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► **To cite this version:**

Kevin Olsen, Franck Lefèvre, Franck Montmessin, A. A. Fedorova, A. Trokhimovskiy, et al.. The vertical structure of CO in the Martian atmosphere from the ExoMars Trace Gas Orbiter. *Nature Geoscience*, 2021, 14, pp.67-71. 10.1038/s41561-020-00678-w . insu-03313378

**HAL Id: insu-03313378**

**<https://insu.hal.science/insu-03313378>**

Submitted on 3 Aug 2021

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# The vertical structure of CO in the Martian atmosphere from the ExoMars Trace Gas Orbiter

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November 11, 2020

## Abstract

Carbon monoxide (CO) is the main product of CO<sub>2</sub> photolysis in the Martian atmosphere. Production of CO is balanced by its loss reaction with OH, which recycles CO into CO<sub>2</sub>. CO is therefore a sensitive tracer of the OH-catalyzed chemistry that contributes to the stability of CO<sub>2</sub> in the atmosphere of Mars. To date, CO has only

6 been measured in terms of vertically-integrated column abundances and the upper  
7 atmosphere, where CO is produced, is largely unconstrained by observations. Here  
8 we report vertical profiles of CO from 10 to 120 km, and from a broad range of lat-  
9 itudes, inferred from the Atmospheric Chemistry Suite onboard the ExoMars Trace  
10 Gas Orbiter. At solar longitudes 164–190°, we observe an equatorial CO mixing ra-  
11 tio of  $\sim 1000$  ppmv (10–80 km), increasing towards the polar regions to more than  
12 3000 ppmv under the influence of downward transport of CO from the upper atmo-  
13 sphere, providing a view of the Hadley cell circulation at Mars equinox. Observations  
14 also cover the 2018 global dust storm, during which we observe a prominent depletion  
15 in the CO mixing ratio up to 100 km. This is indicative of increased CO oxidation in  
16 a context of unusually large high-altitude water vapour, boosting OH abundance.

