The climatology of carbon monoxide on Mars as observed by NOMAD nadir-geometry observations

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1 Abstract

- 2 More than a full Martian year of observations have now been made by the Nadir
- 3 Occultation for MArs Discovery (NOMAD) instrument suite on-board the ExoMars
- 4 Trace Gas Orbiter. Radiative transfer modeling of NOMAD observations taken in the
- 5 nadir geometry enable the seasonal and global-scale variations of carbon monoxide gas in
- 6 the Martian atmosphere to be characterized. These retrievals show the column-averaged
- 7 volume mixing ratio of carbon monoxide to be about 800 ppmv, with significant
- 8 variations at high latitudes caused by the condensation and sublimation of the background
- 9 CO₂ gas. Near summer solstice in each hemisphere, the CO volume mixing ratio falls to
- 10 400 ppmv in the south and 600 ppmv in the north. At low latitudes, carbon monoxide
- 11 volume mixing ratio inversely follows the annual cycle of surface pressure. Comparison
- 12 of our retrieved CO volume mixing ratio against that computed by the GEM-Mars
- 13 general circulation model reveals a good match in their respective seasonal and spatial
- 14 trends, and can provide insight into the physical processes that control the distribution of
- 15 CO gas in the current Martian atmosphere.

16 **1. Introduction**

- 17 Carbon monoxide is a minor constituent in the Martian atmosphere, but it plays an
- 18 important role in the photochemical cycle of CO₂ production and loss, and it serves as an
- 19 important tracer for atmospheric circulation patterns (see Lefèvre and Krasnopolsky,
- 20 2017 and references therein for a review). As a non-condensable gas, the volume mixing
- 21 ratio (vmr) of CO is expected to have seasonal and spatial variations on a global and
- 22 seasonal scale (e.g., Forget et al., 2008; Daerden et al., 2019). Therefore, the seasonal and
- 23 spatial variation of CO vmr provides a key observational constraint for understanding the
- 24 dynamical and photochemical processes that operate in the current Martian atmosphere.
- 25 Since its initial detection by Kaplan et al. (1969) using high spectral resolution infrared
- 26 spectroscopy, carbon monoxide has been observed in the Martian atmosphere by a
- 27 number of ground-based observers at infrared, millimeter, and sub-millimeter
- 28 wavelengths (e.g., Clancy et al., 1983; 1990; Lellouch et al., 1991; Krasnopolsky, 2003;
- 29 2007; Moreno et al., 2009). These observations had limited seasonal and latitudinal
- 30 coverage, but were able to establish an average CO vmr near 800 ppmv and to identify
- 31 some latitudinal gradients.
- 32 Spacecraft observations have provided more information in greater detail about the
- 33 seasonal and spatial distribution of CO vmr. Retrievals using observations from the
- 34 OMEGA (Encrenaz et al., 2006) and PFS (Billebaud et al., 2009; Sindoni et al., 2011)
- 35 instruments on-board the Mars Express spacecraft explored the variation of CO vmr for
- 36 certain locations and seasons, while a more complete climatology of CO vmr was
- 37 characterized using near-infrared observations made by the CRISM instrument on-board

- 38 the Mars Reconnaissance Orbiter (Smith et al., 2009; 2018) and using new retrievals
- 39 from the PFS observations (Bouche et al., 2021). These works confirmed a global
- 40 average CO vmr of about 800 ppmv and revealed the seasonal and spatial patterns
- 41 expected for a non-condensable gas, including relatively low CO vmr at the summertime
- 42 poles and an anticorrelation with the annual cycle of surface pressure at low latitudes.
- 43 Spacecraft observations of CO have been combined with general circulation models
- 44 through data assimilation to demonstrate the capability of models in forecasting the
- 45 atmospheric state (Holmes et al., 2019).
- 46 New near-infrared spectra taken in the nadir geometry by the Nadir Occultation for MArs
- 47 Discovery (NOMAD) instrument suite (Vandaele et al., 2015; 2018) on-board the
- 48 ExoMars Trace Gas Orbiter contain the clear spectral signature of multiple CO
- 49 absorption lines and enable the further characterization and refinement of the CO
- 50 climatology observed by previous missions. In this paper we present a new climatology
- 51 of CO vmr based on radiative transfer modeling of the NOMAD nadir-geometry
- 52 observations of CO. These retrievals provide an independent dataset for comparison
- against the results found by CRISM and PFS, as well as new information about possible
- 54 diurnal variations. We also compare our retrievals of CO vmr against the results
- 55 computed using the GEM-Mars general circulation model (Daerden et al., 2015; Neary
- and Daerden, 2018; Daerden et al., 2019) to put our results into context and to identify
- 57 the physical processes responsible for the observed trends in the CO climatology.
- 58 In Section 2, we describe the NOMAD instrument and the observations used in this
- 59 study. In Section 3 we provide details about the retrieval algorithm including the
- 60 assumptions used and the expected uncertainties in the retrieved quantities. The retrieval
- 61 results are discussed in Section 4, and in Section 5 we compare the retrieved values
- 62 against the output from the GEM-Mars general circulation model. Finally, we summarize
- 63 our findings in Section 6.

64 **2. Data Set**

65 2.1. NOMAD Instrument

- 66 The NOMAD instrument suite was selected for the ExoMars Trace Gas Orbiter mission
- 67 in order to provide a spectroscopic survey of the Martian atmosphere that would advance
- our knowledge of the composition of the Mars atmosphere (Vandaele et al., 2015; 2018).
- 69 The first results from the analysis of NOMAD observations have already been used to
- 70 (among other things) set new stringent upper limits on methane abundance (Korablev et
- al., 2019) and to evaluate the impact of Mars global dust storms on the D/H ratio
- 72 (Vandaele et al., 2019) and the vertical distribution of water vapor (Aoki et al., 2019).
- 73 NOMAD consists of three different spectrometers that cover large portions of the
- visible, and near-infrared spectral ranges from 0.2 to 4.3 μ m. The UVIS

