Did the COVID-19 Crisis Reduce Free Tropospheric Ozone across the Northern Hemisphere?

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Key Points (shortened to less than 140 characters each, and changed as suggested by 56 **Reviewer #2):** 57

- In spring and summer 2020, stations in the northern extratropics report on average 7% (4 58 nmol/mol) less tropospheric ozone than normal. 59
- Such low tropospheric ozone, over several months, and at so many sites, has not been 60 observed in any previous year since at least 2000. 61
- Most of the reduction in tropospheric ozone in 2020 is likely due to emissions reductions 62 related to the COVID-19 pandemic. 63
- 64

65 Abstract

66 Throughout spring and summer 2020, ozone stations in the northern extratropics recorded

unusually low ozone in the free troposphere. From April to August, and from 1 to 8 kilometers

altitude, ozone was on average 7% (\approx 4 nmol/mol) below the 2000 to 2020 climatological mean.

69 Such low ozone, over several months, and at so many stations, has not been observed in any

70 previous year since at least 2000. Atmospheric composition analyses from the Copernicus

Atmosphere Monitoring Service and simulations from the NASA GMI model indicate that the large 2020 springtime ozone depletion in the Arctic stratosphere contributed less than one

large 2020 springtime ozone depletion in the Arctic stratosphere contributed less than one
 quarter of the observed tropospheric anomaly. The observed anomaly is consistent with recent

chemistry-climate model simulations, which assume emissions reductions similar to those caused

by the COVID-19 crisis. COVID-19 related emissions reductions appear to be the major cause

⁷⁶ for the observed reduced free tropospheric ozone in 2020.

77

78 Plain Language Summary

79 Worldwide actions to contain the COVID-19 virus have closed factories, grounded airplanes, and have generally reduced travel and transportation. Less fuel was burnt, and less exhaust was 80 emitted into the atmosphere. Due to these measures, the concentration of nitrogen oxides and 81 volatile organic compounds (VOCs) decreased in the atmosphere. These substances are 82 important for photochemical production and destruction of ozone in the atmosphere. In clean or 83 mildly polluted air, reducing nitrogen oxides and/or VOCs will reduce the photochemical 84 production of ozone and result in less ozone. In heavily polluted air, in contrast, reducing 85 nitrogen oxides can increase ozone concentrations, because less nitrogen oxide is available to 86 destroy ozone. In this study, we use data from three types of ozone instruments, but mostly from 87 ozonesondes on weather balloons. The sondes fly from the ground up to 30 kilometers altitude. 88 In the first 8 kilometers, we find significantly reduced ozone concentrations in the northern 89 extratropics during spring and summer of 2020, less than in any other year since at least 2000. 90 We suggest that reduced emissions due to the COVID-19 crisis have lowered photochemical 91 ozone production and have caused the observed ozone reductions in the troposphere. 92

93

94 **1 Introduction**

95 Widespread measures to contain the COVID-19 pandemic have slowed, or even closed down, industries, businesses, and transportation activities, and have reduced anthropogenic 96 emissions substantially throughout the year 2020. Guevara et al. (2020), or Barré et al. (2020) 97 report European emissions reductions up to 60% for NOx, and up to 15% for Non-Methane 98 Volatile Organic Compounds (NMVOC) in March/April 2020. Based on satellite observations of 99 NO₂ columns (Bouwens et al., 2020), comparable NO_x emissions reductions are reported for 100 Chinese cities in February 2020 (Ding et al., 2020; Feng et al., 2020). Globally averaged CO₂ 101 emissions decreased by 8.8% during the first half of 2020 (Liu et al., 2020), consistent in timing 102 and magnitude with the aforementioned NO₂ emission reductions. The largest relative reductions 103 104 occurred for air traffic, where emissions decreased by $\approx 40\%$, on average, in the first half of 2020 (Le Quéré et al., 2020a; Liu et al., 2020), and remained low during the second half of 2020 (Le 105

106 Quéré et al., 2020b).

107 These COVID-19 emissions reductions are large enough to affect ozone levels in the 108 troposphere (Dentener et al., 2011). Tropospheric O₃-NO_x-VOC-HO_x chemistry is, however,