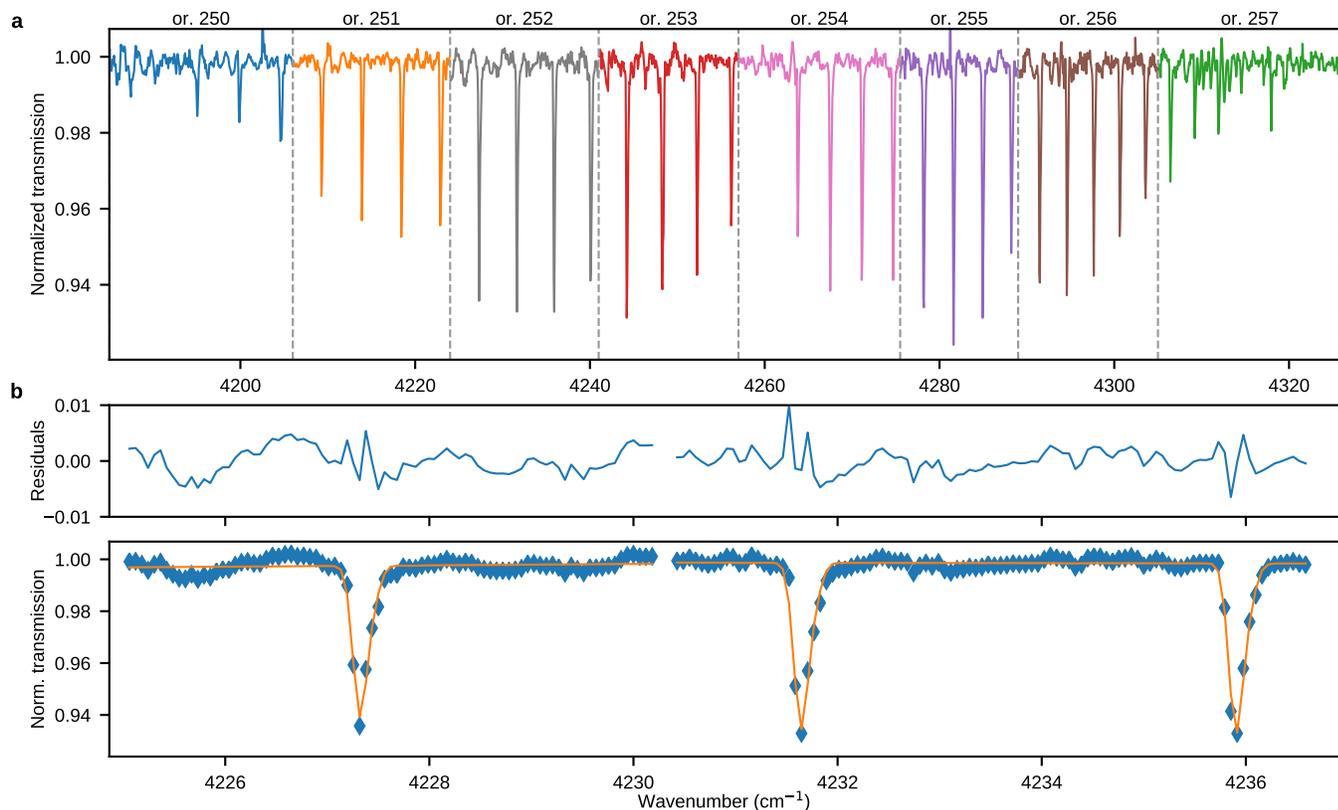
17 Carbon monoxide (CO) plays an important part in the Martian atmosphere, with an  
18 integral role in both hydrogen and oxygen chemical cycles. It is mainly governed by simple  
19 chemistry, being produced in the upper atmosphere (60 km) by CO<sub>2</sub> photolysis, and de-  
20 stroyed in the lower atmosphere by the hydroxyl (OH) radical, a product of water vapour  
21 (H<sub>2</sub>O) photolysis [1, 2, 3, 4, 5]. Tracking atmospheric CO on Mars is an effective method for  
22 exploring the oxidizing capacity of the Martian atmosphere. The abundance of CO reflects  
23 the rate at which CO<sub>2</sub> is recycled by the catalytic HO<sub>x</sub> chemical cycles, addressing the funda-  
24 mental issue of the long-term stability of Mars’ atmospheric composition. The quantitative  
25 understanding of CO has remained a challenge: most theoretical models predict a long-term  
26 equilibrium value for the CO volume mixing ratio (VMR) that is smaller by a factor 2–4  
27 than those observed [5]. The persistence of this disagreement suggests that an important  
28 part of our understanding of Martian atmospheric chemistry remains missing.

29 Past observations of CO below the thermosphere have been limited to vertically integrated  
30 column abundances, and published abundances are representative of the denser, lower atmo-  
31 sphere. These measurements indicate a slow seasonal variation of CO imparted by the global

32 CO<sub>2</sub> condensation-sublimation cycle (e.g., [6]). The lifetime of CO is  $\sim 5$  terrestrial years, so  
33 such a seasonal cycle is expected [6]. Recent satellite observations made with instruments on  
34 Mars Express (MEx) and Mars Reconnaissance Orbiter (MRO) have observed CO volume  
35 mixing ratios of between 500–1000 ppmv [7, 8, 9, 10]. Variations are more pronounced in  
36 polar regions where the CO VMR is directly affected by near-surface CO<sub>2</sub> sublimation and  
37 condensation, as is argon [11, 12]. The most extensive coverage comes from the Compact  
38 Reconnaissance Imaging Spectrometer (CRISM) [13, 10], which found a mean mixing ratio  
39 at mid-latitudes of around 800 ppmv, with a seasonal low-latitude enhancements reaching  
40 about 1000 ppmv around solar longitude (Ls) of 180°.