- 75 (Ultraviolet and Visible Spectrometer) covers the spectral range from 200 to 650 nm
- 76 (Patel et al., 2017), while the SO (Solar Occultation) spectrometer is dedicated to solar
- occultation observations at near-infrared wavelengths $(2.3-4.3 \mu m)$. In this study, we use
- the spectra taken by the LNO (Limb Nadir and solar Occultation) spectrometer, which
- can be used in both nadir-viewing and limb-viewing geometries, as well as for solar
- 80 occultations.
- 81 The LNO spectrometer is a modified version of the SOIR (Solar Occultation in the IR)
- 82 instrument (Nevejans et al., 2006) that was flown on Venus Express (Titov et al., 2006).
- 83 It uses an echelle grating in combination with an Acousto-Optical Tunable Filter (AOTF)
- to sample the spectral range from 2.3–3.8 μm at a spectral resolving power of roughly
- 85 10,000 (~0.4 cm⁻¹). For each LNO observation, 320 spectral channels cover a spectral
- ⁸⁶ range between 20 and 35 cm⁻¹ depending on the diffraction order. Each order is
- 87 characterized by a specific central wavelength, which is selected by the AOTF. A couple
- 88 dozen different grating orders are used regularly to monitor different atmospheric
- 89 constituents, including carbon monoxide.

90 2.2. Observations Used for this Study

- 91 For this work we use all NOMAD LNO observations taken using grating orders 189 and
- 92 190 (4250–4300 cm⁻¹, or 2325–2350 nm), which are used specifically to monitor carbon
- 93 monoxide. Although solar occultation observations have greater sensitivity and enable
- 94 the vertical distribution to be retrieved, the nadir-geometry observations used here have
- 95 the advantage of higher horizontal spatial resolution and much more complete seasonal
- and (horizontal) spatial coverage. Where available, the LNO observations consist of
- 97 north-south strips across the entire visible disk of Mars containing up to a couple hundred
- 98 individual spectra for each orbit. The projected instantaneous LNO footprint on the
- 99 surface of Mars is approximately 0.5 x 17 km (Vandaele et al., 2015), and observations
- are typically spaced every $\sim 0.7^{\circ}$ of latitude along the orbit track. At the near-IR
- 101 wavelengths used here we depend on reflected solar light for our signal and the
- 102 contribution from thermal radiation is negligible. Therefore, we cannot retrieve carbon
- 103 monoxide for nighttime or winter polar regions, and in addition, observations with solar
- 104 incidence angle greater than 55° are not used because of their low signal-to-noise.
- 105 The NOMAD LNO observations used in this study cover the time period between Mars
- 106 Year (MY) 34, $L_s=150^{\circ}$ and MY 35, $L_s=241^{\circ}$ (28 March 2018 and 18 July 2020). This
- 107 time period covers more than a complete Mars Year, with overlapping coverage between
- 108 two Mars Years during the peak dustiest season when some observations were not used
- 109 because of excessive atmospheric dust loading (see Section 3.4).
- 110 Figure 1 shows the coverage of the 109,159 retrievals used in this work as a function of
- 111 season (L_s) and latitude. During each period of time when nadir observations are possible

112 (each lobe in Figure 1) the local time of the observations changes systematically from late

afternoon to early morning. The solar incidence angle is lowest in the center and

114 increases outward to the edge of each lobe. Observations were taken sufficiently often to

enable a good characterization of the seasonal and global-scale trends in carbon

116 monoxide volume mixing ratio, but are not capable of providing maps on short timescales

117 on the order of days or weeks. Observations using both NOMAD grating orders 189 and

118 190 were taken throughout the time period shown in Fig. 1.

119 A typical set of spectra taken using order 190 on one orbit is shown in Fig. 2. In this

120 image each spectrum is represented by a horizontal row in the image with color

indicating the observed signal. The quantity that is shown here and that is used in this

analysis is the "Reflectance Factor" field provided by the nominal NOMAD data pipeline

123 processing (further detail about the calibration of NOMAD data can be found in Liuzzi et

al., 2019; Thomas et al., 2020). This divides the observed radiance of Mars by the

125 observed solar reference spectrum with corrections for the Mars-Sun distance and mean

126 solar incidence angle. The wavelength of each NOMAD channel depends on the

127 instrument temperature at the time of each measurement (e.g., Liuzzi et al., 2019), so a

128 wavelength correction is performed for each individual observation as a part of the

retrieval by fitting the observed lines to their known wavelengths. The individual

130 absorption lines caused by CO are readily apparent as vertical bands in Fig. 2 (for

131 example, at 4285.0, 4288.3, and 4291.5 cm⁻¹).

132 **3. Retrieval Algorithm**

133 We use seven lines for the retrieval of carbon monoxide using NOMAD LNO

134 observations from the (2-0) ro-vibrational band, with four lines from order 189 (R0, R1,

135 R2, and R3) and three from order 190 (R6, R7, and R8). As shown in Fig. 3., the CO

136 absorption lines in this spectral region are very well separated from each other and from

137 any other significant absorptions from water vapor or CO₂. At the spectral resolution of

138 the LNO observations, the absorptions have a depth that is typically at a level of 5-10%

139 of the continuum level, which is more than sufficient for a reliable retrieval. These CO

140 lines are part of the same absorption band used for retrievals of CO by the CRISM

141 instrument (Smith et al., 2009; 2018).

142 **3.1. Radiative Transfer and Assumptions**

143 The radiative transfer modeling used here for computing synthetic spectra of carbon

144 monoxide is essentially the same as that used in our previous retrievals using CRISM

spectra (Smith et al. 2009; 2018). To perform the retrieval, radiative transfer modeling is

146 used to compute an expected spectrum for a given volume mixing ratio of carbon

147 monoxide, and that CO vmr is then varied until the resulting integrated line depth of the

148 CO absorptions in the computed spectrum matches that from the observed NOMAD LNO149 spectrum.

150 The radiative transfer is modeled using the discrete ordinates approach (e.g., Goody,

151 1989; Thomas and Stamnes, 1999), which explicitly includes multiple scattering by

aerosols. Absorption of carbon monoxide gas is computed using the correlated-k

approximation (Lacis and Oinas, 1991), using the latest version of the HITRAN

154 spectroscopic database for line parameters, which now include line broadening from

155 carbon dioxide (Gordon et al., 2017). The viewing geometry, including the solar

156 incidence angle, the emergence angle, and phase angle are read from spacecraft records

157 and are assumed to be known quantities.