- 109 complex and nonlinear. The net effect of emission changes depends on NO_x and VOC
- 110 concentrations (e.g., Kroll et al., 2020; Sillman, 1999; Thornton et al., 2002). In polluted regions,
- 111 at high NO_x concentrations (>> 1pbb), reducing NO_x concentrations can increase ozone, because
- ozone titration by NO is reduced (e.g., Sicard et al., 2020). At low concentrations (NO_x < 1 nmol/mol), however, in the clean or mildly polluted free troposphere, reducing NO_x lowers
- 113 nmol/mol), however, in the clean or mildly polluted free troposphere, reducing NO_x lov 114 photochemical ozone production (e.g., Bozem et al., 2017), and results in less ozone.

Indeed, for many polluted regions, studies report increased near-surface ozone after

116 COVID-19 lockdowns (e.g., Collivignarelli et al., 2020; Lee et al., 2020; Shi & Brasseur, 2020;
117 Siciliano et al., 2020; Venter et al., 2020). Reduced surface ozone is reported for some rural
118 areas, e.g., in the US and Western Europe (Chen et al., 2020; Menut et al., 2020). Meteorological
119 conditions complicate matters, as they play an important role as well (Goldberg et al., 2020;

120 Keller et al., 2020; Ordóñez et al., 2020; Shi & Brasseur, 2020).

121 In the free troposphere, ozone is an important greenhouse gas, and plays a key role in tropospheric chemical reactions, controlling the oxidizing capacity (e.g. Archibald et al., 2020; 122 Cooper et al., 2014; Gaudel et al, 2018). The Northern Hemisphere free troposphere is dominated 123 by net photochemical ozone production, proportional (albeit nonlinearly) to the availability of 124 ozone precursor gases (e.g., Zhang et al., 2020). In contrast to increases of surface ozone in 125 polluted urban areas after the COVID-19 emissions reductions, we find significant reductions of 126 127 ozone in the northern extratropical free troposphere. These large-scale reductions occurred in late spring and summer 2020, following the widespread COVID-19 slowdowns, and are unique 128 129 within the last two decades.

130 2 Instruments and Data

Regular observations of ozone in the free troposphere are sparse: Only around 50 ozone 131 sounding stations worldwide (e.g. Tarasick et al., 2019), a handful of tropospheric lidars (Gaudel 132 et al., 2015; Leblanc et al., 2018), and about twenty Fourier Transform Infrared Spectrometers 133 (FTIRs, Vigouroux et al., 2015). In-Service Aircraft for a Global Observing System (IAGOS, 134 Nédélec et al., 2015) are another important source of tropospheric ozone data. Due to the 135 COVID-19 slowdowns, however, few IAGOS aircraft were flying in 2020, and IAGOS data 136 became quite sparse, with only about 20 flights per month since April 2020, compared to more 137 than 200 flights per month in 2019. The information content of satellite measurements on ozone 138 in the free troposphere is limited, and accuracy is modest, 10 to 30% (Hurtmans et al., 2012; Liu 139 et al., 2010; Oetjen et al., 2014). The recent Tropospheric Ozone Assessment Report found large 140 differences in tropospheric ozone trends derived from different satellite instruments, and even 141

- 142 different signs in some regions (Gaudel et al., 2018).
- Ozonesondes measure profiles with high vertical resolution, about 100 m, and good accuracy, 5 to 15% in the troposphere, 5% in the stratosphere (Smit et al., 2007; Sterling et al., 2018; Tarasick et al., 2016; Van Malderen et al., 2016; Witte et al., 2017; WMO, 2014). This is adequate to detect ozone anomalies of several percent. We use stations with regular soundings, at least once per month since the year 2000, and with data available until at least July 2020.
- 148 Soundings with obvious deficiencies were rejected (i.e. large data gaps, integrated ozone column
- from the sounding deviating by more than 30% from ground- or satellite-based spectrometer
- 150 measurement). Table 1 provides information on stations, and public data archives.