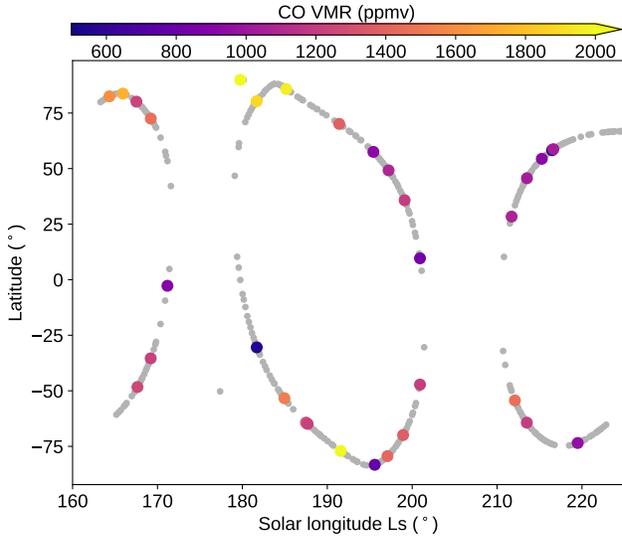
41 Observations of vertical changes to CO density have been made *in situ* by the Neutral  
42 Gas and Ion Mass Spectrometer (NGIMS) on the Mars Atmosphere and Volatile Evolu-  
43 tion (MAVEN) satellite [14]. These measurements are of the particle flux of neutral CO  
44 molecules collected between 140–350 km during low-altitude MAVEN orbits. The density  
45 measurements are restricted to the thermosphere and ionosphere, and full mixing ratio ver-  
46 tical profiles have not been produced. CO<sup>+</sup> ions have also been detected [15].

47 Using the Atmospheric Chemistry Suite (ACS) mid-infrared channel (MIR) onboard the  
48 ExoMars Trace Gas Orbiter (TGO), we present the first vertical profiles of CO in the Martian  
49 atmosphere. These measurements obtained with the solar occultation technique give access  
50 for the first time to the production region of CO in the upper atmosphere (above 60 km),  
51 putting strong constraints on the CO formation rate or, equivalently, the CO<sub>2</sub> photolysis  
52 rate, since there is negligible accompanying CO loss at high altitudes. Above the poles, ACS  
53 measurements of CO characterize the equator-to-pole Hadley circulation that transports CO  
54 from its extra-polar region of production, revealing the pole-ward fluxes of atmospheric mass  
55 involved. Our measurements also cover the particular conditions of the Mars year (MY) 34  
56 global dust storm, supplying direct information on the impact of such events on chemistry  
57 atmospheric circulation, particularly at high altitudes.



**Fig. 1.** | **Spectral fitting with ACS MIR** Example of normalized spectra recorded at 60 km with ACS MIR using secondary grating position 7. Shown are: (a) contributions from diffraction orders 250–257, (b) example fits for spectral windows covering order 252 (bottom) and the associated residuals (top).

58 Fig. 1a shows an example spectrum from a single occultation at 60 km featuring the 2-0  
 59 band of CO. Fig. 1b shows the fit for one order and the associated residuals. Details about  
 60 the ACS MIR instrument, data processing, and VMR vertical profile retrievals are provided  
 61 in Supplementary Methods and Supplementary Figs. 1–3. Vertical profiles of  $T$  and  $P$  were  
 62 retrieved from simultaneous observations made by the near-infrared (NIR) channel of ACS  
 63 [16, 17]. Details about NIR are given in the Supplementary Methods, as are vertical profiles  
 64 of the CO volume mixing ratio,  $T$ , and  $P$  that were generated by running the LMD general  
 65 circulation model (GCM) [18, 19] at coincident times and locations using a reconstructed  
 66 dust climatology for MY 34 [20] (see Supplementary Methods and Supplementary Figs. 4–5