158 The thermal state of the atmosphere for each observation is provided by the OpenMARS

159 database, which is a reanalysis product that combines spacecraft observations with a

160 Mars General Circulation Model (Holmes et al., 2020). Thus, the temperature profiles

161 read from OpenMARS and used in this work include the effects of the global dust storm

162 that occurred during Mars Year 34 (e.g., Guzewich et al., 2018; Smith, 2019). Surface

163 pressure is taken from the Mars Climate Database v.5.3 (Forget et al., 1999; Millour et

164 al., 2018) using its high-resolution setting to resolve sub-grid topography. Surface albedo

- 165 is taken from a map based on Thermal Emission Spectrometer observations (Christensen
- 166 et al., 2001).

167 Scattering from dust and water ice aerosols affects the observed depth of gas absorptions

and must be included in the model for an accurate retrieval. The optical depth of dust and

169 water ice aerosols for each NOMAD LNO spectrum is estimated from concurrent

170 observations by the THEMIS instrument on Mars Odyssey (Smith, 2018; 2019). The

171 scattering properties of dust and water ice aerosols are taken from the analysis of CRISM

172 near-infrared observations (Wolff et al., 2009), while the aerosol particle size (effective

173 radius of 1.5 μ m for dust and 2.0 μ m for water ice) is an average value from the analysis

174 of many previous spacecraft observations (e.g., Wolff and Clancy, 2003; Clancy et al.,

175 2003; Wolff et al. 2006; 2009; Vincente-Retortillo, 2017). The dust aerosol is assumed

176 here to be well-mixed with the background atmosphere. Water ice aerosol is assumed to 177 form clouds above the water condensation level, with no cloud below and a well-mixed

178 cloud above. Discussion of the uncertainties related to the above assumptions is presented

179 in Section 3.4.

180 **3.2. Retrieval Algorithm Process**

181 Figure 4 shows representative averaged NOMAD LNO spectra for the seven lines chosen

182 for the retrieval. The center locations for these lines for order 189 are: 4263.8, 4267.5,

- 183 4271.2, and 4274.7 cm⁻¹, and for order 190 are: 4285.0, 4288.3, and 4291.5 cm⁻¹. For
- 184 each line, we divide the observed reflectance factor by the continuum level and compute

185 the total integrated line depth for the line. The integrated line depth retains all the

- 186 information from the line since these lines are spectrally unresolved in the observations.
- 187 The observation to be fit is then the sum of the integrated line depths for all of the lines in
- 188 each spectrum.
- 189 The retrieved value of CO volume mixing ratio is determined by computing synthetic
- 190 spectra with a given CO vmr and computing the integrated line depth for the three or four
- 191 lines in the computed spectrum (with the continuum divided out) in the same way as done
- 192 for the observation. In the computation, CO is assumed to be well-mixed vertically,
- 193 which is supported by previous studies (e.g., Smith et al., 2019) and by model results for
- 194 the lower part of the atmosphere where most of the CO molecules lie (e.g., Daerden et al.,
- 195 2019). The final retrieved value of CO vmr is simply that value where the computed
- 196 integrated line depth matches the observed value.
- 197 Using this algorithm makes the retrieval robust to uncertainties in the instrumental
- 198 spectral response function and to errors or uncertainties in the overall calibration of the
- 199 observed reflectance factor. The drawback of using integrated line depth as a metric is its
- 200 sensitivity to the choice of the continuum. To minimize this sensitivity, we define the
- 201 continuum as being linear in reflectance factor and computed using spectral channels that
- are the same constant distance (plus and minus 6 LNO channels, or about ± 0.64 cm⁻¹)
- from the line center for each line. These computed continua are shown in Fig. 4 as red line segments
- 204 line segments.
- Along with the retrieved value of CO vmr, for each spectrum we also record the observed noise level in the observation by computing the root mean square (rms) variation of the reflectance factor in the spectral regions between the lines used in the retrieval. This quantity is used as a quality control parameter. Retrievals with a noise level greater than a
- 209 threshold value are rejected.
- 210 **3.3. Modeling the AOTF**
- 211 The presence of the AOTF complicates modeling of the observed NOMAD spectrum
- since the observed signal for a given diffraction order will also contain some amount of
- 213 signal from neighboring orders. Since the CO absorption lines used for this analysis are
- 214 well spaced with essentially no contribution from other species (Fig. 3), the main concern
- 215 is the addition of continuum signal from neighboring orders to the observed signal of the
- 216 CO lines in the orders being studied. The effect of adding this continuum from
- 217 neighboring orders would be to artificially reduce the integrated line depth that we use as
- 218 our observation to be retrieved, and thus to systematically reduce the CO vmr values that
- 219 we retrieve.
- 220 Fortunately, this effect is observed to be relatively small for the NOMAD LNO
- 221 observations in orders 189 and 190 that are used for our CO retrievals. Ideally,