152

- 153 **Table 1.** Stations in this study, mostly ozonesonde stations. *FTIR and LIDAR stations are*
- 154 *italicized*. Data sources: **W**=World Ozone and UV Data Centre
- 155 (<u>https://woudc.org/archive/Archive-NewFormat/OzoneSonde_1.0_1/</u>), N=Network for the
- 156 Detection of Atmospheric Composition Change (<u>ftp://ftp.cpc.ncep.noaa.gov/ndacc/station/;</u>
- 157 <u>ftp://ftp.cpc.ncep.noaa.gov/ndacc/RD/</u>), **E**= European Space Agency Validation Data Center
- 158 (<u>https://evdc.esa.int/</u> requires registration, or
- 159 <u>ftp://zardoz.nilu.no/nadir/projects/vintersol/data/o3sondes</u> requires account), G=Global
- 160 Monitoring Laboratory, National Oceanic and Atmospheric Administration
- 161 (<u>ftp://aftp.cmdl.noaa.gov/data/ozwv/Ozonesonde/</u>)
- ¹Due to COVID-19 restrictions, most Canadian ozonesonde data were available only up to March or April 2020.
- ² Tateno data were corrected for the change from Carbon Iodine to ECC ozonesondes in December 2009.

 3 Stations affected by a drop-off in ECC sonde sensitivity > 3% in the stratosphere, after 2015 (see Stauffer et al.,

2020). The drop-off is much smaller (< 1%) in the troposphere, and should be negligible here. At many of the

affected stations, ECC sondes behaved normally again in 2019/2020.

Station	Latitude (deg N)	Longitude (deg E)	Data source (see caption)	Data until	Profiles / spectra per month in 2020
Alert, Canada ^{1,3}	82.50	-62.34	W	4/2020	3.75
Eureka, Canada ³	80.05	-86.42	W, E	9/2020	4.89
Ny-Ålesund, Norway	78.92	11.92	W, E	10/2020	7.10
Ny-Ålesund FTIR, Norway	78.92	11.92	Ν	7/2020	12.86
Thule FTIR, Greenland	76.53	-68.74	Ν	9/2020	73
Resolute, Canada ¹	74.72	-94.98	W	4/2020	5.50
Scoresbysund, Greenland	70.48	-21.95	Е	11/2020	4.00
Kiruna FTIR, Sweden	67.41	20.41	N	7/2020	46
Sodankylä, Finland	67.36	26.63	W, E	12/2020	2.83
Lerwick, United Kingdom	60.13	-1.18	W, E	12/2020	3.92
Churchill, Canada ^{1,3}	58.74	-93.82	W	3/2020	3.33
Edmonton, Canada ^{1,3}	53.55	-114.10	W	3/2020	3.67
Goose Bay, Canada ¹	53.29	-60.39	W	3/2020	2.67
Bremen FTIR, Germany	53.13	8.85	N	10/2020	5.27
Legionowo, Poland	52.40	20.97	W	10/2020	4.00
Lindenberg, Germany	52.22	14.12	W	11/2020	4.73
DeBilt, Netherlands	52.10	5.18	W, E	12/2020	4.33
Valentia, Ireland	51.94	-10.25	W, E	12/2020	2.50

Uccle, Belgium	50.80	4.36	W, E	12/2020	12.00
Hohenpeissenberg, Germany	47.80	11.01	W	12/2020	10.50
Zugspitze FTIR, Germany	47.42	10.98	N	9/2020	73
Jungfraujoch FTIR, Switzerland	46.55	7.98	N	12/2020	46
Payerne, Switzerland	46.81	6.94	W	10/2020	11.10
Haute Provence, France	43.92	5.71	Ν	8/2020	2.50
Haute Provence LIDAR, France	43.92	5.71	N	8/2020	3.50
Toronto FTIR, Canada	43.66	-79.40	N	10/2020	59
Trinidad Head, California, USA	41.05	-124.15	G	12/2020	3.58
Madrid, Spain	40.45	-3.72	W	11/2020	4.09
Boulder, Colorado, USA	39.99	-105.26	G	12/2020	4.83
Boulder FTIR, Colorado, USA	39.99	-105.26	N	10/2020	56
Tateno (Tsukuba), Japan ²	36.05	140.13	W	10/2020	2.70
Table Mountain LIDAR, California, USA	34.40	-117.70	Ν	8/2020	19
Izana, Tenerife, Spain	28.41	-16.53	W	8/2020	2.00
Izana FTIR, Tenerife, Spain	28.30	-16.48	N	9/2020	28
Hong Kong, China	22.31	114.17	W	9/2020	4.11
Hilo, Hawaii, USA ³	19.72	-155.07	G	12/2020	4.08
Mauna Loa FTIR, Hawaii, USA	19.54	-155.58	N	10/2020	36
Paramaribo, Suriname	5.81	-55.21	N, E	10/2020	3.60
Pago Pago, American Samoa ³	-14.25	-170.56	G	12/2020	3.08
Suva, Fiji ³	-18.13	178.32	G	9/2020	1.44
Wollongong FTIR, Australia	-34.41	150.88	N	10/2020	43
Broadmeadows, Australia	-37.69	144.95	W	7/2020	4.29
Lauder, New Zealand	-45.04	169.68	W	10/2020	4.40
Lauder FTIR, New Zealand	-45.04	169.68	N	10/2020	99
Macquarie Island, Australia	-54.50	158.94	W	7/2020	4.29