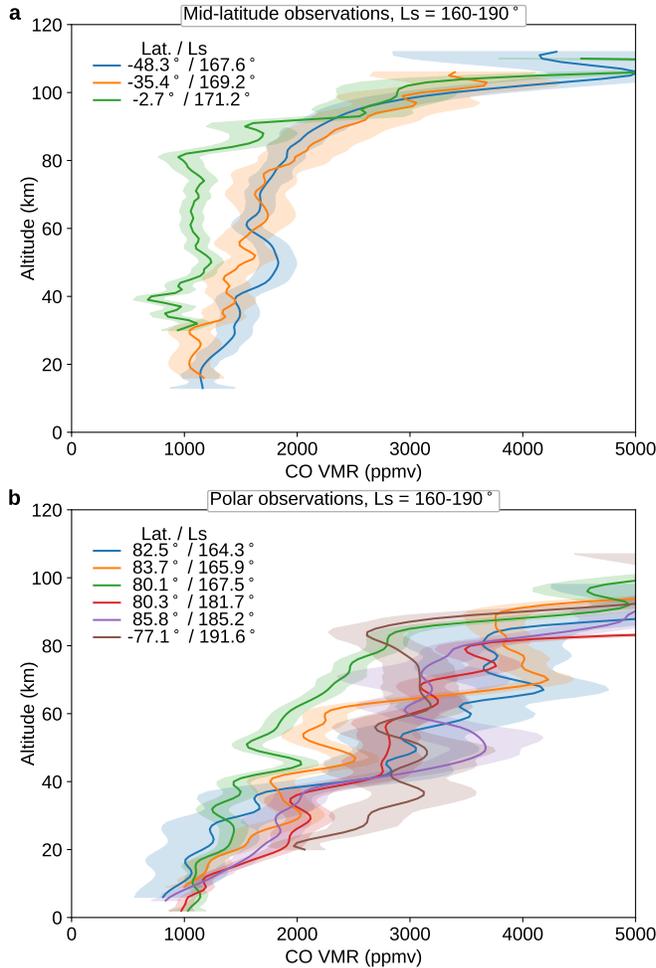


**Fig. 2.** | **Solar occultation latitudes** Distribution over latitude and solar longitude of ACS MIR occultation using position 7, and all other ACS occultation points. The colour scale indicates the retrieved CO VMR between 20–40 km. The solar longitudes correspond to an observational period of April to September 2018.

67 ACS MIR was used in a configuration suitable for CO retrievals (with secondary grating  
 68 position 7) for only a small fraction of observations. Herein we present analysis of the first 32  
 69 occultations recorded between Ls 164° and 220° (April 24 2018 and June 28 2018). The lat-  
 70 itudinal distribution of these observations is shown in Fig. 2 as a function of solar longitude.  
 71 Fig. 2 also indicates the mean VMR of CO between 20–40 km, and the distribution of all  
 72 other ACS MIR occultations. A list of location and time information for these observations  
 73 is given in Supplementary Table 1.

## 74 Mid-latitude observations

75 The first set of CO retrievals we present were measured at mid-to-low latitudes around the  
 76 northern hemisphere autumnal equinox, prior to Ls 190°. Previous measurements of the  
 77 CO vertical column indicate that this is a period when the CO outside the polar regions is  
 78 largest [21, 10]. Fig. 3a presents the retrieved vertical profiles of the CO mixing ratio from



**Fig. 3. | CO VMR vertical profiles at different latitudes** Vertical profiles of retrieved CO mixing ratio as observed by ACS MIR at: a) equatorial and mid-latitudes prior to Ls 190°, and b) at polar latitudes greater than  $\pm 75^\circ$ . The shaded areas show the magnitude of the uncertainty of each retrieved VMR value. Lower altitude ranges are limited by signal attenuation due to aerosols. Uncertainties are derived from the non-linear least squares inversion using partial derivatives.

79 ACS measurements outside the polar regions, and prior to the onset of the global dust storm  
 80 (individual retrievals are shown in Supplementary Figs. 6–7). The altitudes at which a  
 81 reliable retrieval can be performed have upper and lower limits caused by signal attenuation  
 82 due to aerosols that become significant below 30 km, and by the decrease of absorption line  
 83 depths relative to noise above 100 km.