- absorptions that are very optically thick at the spectral resolution of NOMAD would be
- 223 used to estimate this out-of-order continuum contribution. In this case, the expected
- signal would be zero inside the absorption and any non-zero signal that is observed could
- be attributed to the continuum from other orders. There are no such optically thick
- absorptions for these diffraction orders, so instead we use two different means to estimate
- 227 the contribution from neighboring orders.
- 228 The most straightforward method is to look at the relative amplitude of spectral lines 229 aliased from neighboring orders by the AOTF. Figure 5 shows four of the same spectra 230 for order 189 as shown in Fig. 4. The black vertical lines indicate the frequency of the 231 CO absorptions for order 189. As expected, they line up with the observed spectral 232 features. The red and blue vertical lines indicate the frequencies where we would expect 233 to see CO absorptions aliased from orders 188 (red lines) and 190 (blue lines) by the 234 AOTF. Spectral features aliased from neighboring orders are clearly visible in many 235 NOMAD observations, for example, the solar occultation observations analyzed by Aoki 236 et al., (2019), and the analysis of Liuzzi et al., (2019). However, the LNO observations 237 for orders 189 and 190 show essentially no features at the expected locations, which
- 238 indicates that the contribution from other orders is small.
- 239 The top panel of Fig. 6 shows how comparing the observed line depth of a CO absorption 240 aliased from a neighboring order (4268.8 cm-1, the middle blue line in Fig. 5) against the observed line depth of a CO absorption from the order being observed (4267.55 cm-1, the 241 242 middle black line in Fig. 5) can be used to estimate the continuum contribution from 243 neighboring orders. The greater the observed line depth ratio, the greater the contribution 244 from neighboring orders, and the smaller the line depth would be for the CO lines used in 245 the retrieval. A detailed analysis looking at all the LNO order 189 observations shows 246 this observed line depth ratio to be no larger than 0.02. Using Fig. 6, this implies that the 247 observed CO lines used in this retrieval are at least 0.92 times as strong as they would be 248 with no AOTF. In other words, the true CO line depths at Mars (with no AOTF) are at 249 most 1.08 times stronger than observed.
- A second way to estimate the out-of-order continuum contribution is to look at how
 retrieved CO varies as a function of a known quantity where the expected variation is
 non-linear. The variation of CO with surface pressure is the best example of this. Surface
- 253 pressure is (essentially) known from model results, and the expected line depth of CO
- varies non-linearly with surface pressure both because the curve of growth is non-linear
- 255 (i.e., doubling the amount of CO leads to a line depth less than twice as large) and
- because of the effects of pressure broadening of the absorption lines.
- 257 The bottom panel of Fig. 6 shows the results of a numerical experiment where we use a
- constant CO vmr and a range of surface pressures to compute expected line depths. We
- 259 multiply each of these line depths by factors ranging from 0.9 to 1.0 (as indicated in Fig.

260 6 by the numbers at the ends of the black curves) and then perform the retrieval on these 261 reduced "observed" line depths assuming no AOTF. The results, normalized to their value at 6 mbar, show a systematic variation with surface pressure. The actual CO 262 retrievals using NOMAD LNO observations (presented in detail in Section 4) for orders 263 264 189 and 190 are shown by the red and blue points. Here we have used only retrievals 265 between 30° S and 30° N latitude to minimize real variations of CO vmr, and we have 266 smoothed the results by convolving with a function 1 mbar wide in surface pressure. As 267 expected, the retrievals do show a tendency toward lower CO vmr values at lower surface 268 pressures, but the implied continuum contribution from other orders is not large, with the 269 resulting strength of the observed CO lines being at least 0.95 times as strong as they 270 would be with no AOTF.

271 Taken together, the two independent analyses above present strong evidence that the 272 AOTF contribution to the continuum from neighboring orders is relatively minor for the 273 specific case of the LNO observations for orders 189 and 190. Again, we note that AOTF 274 is observed to produce significant contributions from neighboring orders for other orders 275 and modes (e.g., Liuzzi et al., 2019; Aoki et al., 2019). For this case of these retrievals, 276 the observations are consistent with the CO lines being at least 0.92-0.95 times as strong 277 as they would be with no AOTF. Therefore, for simplicity we have chosen to not include 278 the AOTF in our retrieval. Systematically underestimating line depths by 5–8% would 279 lead to a systematic underestimate of retrieved CO by as much as 20%. However, any 280 such effect of the AOTF not accounted for in the retrieval would be a nearly constant 281 factor applied to every individual retrieval, so that while this uncertainty would affect the 282 overall average CO vmr it would have essentially no effect on the seasonal and latitudinal 283 variations of CO that are the focus of this work.

284 **3.4. Uncertainties**

In addition to uncertainties from the AOTF, there are a number of other sources that could contribute systematic uncertainties. Given the form of our retrieval algorithm, the uncertainty in retrieved results is most easily estimated through the use of numerical experiments. For each source of uncertainty to be evaluated, the retrieval algorithm can be performed over a range of different assumptions, approximations, or values to evaluate the resulting change in the retrieved volume mixing ratio of carbon monoxide.

The most straightforward quantities to test are model-related assumptions, such as the number of vertical layers in the model, the number of radiation streams included in the discrete ordinates formulation, and the number of terms kept in the Legendre polynomial expansion of the scattering phase functions. These parameters can be set large enough so that they do not contribute significantly to the total uncertainty. In each case the model parameters were chosen so that the retrieved CO vmr changed by less than one percent when the number of model layers, radiation streams, or phase function terms was

- doubled. The actual values used in the retrieval are 16 radiation streams, 32 terms in the
 Legendre polynomial expansion, and 25 vertical layers in the model.
- 300 Although thermal radiation is negligible in these observations, atmospheric temperatures
- 301 can still affect the retrievals since the spectroscopic properties of CO, including line
- 302 strengths and widths, are temperature dependent. Since we use atmospheric temperatures
- 303 from OpenMARS that are specifically computed from assimilation of observations during
- 304 Mars Years 34 and 35, we expect them to be well within 10 K of the true value for all
- 305 cases. As a test of the worst case, we found that an offset in the temperature profile by 10
- 306 K over the entire atmosphere leads to a 2–6% change in retrieved CO vmr.
- 307 It is not possible to reliably retrieve aerosol optical depth from the individual NOMAD
- 308 LNO spectra themselves because of the very limited spectral range in each spectral order.
- 309 However, the use of dust and water ice aerosol optical depth from concurrent THEMIS
- 310 observations provides a useful estimate. Numerical experiments show that doubling the
- aerosol optical depth found outside of major dust storms leads to changes in the retrieved
- 312 CO vmr by 5% or less. During large dust storms the optical depth of the dust can become
- 313 large enough to effectively screen the lower part of the atmosphere so that the entire
- 314 column is not sampled by the observations. For this reason, we choose to reject any
- 315 observation for which the extinction optical depth at 9-µm is greater than unity. This
- 316 corresponds to an extinction optical depth at the wavelength used in this retrieval of about
- 317 1.8. Our assumptions for the vertical profile of dust and water ice aerosol are found by
- numerical experiment to cause changes in the retrieved values by 10% or less for any
- 319 reasonable choices.
- 320 Uncertainty related to our definition of the continuum level was tested by performing the
- 321 retrieval for a large suite of cases using different offsets from the line center for
- 322 computing the continuum (including cases with asymmetric offsets). The retrieval results
 323 were found to vary by as much as 10% for reasonable choices for the definition of the
- 324 continuum. Considering all of the above sources of uncertainty, we estimate that the total
- 325 systematic one-sigma uncertainty in an individual retrieval of CO vmr to be 20% or less
- 326 from sources other than the AOTF.
- 327 Perhaps the most straightforward way to estimate an overall uncertainty estimate is by
- 328 looking at the retrieval values themselves. In addition to the uncertainties described
- 329 above, the amplitude of random noise in the observed spectra relative to the observed
- 330 continuum level varies significantly as a function of solar incidence angle, surface
- albedo, and the distance between Mars and the Sun. As mentioned earlier, we compute
- the amplitude of the random noise relative to the continuum level for each spectrum to
- 333 use as a quality criterion. We select the maximum allowed value for the noise level based
- 334 on a tradeoff between the desire to retain as many retrievals as possible, while keeping
- the observation-to-observation variation of retrieved CO vmr to a minimum.