Apart from the sondes, FTIR spectrometers from the Network for the Detection of 169 Atmospheric Composition Change (NDACC, De Mazière et al., 2018) provide independent 170 171 information, based on a completely different method (ground-based solar-infrared absorption spectrometry). The altitude resolution of FTIR ozone profiles in the troposphere is much coarser 172 (5 to 10 km) than that of the sondes, while accuracy is similar, 5 to 10% (Vigouroux et al., 2015). 173 Finally, we use data from tropospheric lidars (Gaudel et al., 2015, Granados-Muñoz & Leblanc 174 2016), which provide ozone profiles from \approx 3 to 12 km altitude, with accuracy comparable to the 175 sondes (5 to 10%; Leblanc et al., 2018), and slightly coarser altitude resolution (100 m to 2 km). 176 We also use global atmospheric composition re-analyses from the Copernicus 177

Atmosphere Monitoring Service for the years 2003 to 2019, and operational analyses for the year

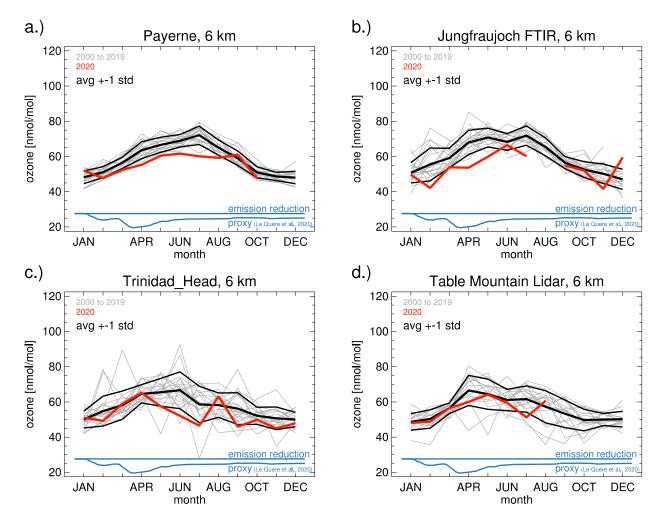
- 179 2020 (CAMS, Inness et al., 2019; see also Park et al., 2020). The CAMS data are taken at the
- 180 grid-points closest to the stations in Table 1. The analyses (in 2020) are adjusted for the small
- average difference to the re-analyses in 2018 and 2019. CAMS (re-)analyses are based on
- 182 meteorological fields, and assimilation of satellite observations of ozone and NO₂. However, for
- NO₂ the impact of the assimilation is small and frequently insignificant, so that tropospheric NO_x in CAMS is essentially controlled by the prescribed emissions (Inness et al., 2019). Similarly, the
- limited information content of current satellite measurements of tropospheric ozone means that
- 186 tropospheric ozone in CAMS is also driven largely by the prescribed emissions (and the
- 187 chemistry module). Stratospheric ozone, however, is constrained well by the assimilated satellite
- data. Thus, CAMS analyses account for the large Arctic stratospheric depletion in spring of 2020
- 189 (Manney et al., 2020; Wohltmann et al., 2020), for 2020 meteorological conditions, and for
- ozone transport, e.g. from the stratosphere to the troposphere (Neu et al., 2014). However, since
- 191 they rely on "business as usual" emissions for 2020, the CAMS analyses do not account for the
- 192 effects of COVID-19 emissions reductions in 2020 on tropospheric ozone (and NO_x).

