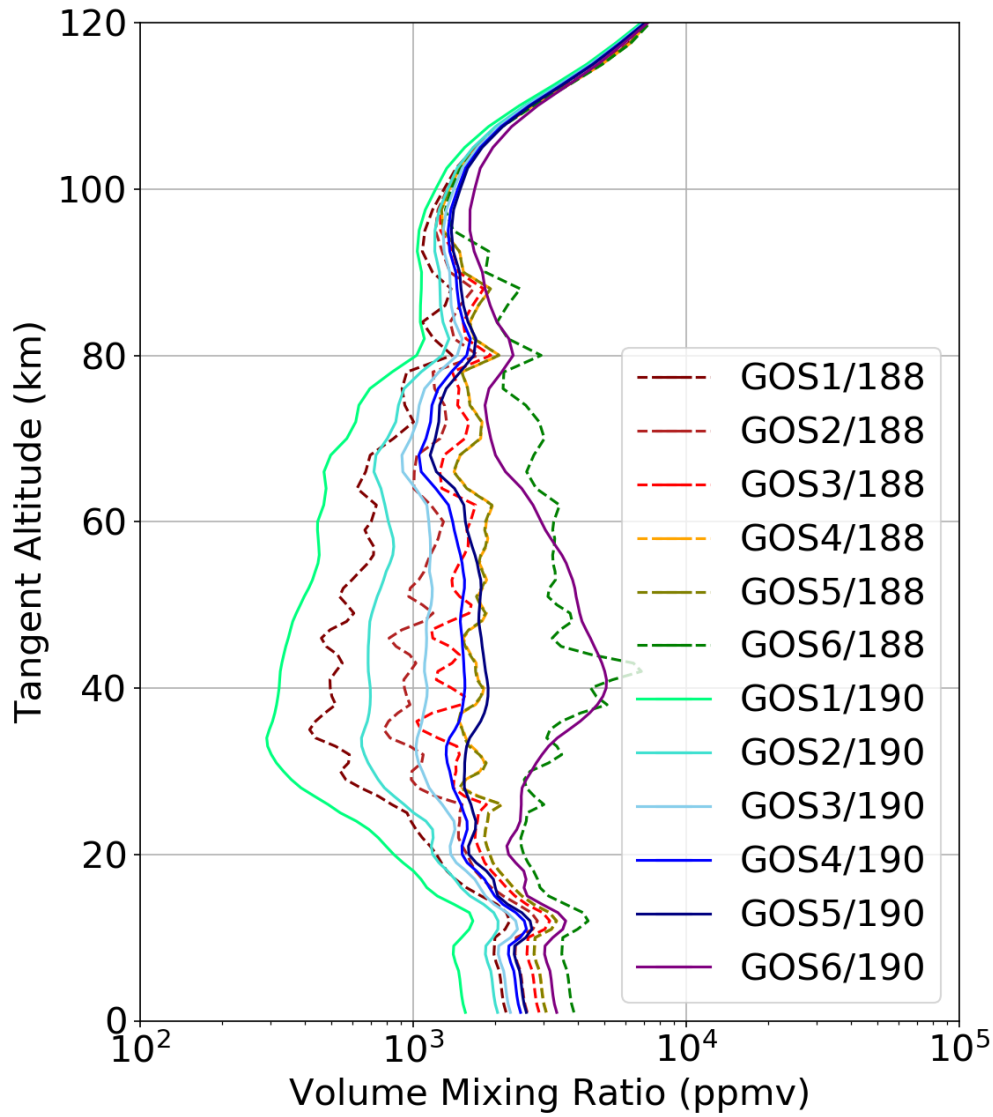


Figure A2. Response of the retrieved CO profiles to variations in the Gaussian offset.

GOS1,GOS2,GOS3,GOS4,GOS5, and, GOS6 correspond to values 0.1, 0.15, 0.18, 0.2, 0.3, and 0.35 respectively. The nominal GOS is 0.2132 for order 190. See text for details.