336 For a given maximum allowed value for the noise level, we compute the rms difference between individual retrievals and a smoothed average formed by a 2-D convolution of the 337 retained retrievals using a bin size 45° in L_s and 15° in latitude. Figure 7 shows the results 338 for this analysis, which was performed separately for retrieval results from orders 189 339 340 and 190. For the strictest cases (lowest maximum allowed noise level) the fraction of the 341 retrievals retained is low, but the overall uncertainty in the retrievals estimated by the rms from their smoothed average is also relatively low. As the noise level criterion is relaxed 342 343 to allow retrievals from observations with higher noise levels, progressively more 344 retrievals are retained at the cost of more scatter in the retrieval results. Given the 345 relatively gradual slopes in Fig. 7, we select values (indicated by arrows) for the 346 maximum allowed noise level to retain 85-90% of the retrievals. Specifically, we set the 347 maximum allowed noise level relative to the continuum level to be 0.025 for order 189 348 and 0.035 for order 190. This leads to our best estimate for the overall uncertainty in 349 individual retrievals of CO vmr to be about 240 ppmv for order 189 and 290 ppmv for order 190. Given that the global, seasonal average is about 800 ppmv (see section 4), this 350 corresponds to 30–35%, which is comparable to or slightly better than that for the 351 352 CRISM retrievals of CO vmr (Smith et al., 2009; 2018). This total uncertainty is larger 353 enough than the 20% systematic uncertainty described above that these retrievals are not 354 dominated by systematics.

355 4. Retrieval Results

Here we present the results of the retrieval of the column-integrated carbon monoxide 356 357 volume mixing ratio for all NOMAD LNO observations using orders 189 and 190 taken 358 between Mars Year (MY) 34, L_s=150° and MY 35, L_s=241° (28 March 2018 and 18 July 359 2020). This covers 1.25 Mars Years, providing a climatological view of CO and its seasonal and spatial variations. Excluded are observations with a solar incidence angle 360 361 greater than 55°, those taken during major dust storms, and those with noise levels that 362 exceed the threshold value given in the previous section. These retrieval results are 363 available for download at http://dx.doi.org/10.17632/px89dk6ck9.1

364 4.1. Climatology of Retrieved Carbon Monoxide

The retrieved column-integrated carbon monoxide volume mixing ratio is shown in Fig. 8 as a function of season (L_s) and latitude separately for the retrievals using order 189 and

367 190. As described above in detail, the uncertainty in individual retrieved values are

- relatively large (30–35%) so the CO vmr shown in Fig. 8 has been smoothed to highlight
- 369 the trends. The size of the smoothing box is 15° in L_s and 15° in latitude and is shown in
- the figure for comparison. In total, there are 34,152 retrievals for order 189 and 75,007
- 371 retrievals for order 190.

- 372 The overall level of CO vmr and its seasonal and latitudinal variations are broadly similar
- between the two orders. While there are minor differences, perhaps most notably the
- 374 generally higher CO vmr in order 189 retrieved around $L_s=180^\circ$, these differences are
- 375 well within the stated uncertainties and do not appear from our analysis to arise from any
- 376 systematic differences between the two orders. Therefore, from this point forward we will
- 377 describe the combined results from each of the two orders under the assumption that this
- 378 provides the most accurate value possible.
- The CO climatology displayed in Fig. 8 bears a strong resemblance to those retrieved previously from CRISM (Smith et al., 2008; 2019) and from Mars Express PFS (Bouche
- previously from CRISM (Smith et al., 2008; 2019) and from Mars Express PFS (Bouch et al., 2021), and is largely as expected for a non-condensable gas (e.g., Forget et al.,
- 2008; Daerden et al., 2019; Holmes et al., 2019). There is depletion of carbon monoxide
- in the summertime polar regions in both hemispheres, although it is significantly stronger
- in the south. The southern hemisphere summertime depletion leads to CO vmr values of
- 385 400 ppmv or less poleward of 70°S, while in the north at least ~600 ppmv of CO is
- 386 maintained at all observed locations. In both hemispheres the minimum CO vmr values
- are found somewhat before solstice (by up to $\sim 30^{\circ}$ of L_s), especially at latitudes further
- 388 removed from the pole. The summertime depletion of CO extends to roughly 40° latitude
- in both hemispheres, with a roughly constant gradient at higher latitudes trending to
- 390 lower CO vmr toward the pole. Model results indicate a corresponding maximum in CO
- 391 vmr over the winter poles (Forget et al., 2008; Daerden et al., 2019; Holmes et al., 2019),
 392 but this cannot be directly confirmed with retrievals using NOMAD LNO, which rely on
- solar illumination for the observed signal.
- 394 The NOMAD LNO retrievals show that the CO vmr at low latitudes follows a seasonal
- 395 variation that ranges between roughly 700 ppmv near $L_s=0^\circ$ and 900–950 ppmv near
- $L_s=180^\circ$, with an annually averaged value near 800 ppmv. Moving from south to north,
- 397 the peak annual value tends to occur at gradually later seasonal dates, from about
- 398 $L_s=140^\circ$ at 30° S latitude to about $L_s=220^\circ$ at 30° N latitude (this trend is more easily
- 399 seen in the combined and smoothed version of the retrievals discussed in Section 5.1 and
- 400 shown in Fig. 10). As expected for a non-condensable gas, the overall seasonal variation
- 401 of CO vmr is observed to roughly follow an inverse relation with the annual variation of
- 402 surface pressure observed from the surface of Mars (e.g., Tillman et al., 1993; Martínez
- 403 et al., 2018), although the pattern is also modified by the latitudinal transport of CO (e.g.,
- 404 Daerden et al., 2019; Smith et al., 2018) as will be further discussed in Section 5.3.
- 405 Given the relatively long chemical lifetime of CO lived (~6 years; Krasnopolsky, 2007),
- 406 we do not expect to see interannual variations in these retrievals. The differences between
- 407 the MY 34 retrievals at the beginning of the time period shown in Fig. 8 and the MY 35
- 408 retrievals at the end are instead indicative of the level of uncertainty in the retrievals.