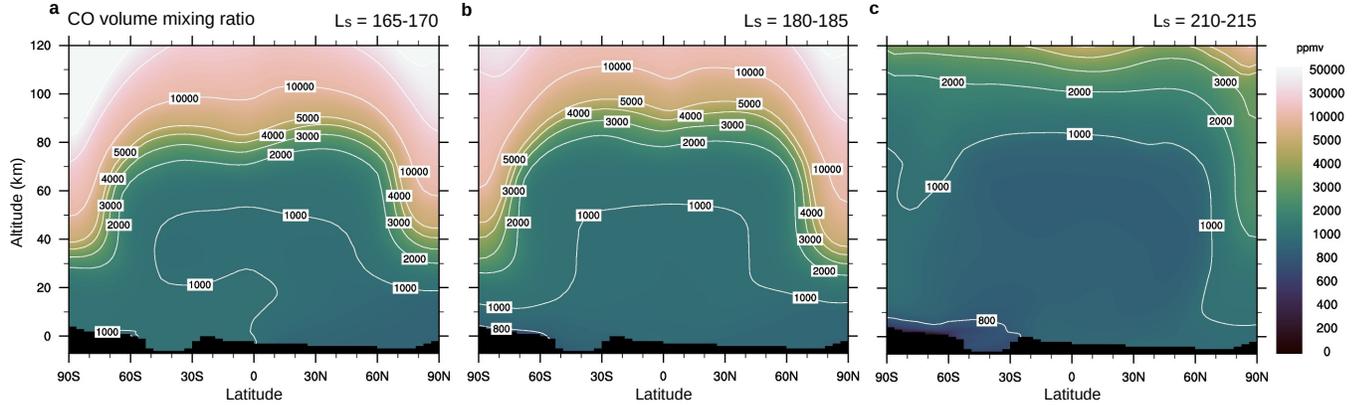
84 The north-most ACS MIR observation, made equatorially at  $-2.7^\circ$  is about 1000 ppmv,

85 in good agreement with the CRISM observations. The more southern observations are  
86 still impacted by the south polar winter CO enrichment, which is also observed by CRISM  
87 [10, 22], and the mean CO mixing ratio rises towards 1260 ppmv. Since the CO mixing  
88 ratios are expected to be nearly constant at lower altitudes, this should be comparable to  
89 column-averaged abundances. The mean mid-latitude value reported by CRISM during this  
90 period is 900–1000 ppmv [10], by PFS is 500–1500 ppmv, and by terrestrial observations is  
91 850–1100 ppmv [21].

92 The neutral CO densities reported by the NGIMS team approach  $10^{10}$  cm<sup>-3</sup> at the  
93 lowest altitude limits of around 130 km. CO number densities computed from the ACS MIR  
94 retrievals and the LMD GCM are in broad agreement at 120 km, covering a range of  $10^9$ –  
95  $10^{10}$  cm<sup>-3</sup>. Thus the ACS MIR observations are in agreement with the NGIMS results, but a  
96 direct comparison is made cautiously as the uncertainty of ACS MIR retrievals is very large  
97 above 100 km, the NGIMS results are likely only reliable above 160 km, and the difference  
98 in altitude coverage will have a very significant impact on composition.

99 Our initial set of observations occurs at a unique time, around the autumnal equinox.  
100 This is a period between polar CO enhancements during the past southern winter and up-  
101 coming northern winter. It is characterized by a CO meridional distribution expected to  
102 be symmetrical across the Equator, under the influence of the mean meridional circulation  
103 dominated by a double Hadley cell structure. Fig. 4 shows the evolution of the CO zonal  
104 mean modelled by the LMD GCM with photochemistry [19]. Panel (a) shows a symmetric  
105 CO distribution at the start of observations, and panel (b) reveals an oncoming change in  
106 the downwelling strength over the polar regions. Around Ls 190°, the Martian atmosphere  
107 was severely affected by a global dust storm that began in the southern hemisphere [23, 24],  
108 shown in panel (c) and discussed below.

109 The vertical profiles observed by ACS are fairly uniform up to around 80 km, at which  
110 point the mixing ratios begin to increase rapidly due to CO<sub>2</sub> photolysis. This is distinct



**Fig. 4. | Modelled CO distributions** Zonally-averaged CO volume mixing ratio (ppmv) calculated by the LMD global climate model using the MY 34 dust climatology [20] for three Ls periods: a) 165–170°, b) 180–185°, c) 210–215°. The onset of the global dust storm was around Ls 190°. The simulation shown here was initialized at Ls = 180° of MY33 and therefore avoids known issues with photochemical models underestimating the long-term equilibrium value of CO.

111 from GCM predictions which have more pronounced mixing ratio increases occurring just  
 112 above 60 km. This indicates that we are observing stronger upward transport of CO, the  
 113 equatorial ascending branch of Hadley circulation, than modelled in the GCM. Zonal means  
 114 at all latitudes derived from the GCM prior to Ls 190° are shown in Fig. 4a and b. This shows  
 115 a well mixed atmosphere between  $-50^\circ$  and  $50^\circ$  with increasing CO abundance pole-ward.  
 116 CO vertical profiles measured by ACS at southern mid-latitudes show a more distinct increase  
 117 in CO compared to the equatorial profile. This is likely to be related to the downwelling  
 118 motion that occurs above both poles around equinox as observed seasonally by CRISM [10].  
 119 That it is also more pronounced than in the GCM again suggests a more intense Hadley  
 120 circulation in the real atmosphere than modelled.