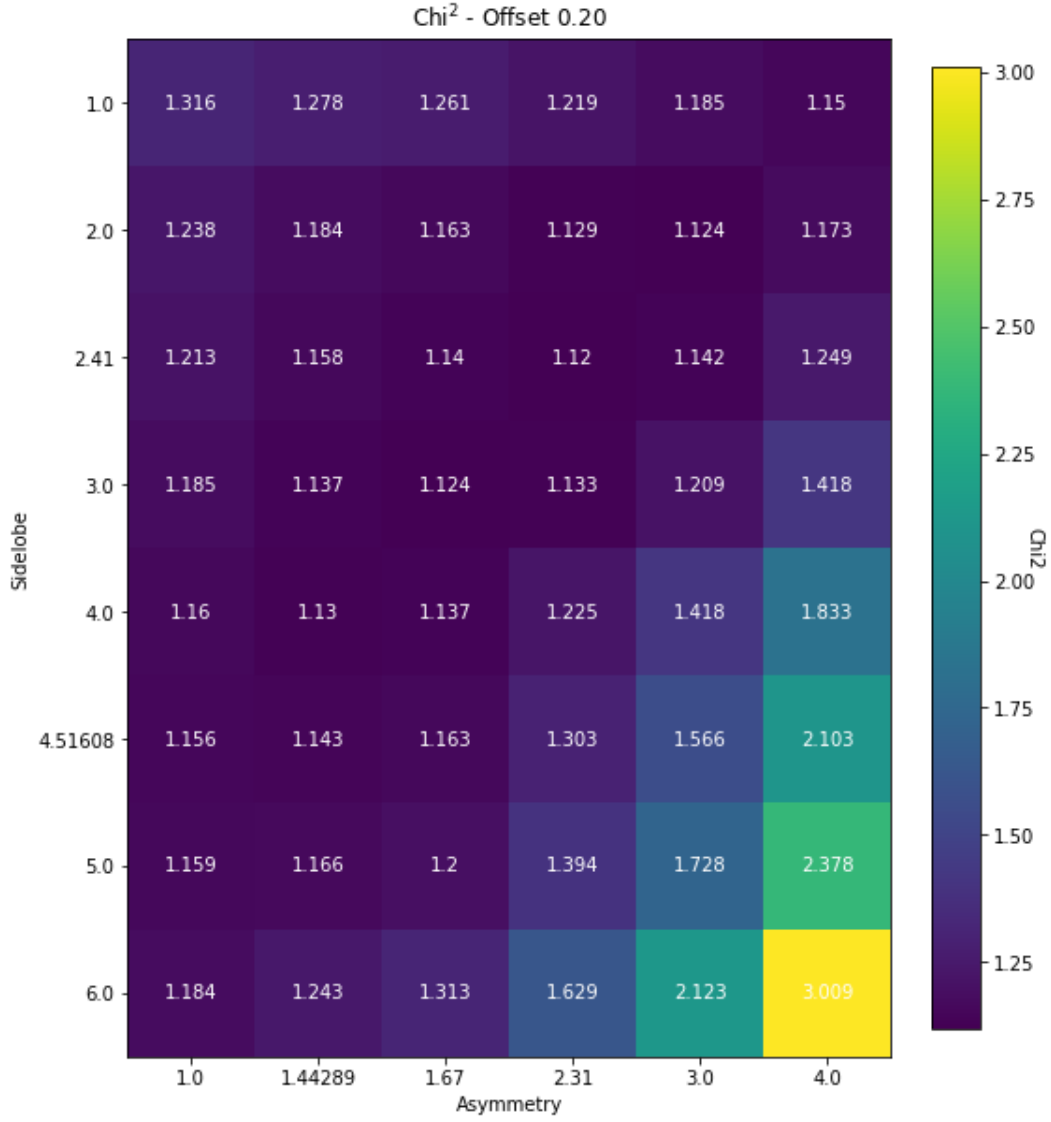


Figure A3. The χ^2 values tabulated for different values of sidelobe ratios and asymmetry factors. The color in the table indicate the values of the χ^2 . The lowest χ^2 is obtained for a sidelobe ratio 2.41 and asymmetry factor 2.31.

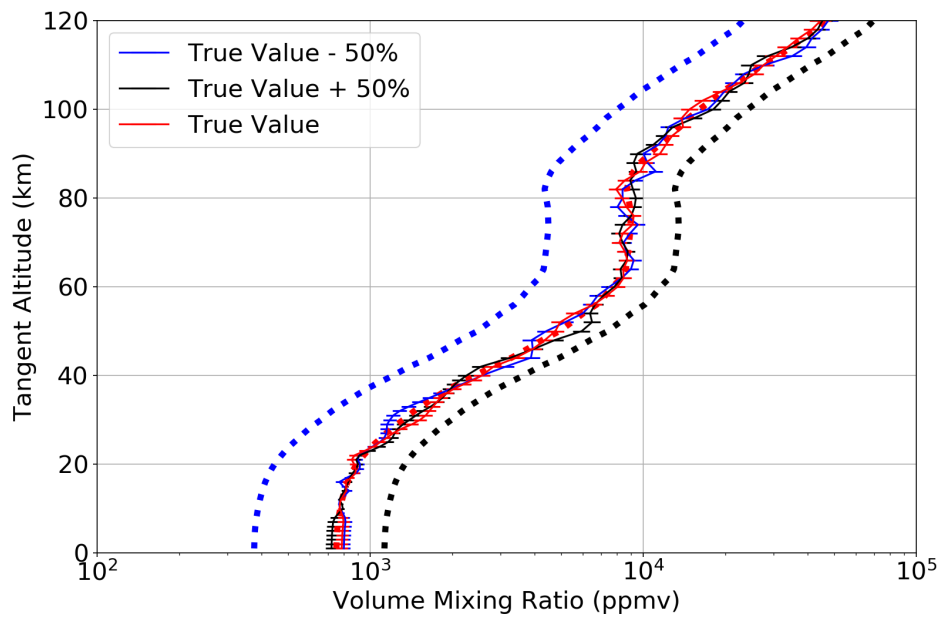


Figure B1. CO profiles retrieved from the synthetic transmittance generated with true value of CO. Colored solid line with errorbar is the retrieved CO with an *a priori* indicated by the dotted line with same color.

830 In this section, we present a study devoted to test the dependence on the *a priori*
831 CO densities. In Figure B1 we show three CO profiles, from retrievals performed with
832 three different *a priori* CO profiles: (i) the nominal VMR from the GCM, which also rep-
833 represents the "true profile", (ii) the nominal profile reduced by 50% at all altitudes, and
834 (iii) the nominal profile increased by 50% at all altitudes. In the three exercises, the re-
835 trieved CO densities essentially coincide among them and with the true profile, with de-
836 viations lower than the retrieval errors. Thus, in these conditions, our retrieval scheme
837 gives correct and robust results against *a priori* assumptions.

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856 **Open Research/Data availability**

857 The HDF5 datafiles of NOMAD SO Level 1a transmittances used in this study are
858 available at European Space Agency (ESA) planetary archive (<https://archives.esac.esa.int/psa>),
859 and also at the NOMAD data center (<https://nomad.aeronomie.be/index.php/data>, (I. R. Thomas
860 et al., 2022; Vandaele et al., 2019))

861 The retrieved results, presented in this work are archived at the zenodo repository
862 and can be accessed from Ashimananda Modak (2022).

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