409 4.2. Spatial and Diurnal Variations of Retrieved Carbon Monoxide

- 410 Figure 9 shows maps of the spatial variation of CO vmr for the four cardinal seasons of
- 411 the Martian year. To attempt to identify trends, a spatial smoothing has been performed
- 412 using a box 45° in longitude by 15° in latitude. The largest variations are the latitudinal
- 413 gradients that describe the annual climatology described in the previous section. At
- 414 $L_s=270^\circ$ there is a clear gradient from higher values of CO vmr in the north where it is
- 415 winter to lower values in the south where it is summer.
- 416 Overall, there appears to be limited variation of CO vmr with longitude. The relative lack
- 417 of correlation between CO vmr and surface topography supports our assumption that CO
- 418 is largely well-mixed, at least in the lower portion of the atmosphere containing the bulk
- 419 of the column mass. Most of the small amplitude longitudinal variations that do appear in
- 420 Fig. 9 are not statistically significant and do not correspond to features in similar maps of
- 421 CO vmr based on CRISM retrievals presented in Smith et al. (2018). The exception is
- 422 that the Hellas region at $L_s=90^\circ$ stands out as having a higher CO vmr than neighboring
- 423 locations. This enhancement in Hellas during winter was also observed in CRISM
- 424 retrievals of CO (Smith et al., 2018), although the corresponding enhancements observed
- 425 in regions with low-lying topography in the north at $L_s=270^\circ$ are not observed in the
- 426 LNO retrievals shown here.
- 427 Recalling the top panel of Fig. 1, the precessing orbit of the Trace Gas Orbiter enables
- 428 NOMAD to view a range of local times between roughly 08:00 and 16:00 local true solar
- 429 time over a relatively short seasonal timescale. However, our analysis of the retrievals
- 430 does not reveal any systematic variation of column-integrated CO vmr as a function of
- 431 local time at levels greater than 10%, at least for the daytime hours observable by
- 432 NOMAD LNO.

433 **5. Discussion**

434 **5.1.** Comparison with CRISM and Other Previous Results

- 435 The retrievals of carbon monoxide volume mixing ratio from this work can be directly
- 436 compared against previous retrievals from other spacecraft observations. In particular, we
- focus here on a comparison against the retrievals from CRISM, which use the same CO
- 438 absorptions and have a similar uncertainty level (Smith et al., 2009; 2018). The CRISM
- 439 observations were taken from MY 28 to 33. The top two panels in Fig. 10 show a side-
- 440 by-side comparison of the CO climatology retrieved from CRISM and the current work.
- 441 For ease of comparison, each has been smoothed with a box 45° in L_s and 15° in latitude
- to highlight global/seasonal trends, and the CRISM climatology has been resampled to
- the same L_s and latitude values as given by the NOMAD LNO retrievals.
- 444 The overall annual average value is roughly the same in the NOMAD LNO and CRISM
- retrievals at 800 ppmv. And, the overall pattern of variation described in Section 4.1, with
- 446 reduced CO vmr near the south pole during summer solstice, a smaller reduction in CO

- 447 vmr near the north pole during summer solstice, and low-latitude CO vmr inversely
- 448 correlated with the annual cycle of surface pressure, is observed in both the NOMAD
- LNO and the CRISM datasets. However, one difference is that the amplitude of the
- 450 variations is noticeably less in the NOMAD LNO retrievals, being perhaps half as large
- 451 as that in the CRISM retrievals.
- 452 Other differences are present, but smaller. The northern summer minimum in the CRISM
- retrievals is more extensive than in the NOMAD LNO retrievals, both in terms of
- 454 latitudinal extent and seasonal duration. On the other hand, the southern summer
- 455 minimum has very similar latitudinal extent and seasonal duration in the two datasets.
- The NOMAD LNO retrievals tend toward somewhat greater CO vmr values than those
- 457 from CRISM at low northern latitudes during the second half of the Martian year
- 458 (L_s=180°-360°) and do not show the slight decrease apparent in CRISM southern
- 459 hemisphere retrievals between $L_s=0^\circ$ and 120° .
- 460 Recently, another climatology of CO vmr has been retrieved using observations from the
- 461 Planetary Fourier Spectrometer (PFS) on the Mars Express spacecraft (Bouche et al.,
- 462 2021) spanning MY 26 to 34. This retrieval uses a different set of CO absorptions at
- 463 \sim 2100 cm⁻¹ (4.7 μ m), but are still directly comparable to the NOMAD LNO results.
- Bouche et al. (2021) find an overall annual average CO vmr of 820 ppmv and many of
- 465 the same climatological variations described above. Their southern summer minimum has
- 466 a somewhat smaller amplitude (~500 ppmv for PFS, ~400 ppmv for NOMAD LNO), but
- the northern summer minimum is somewhat deeper (~600 ppmv for PFS, ~650 ppmv for
- 468 NOMAD LNO) and there is a larger annual variation at low latitudes in the PFS469 retrievals.
- 470 It is unlikely that the observed differences between the CRISM, PFS, and NOMAD
- 471 retrievals are caused by real interannual variations in CO vmr. The chemical lifetime for
- 472 CO is relatively long, and the different retrievals are generally within the uncertainties of
- the different retrievals. Furthermore, all three of the climatologies compared here (from
- 474 NOMAD, CRISM, and PFS) are of retrieved values that have been averaged over all
- 475 Mars Years observed by each instrument.