121 We have found that when using  $T$  and  $P$  derived by the GCM, rather than ACS NIR  
 122 retrievals, our retrieved CO VMR tends toward closer agreement with the GCM CO. This  
 123 implies that any underestimation of the CO mixing ratio by the GCM is also linked to  
 124 differences in the physical state of the atmosphere between model and observation.

## 125 **High-latitude observations**

126 Fig. 3b shows the CO VMR vertical profiles retrieved at latitudes greater than  $75^\circ$  in both  
127 hemispheres. Close to the surface the CO mixing ratio measured by ACS near the poles  
128 remains between 900–1200 ppmv as in mid-to-low latitudes. The increase in CO with altitude  
129 is however much more pronounced at high latitudes, where the mixing ratios are of the order  
130 of 2000–3000 ppmv at 40 km. Such values are only measured above 80 km at lower latitudes  
131 (Fig. 3a). This is a result of the Hadley circulation, which, around equinox, consists in  
132 two branches symmetrical across the Equator. Ascending motion prevails at low latitudes,  
133 whereas downwelling over both poles brings large quantities of CO produced in the upper  
134 atmosphere to the lower atmosphere (see Fig. 4b). During this time period, H<sub>2</sub>O VMRs are  
135 reduced over both poles, reducing OH availability, but a reduction in CO depletion rates is  
136 much smaller than the dynamical changes. The fine structures observed over the altitude  
137 range are correlated with those seen in NIR data and are suggestive of horizontal transport,  
138 which mixes CO-rich layers coming from the pole with CO-poor air farther from the location  
139 of downwelling motion.

140 While the GCM and retrievals are in qualitative agreement, the rate at which CO in-  
141 creases dramatically over altitude in Fig. 4, and the altitude at which this occurs, are in  
142 quantitative disagreement. We see higher mixing ratios than predicted below 40 km, and  
143 lower ones above. Such disagreement likely stems from the dynamic model in the GCM. On  
144 one hand, the GCM may be over-estimating the strength of polar downwelling. Otherwise,  
145 it may be forcing the downwelling over too narrow a latitude range. In the downwelling  
146 regions of Fig. 4b, the gradient of the CO vertical profiles changes rapidly with latitude, and  
147 GCM results from more southerly latitudes may be more representative of the sampled air  
148 masses.

## 149 **Impact of the global dust storm**

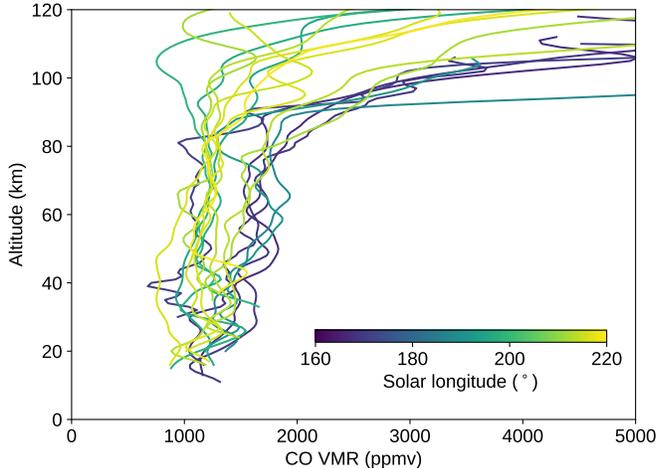
150 From Ls 190° until after Ls 230°, nearly the entire Martian atmosphere was affected by a  
151 global dust storm. Fig. 5 shows the evolution of observed vertical profiles over Ls. During this  
152 period we observe a strong decrease in the CO VMR over time, as well as increasing vertical  
153 transport. The latter is indicated by the rise in the height at which the CO abundance starts  
154 to increase. These correspond to the onset of the global dust storm around Ls 190°.