476 5.2. Comparison with GCM Modeling Results

- 477 The retrievals of CO vmr from the NOMAD LNO observations can also be compared
- 478 against model results. Such a comparison can provide insight into the physical processes
- that drive the observed climatology, and differences between observations and model
- 480 results can identify areas where improvement is needed in the model or retrieval process.
- 481 The model used here is the GEM-Mars General Circulation Model (Daerden et al., 2015;
- 482 Neary and Daerden, 2018; Daerden et al., 2019). This model uses 103 vertical levels
- 483 extending from the surface to \sim 150 km, and it is operated on a grid with a horizontal

484 resolution of 4°×4° in latitude and longitude. More details on the physical 485 parameterizations in the model and evaluations of simulations are provided in Neary and 486 Daerden (2018), Smith et al. (2018), Daerden et al. (2019), Neary et al. (2020), and 487 Bouche et al. (2021). Here we summarize the parameterization for the non-condensable 488 gas enrichment. Deposition (sublimation) of CO₂ above a model point results in a change 489 of the surface pressure. The dynamical core of the GCM then readjusts the atmospheric 490 mass globally, and the impact of the mass loss at the poles is spread out instantaneously 491 over the entire planet. Locally, the enrichment (or depletion) of minor species will build 492 up more gradually and spread out on a longer timescale by eddy mixing and global 493 circulation. Because of the definition of the model levels in a GCM in terms of surface 494 pressure (implying that the model levels themselves will be modified upon deposition or 495 sublimation), this local process is difficult to disentangle from the global pressure 496 correction. Forget et al. (2008) and Lian et al. (2012) developed methods to treat this 497 problem in their GCMs. In GEM-Mars we apply a simple parameterization that corrects 498 the local vmr after deposition (or sublimation) in two steps. First the vmr of CO₂ is modified at all model levels by a single factor that corrects its column abundance to 499 500 match the local surface pressure change. Then it is additionally modified at those vertical 501 model levels where CO_2 ice deposition occurred, by a factor that is proportional to the 502 vmr of condensing (or sublimating) ice particles. The factors of proportionality (one for 503 deposition and one for sublimation, see Smith et al. (2018) and Bouche et al. (2021) for 504 details) do not necessarily have the same values, as they describe different processes that involve different timescales, but they should be well balanced to ensure that the total 505 506 atmospheric content of noncondensing species does not change.

507 For the simulations presented here, the atmospheric dust was constrained by the dust 508 climatologies provided by Montabone et al. (2015, 2020). For total CO columns, no 509 significant difference was found between using the dust climatology for a year with 510 nominal dust loading (e.g., MY 33) and using the observed dust climatology for MY 34 511 (when the early NOMAD observations were taken), which included a global dust storm 512 (e.g., Smith, 2019). It was found that the global dust storm had an impact on the higher 513 altitude photochemistry through the redistribution of water vapor (Neary et al., 2020), 514 and therefore also on CO vmr in the upper atmosphere, but did not have an impact on the 515 lower atmospheric CO and on the total column amounts such as retrieved by NOMAD 516 LNO observations.

517 The bottom two panels of Fig. 10 show a comparison of CO vmr climatology as retrieved 518 by NOMAD LNO observations against that computed by GEM-Mars. Here, the overall 519 average initial value of CO vmr has been adjusted in the model, compared to the results 520 shown in Smith et al. (2018), to match the NOMAD observations, so it is the seasonal 521 and latitudinal variations that should be compared. The GEM-Mars model closely 522 matches the observed climatological patterns. In particular, the southern summer

- 523 minimum is well represented as is the modest increase in low-latitude CO vmr between
- 524 $L_s=120^\circ$ and 210° in response to the annual minimum in surface pressure. The amplitudes
- 525 of those two features as computed by GEM-Mars are more similar to those observed by
- 526 NOMAD LNO than to the larger amplitude features observed by CRISM. Perhaps the
- 527 largest difference between the model and the retrievals is the northern summer minimum.
- 528 The NOMAD LNO retrievals show a much smaller decrease than the CRISM retrievals,
- 529 but the minimum in the GEM-Mars has even smaller amplitude.
- 530 Figure 11 shows a comparison between the NOMAD LNO retrievals and the GEM-Mars
- 531 model results in some more detail. Both datasets have been binned in latitude bands as
- stated, and then binned in L_s using a box 15° wide sliding 5° of L_s between each point.
- 533 Again, there is a close correspondence with the largest differences being at high northern
- 534 latitudes where the model predicts higher CO vmr than observed, and a smaller latitudinal
- 535 gradient at low latitudes (-40° to $+40^{\circ}$) during the second half of the Martian year
- 536 (L_s=180°-360°). The remaining differences, with a root-mean-square amplitude of ~50
- 537 ppmv, are within the noise level in the retrievals.
- 538 In general, the GEM-Mars General Circulation Model can reproduce the relative
- 539 distribution of the observed climatology of CO vmr and explain its broad features
- 540 (Daerden et al., 2019). Carbon monoxide is a relatively long-lived (~6 years;
- 541 Krasnopolsky, 2007) and non-condensable species. As such, we do not expect significant
- 542 interannual variations, and the column-averaged volume mixing ratio of CO is controlled
- 543 by the condensation of CO_2 and latitudinal transport. The condensation of CO_2 onto the
- southern seasonal cap during winter is followed by this CO-enriched air being transported
- 545 equatorward and into the northern hemisphere between $L_s=120^\circ$ and 180° forming the
- 546 low-latitude maximum in CO vmr that is observed. Sublimation of the seasonal CO₂ cap 547 during Spring and Summer releases large amounts of CO₂ leading to a minimum in CO
- 548 vmr. The amount of CO₂ condensed onto the southern cap is much greater than that
- 549 condensed onto the northern cap, which explains the difference in the amplitude of the
- two summertime minima in CO vmr. Assimilation of the CO climatology observed by
- 551 NOMAD LNO could help improve the parameterization of model processes and thereby
- reduce the difference between the model and retrieval results, which is the subject of
- 553 active research.

554 6. Summary

- 555 The nadir-geometry LNO observations taken by the NOMAD instrument enable the
- 556 characterization of the climatology of the column-averaged volume mixing ratio of
- 557 carbon monoxide gas in the Martian atmosphere. Comparison of these retrieved values
- 558 with the results from the GEM-Mars GCM model allow for physical interpretation of the
- retrieved seasonal and spatial variations, and also serve to highlight possible areas for
- 560 improvement in both the model and the retrieval process.

- 561 NOMAD LNO observations taken covering more than one full Martian Year reveal the
- same general climatological trends that have been observed by other spacecraft (e.g.,
- 563 Smith et al., 2009; 2018; Bouche et al., 2021), but with some differences. The global,
- annual average value of CO vmr is found to be \sim 800 ppmv for the daylight portions of
- 565 Mars observable by the NOMAD LNO observations. A minimum in CO vmr is observed
- 566 at high latitudes around the summer solstice in both hemispheres, although the decrease 567 in CO vmr is much more pronounced in the south than in the north because of the greater
- 567 in CO vin is inder more pronounced in the south than in the north because of the greater amount of CO₂ released from the seasonal cap in the south. At low latitudes, the seasonal
- 569 trend generally follows that expected of a non-condensable gas (e.g., Forget et al., 2008;
- 570 Daerden et al., 2019; Holmes et al., 2019) with maximum CO vmr in the season around
- 571 $L_s=180^\circ$ as air enriched with CO is transported northward across the equator. Maps of
- 572 CO vmr show relatively little spatial variation outside of a noticeable enhancement over
- 573 Hellas and other areas of low-lying topography during southern summer. There is no
- 574 obvious trend in CO vmr as a function of local time for the daytime hours observed by
- 575 NOMAD LNO. Compared to retrievals of CO vmr from CRISM (Smith et al., 2018), the
- 576 NOMAD results show less overall variation with the high-latitude summer minima in
- 577 each hemisphere having an amplitude roughly half as large.
- 578 Model results of CO vmr from the GEM-Mars GCM model broadly agree with the
- 579 NOMAD LNO retrievals. The correspondence is closer than that between the GEM-Mars
- 580 results and the CRISM retrievals of CO vmr (Smith et al., 2018), capturing both the
- 581 amplitude of the high-latitude summertime minima and low-latitude seasonal variations
- 582 observed by NOMAD. Small differences between the model and retrievals may be
- 583 reduced with further improvement in model parameterizations.
- 584 The ExoMars Trace Gas Orbiter and NOMAD instrument continue to operate at the time
- 585 of this writing taking new LNO observations in orders 189 and 190 to characterize CO.
- 586 Further observations will extend the existing record of carbon monoxide retrievals to
- 587 provide an even better estimate of CO climatology through better statistics and by filling
- 588 in the gaps between lobes seen in Fig. 10, and will enable the study of the limits of
- 589 interannual variation in CO and the response of CO to large dust storms.

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Figure 1. The seasonal and latitudinal coverage of the NOMAD LNO observations in orders 189 and 190 used in this study. Shown are (top) the local true solar time for each observation, and (bottom) the solar incidence angle.



Figure 2. A typical NOMAD LNO observation for order 190 shown in terms of reflectance factor. The data were taken on 5 November 2018 (MY 34, Ls=283°). Spectral features caused by CO are visible as vertical lines in the image. In this representation, the continuum level signal is controlled largely by surface albedo, solar incidence angle, and aerosol scattering.



Figure 3. A computed spectrum of the absorptions from CO (black), water vapor (blue), and CO_2 (green) convolved to a spectral resolution of 0.4 cm⁻¹. The spectral range covered by different NOMAD LNO grating orders are indicated by the numbers 186 through 191. The orders used in this study (189 and 190) contain strong CO lines with no interference from other gases.



Figure 4. Averaged spectra for (top) order 189 and (bottom) order 190 showing the CO absorptions used for the retrieval. These are lines from the (2-0) ro-vibrational band of CO, lines R0, R1, R2, and R3 for order 189, and R6, R7, and R8 for order 190. The red line segments show the estimated continua and the spectral range over which the integrated line depths are computed for each line.



Figure 5. Averaged spectra for order 189 showing the expected spectral locations of spectral features. The black vertical lines show the frequencies of three of the main CO lines in order 189. The red and blue vertical lines show the frequencies where CO absorptions from orders 188 (red) and 190 (blue) would appear if aliased by the AOTF.

Figure 6. Two ways to estimate the continuum contribution from neighboring orders from the AOTF. (Top panel) The expected line depth of CO lines as a fraction of that with no AOTF is shown as a function of the observed line depth of a CO absorption aliased from a neighboring order (4268.8 cm-1, the middle blue line in Fig. 5) against the observed line depth of a CO absorption from the order being observed (4267.55 cm-1, the middle black line in Fig. 5). (Bottom panel) For line depths multiplied by a given constant value (the numbers given at each end of the black lines), the retrieved CO vmr as a function of surface pressure has a systematic variation as shown by the black curves. The CO vmr retrieved from the observations (red and blue points) are consistent with this factor being 0.95 or greater.

Figure 7. The observed level of uncertainty in the retrieved value of CO vmr as a function of the fraction of retrievals retained. Each point represents a different upper limit for the maximum allowed noise level in the observations. The arrows show the selected upper limit for each order.

Figure 8. The column-averaged carbon monoxide volume mixing ratio retrieved from NOMAD LNO spectra as a function of season, latitude, and grating order.

Figure 9. Maps showing the spatial variation of CO vmr retrieved from NOMAD LNO spectra for four different seasons.

Figure 10. A comparison of CO vmr climatologies from (top) CRISM, (middle) NOMAD LNO, and (bottom) the GEM-Mars model. The CRISM and GEM-Mars model data have been interpolated to the times and locations of the NOMAD LNO retrievals, and all three datasets have been smoothed (45° in L_s and 15° in latitude) for easier comparison.

Figure 11. Comparison of GEM-Mars model simulation of CO vmr (red points) with NOMAD LNO retrievals of CO vmr (black points). Both datasets have been binned 15° in L_s.