155 The main impact of a dust event on our ability to study CO is to limit how low in  
156 the atmosphere we can probe. As atmospheric density and aerosol loading increase with  
157 decreasing altitude, the solar occultation transmission level often falls from unity to zero  
158 within 5–10 km (see [16, 17]).

159 The impacts of the storm on the physical state of the atmosphere and on the water cycle  
160 have been studied extensively [e.g., 16, 17, 20]. When dust is lifted into the atmosphere,  
161 it absorbs radiation and stores heat. This results in an expansion of the lower layers of  
162 the atmosphere, amplified Hadley cell circulation, deep convection, strongly increased wa-  
163 ter vapour content, and reduced cloud formation. During our period of observation, the  
164 hygropause altitude moved from  $\sim 50$  km to between 50–80 km, the VMR of H<sub>2</sub>O was en-  
165 hanced by around 75–150 ppmv [25, 17], and temperature increases on the order of 30–50 K  
166 were observed [23].

167 These impacts due to dust loading are well-reproduced in GCM simulations [e.g., 26],  
168 and our own simulations show the resulting changes to CO in Fig. 4c. The zonal mean CO  
169 mixing ratio profiles reveal increased Hadley cell circulation: strong upwelling in northern  
170 summer as well as at low-to-mid latitudes in the southern winter atmosphere.

171 If we consider the observations made outside the polar enhancements, we can compute  
172 the mean CO mixing ratio below 40 km for the two time periods (before and during the  
173 dust storm). After the onset of the dust storm, the mean CO mixing ratio value in our



**Fig. 5. | Time evolution of CO VMR vertical profiles** Evolution of the ACS vertical profiles of CO during the MY34 dust storm. Latitudes are between  $-75^\circ$  to  $75^\circ$  and colours indicate Ls.

174 observations has fallen to  $1070 \pm 17$  ppmv from  $1260 \pm 30$  ppmv, which is greater than a 20%  
 175 decrease (95% confidence intervals are 1034–1098 ppmv and 1198–1319 ppmv, respectively).  
 176 These differences are distinctly seen in Fig. 5 and are likely to be due to the large amounts  
 177 of  $\text{H}_2\text{O}$  present during the dust storm and the subsequent accelerated CO loss by reaction  
 178 with OH.

179 At high altitudes, ACS observations of decreased CO mixing ratios after the onset of  
 180 the global dust storm is explained by the same expansion that raised the hygropause level.  
 181 Intense warming and increased pressure produce an upward motion that leads to intensi-  
 182 fied circulation, consistent with our GCM results (Fig. 4). The dramatic increase in the  
 183 hygropause altitude also increases the vertical range over which OH becomes available to  
 184 convert CO into  $\text{CO}_2$ . After the dust storm, we observe CO VMRs that are fairly constant  
 185 until above 100 km, much higher than observations prior to the dust storm and GCM predic-  
 186 tions. This suggests strong vertical mixing, and that the altitude at which the  $\text{CO}_2$  mixing  
 187 ratios are assumed to begin to drop off is elevated.

188 CO is a tracer for upper atmosphere transport and its quantification is also important

189 for understanding the oxidizing power of the Mars atmosphere. These continuing observa-  
190 tions will help us constrain the long-term equilibrium of the Mars atmosphere composition.  
191 The observations presented here, covering an altitude range from the near-surface to above  
192 110 km, provide new insights into both atmospheric dynamics and photochemistry.

193 Further observations will enable us to study the seasonal cycle and ratios of oxygen  
194 isotopes of CO. By incorporating data from other ACS channels and MIR grating positions,  
195 we will be able to compare CO mixing ratio profiles to simultaneous observations of aerosols  
196 and other gases, such as water vapour, ozone, and oxygen, allowing us to investigate chemical  
197 interactions, atmospheric waves or cloud interactions.

## 198 **Data availability**

199 The datasets generated by the ExoMars Trace Gas Orbiter instruments, including ACS, and  
200 analyzed during the current study are being made available in the ESA Planetary Science  
201 Archive (PSA) repository, <https://archives.esac.esa.int/psa>, following a six months prior  
202 access period, following the ESA Rules on Information, Data, and Intellectual Property. Data  
203 used herein can be found by searching for the “ExoMars 2016” mission and then selecting  
204 the ACS and MIR instruments. Derived producted (CO VMR vertical profiles) have been  
205 deposited in the Oxford University Research Archive at: DOI: 10.5287/bodleian:wxxq2m6jo.

## 206 **Code availability**

207 The GGG software suite is maintained by NASA’s Jet Propulsion Laboratory (JPL) and the  
208 California Institute of Technology. GGG is available at <https://tcon-wiki.caltech.edu> and  
209 distributed under a non-commercial software license. The LMD GCM is maintained at LMD.  
210 It can be obtained using the Subversion version control system following the instructions

211 made available here: [https://www.lmd.jussieu.fr/~lmdz/planets/mars/user\\_manual.pdf](https://www.lmd.jussieu.fr/~lmdz/planets/mars/user_manual.pdf)

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## 274 **Acknowledgements**

275 The ExoMars mission is a joint mission of the European Space Agency (ESA) and Roscosmos.  
276 The ACS experiment is led by the Space Research Institute (IKI) in Moscow, assisted by  
277 LATMOS in France. This work was funded by Roscosmos, the National Centre for Space  
278 Studies of France (CNES), the Ministry of Science and Education of Russia, the Natural  
279 Sciences and Engineering Research Council of Canada (NSERC) (PDF - 516895 - 2018),  
280 and the UK Space Agency (ST/T002069/1, ST/R001502/1 and ST/P001572/1). Science  
281 operations are funded by Roscosmos and ESA.

## 282 **Author contributions**

283 Spectral fitting of ACS MIR spectra was performed by KSO using the GGG software suite.  
284 Input and aid on spectral fitting was given by JA, CW, DB, AF, and FM. Processing of ACS  
285 spectra is done at LATMOS by LB and at IKI by AT. AF supplied ACS NIR retrievals of  
286  $P$ - $T$  profiles, and preliminary CO VMR profiles for comparison. The LMD GCM was run  
287 by FL with support from FF. The ACS instrument was operated by AT, AP, AG, and AS.  
288 All co-authors have contributed to the preparation of the manuscript, written by KSO, FL,  
289 FM and OK.

## 290 **Competing interests**

291 The authors declare no competing interests.

## 292 **Figure legends**

293 **Fig. 1 | Spectral fitting with ACS MIR** Example of normalized spectra recorded at  
294 60 km with ACS MIR using secondary grating position 7. Shown are: (a) contributions  
295 from diffraction orders 250–257, (b) example fits for spectral windows covering order 252  
296 (bottom) and the associated residuals (top).

297 **Fig. 2 | Solar occultation latitudes** Distribution over latitude and solar longitude of  
298 ACS MIR occultation using position 7, and all other ACS occultation points. The colour  
299 scale indicates the retrieved CO VMR between 20–40 km. The solar longitudes correspond  
300 to an observational period of April to September 2018.

301 **Fig. 3 | CO VMR vertical profiles at different latitudes** Vertical profiles of  
302 retrieved CO mixing ratio as observed by ACS MIR at: a) equatorial and mid-latitudes  
303 prior to Ls 190°, and b) at polar latitudes greater than  $\pm 75^\circ$ . The shaded areas show the  
304 magnitude of the uncertainty of each retrieved VMR value. Lower altitude ranges are limited  
305 by signal attenuation due to aerosols. Uncertainties are derived from the non-linear least  
306 squares inversion using partial derivatives.

307 **Fig. 4 | Modelled CO distributions** Zonally-averaged CO volume mixing ratio  
308 (ppmv) calculated by the LMD global climate model using the MY 34 dust climatology  
309 [20] for three Ls periods: a) 165–170°, b) 180–185°, c) 210–215°. The onset of the global  
310 dust storm was around Ls 190°. The simulation shown here was initialized at Ls = 180° of  
311 MY33 and therefore avoids known issues with photochemical models underestimating the  
312 long-term equilibrium value of CO.

313 **Fig. 5 | Time evolution of CO VMR vertical profiles** Evolution of the ACS vertical  
314 profiles of CO during the MY34 dust storm. Latitudes are between  $-75^\circ$  to  $75^\circ$  and colours  
315 indicate Ls.