193 **3 Results**

For selected stations, Fig. 1 presents the annual cycles of tropospheric ozone over the last 20 years, at 6 km, a representative altitude for the free troposphere. Monthly means (over 1-km wide layers) reduce synoptic meteorological variability and measurement noise, and focus on longer-term, larger-scale variations.

Payerne, Jungfraujoch, and Trinidad Head show an annual cycle with low ozone in
winter and high ozone in summer. This is the case for most stations in the northern extratropics
(Cooper et al., 2014; Gaudel et al., 2018; Parrish et al., 2020). Increased photochemical
production due to more sunlight and warmer temperatures is the main driver for the summer
ozone maximum in the northern extratropics (Wu et al., 2007; Archibald et al., 2020).

Figure 1 shows substantial yearly variability, but ozone levels are notably below average 203 in 2020, at all four stations (thick red lines in Fig. 1). At Payerne and Jungfraujoch, and a number 204 of other stations, monthly means in spring and summer 2020 were actually the lowest, or close to 205 the lowest, since 2000. For context, the dark blue lines in Fig. 1 provide global CO₂ emission 206 reductions due to the COVD-19 pandemic (Le Quéré et al., 2020b). Comparable reductions 207 apply to global ozone precursor emissions (NOx and VOCs). The (daily) emission proxy in Fig. 1 208 indicates that the largest effect for ozone might be expected after March 2020. However, Fig. 1 209 does not show any clear or close correspondence between unusual ozone monthly means in 2020 210 (red lines) and the emission reduction proxy (dark blue lines). 211



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Figure 1. Observed ozone monthly means at four typical stations. Results are for 6 km altitude. The thick red line highlights the year 2020. Climatological averages, and standard deviations over the years 2000 to 2020 are indicated by the thick black lines. Payerne (a) and Trinidad Head (c) are sonde stations. Jungfraujoch (b) is an FTIR station. Table Mountain (d) is a lidar station. Dark blue lines: CO₂ emission reductions (arbitrary units) from Le Quéré et al. (2020b), as a proxy for ozone precursor reductions in 2020.

Annual cycles of ozone anomalies, averaged over all northern extratropical stations 221 (stations north of 15°N), are shown in Fig. 2. Anomalies were defined as the relative deviation 222 (in percent) from the 2000-2020 climatological mean of each calendar month at each station. As 223 for the single stations in Fig. 1, the observed northern extratropical average shows exceptionally 224 low ozone throughout spring and summer 2020 (red line in Fig. 2a). This is not reproduced by 225 the CAMS analyses, which do not account for COVID-19 related emissions reductions, and 226 simulate ozone in the usual range in 2020 (red line in Fig. 2b). Again, there is no close temporal 227 correspondence between the unusual behavior of observed ozone in 2020 (red line in Fig. 2a), 228 and the emissions proxy (dark blue line in Fig. 2a). 229

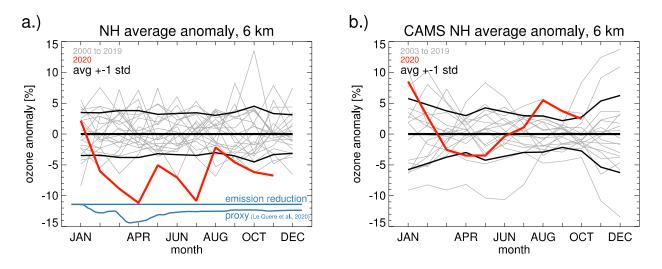


Figure 2. Annual cycles of monthly mean northern extratropical ozone anomalies at 6 km 232 altitude. Anomalies are in percent, relative to the climatological monthly mean calculated for 233 each station/instrument, and for the period 2000 to 2020 (all Januaries, all Februaries, ..., all 234 Decembers). These single station/instrument anomalies are then averaged over all northern 235 extratropical stations/instruments (north of 15°N). Panel a) Results from the station observations. 236 Panel b) Results for CAMS atmospheric composition (re-)analyses at grid points nearest the 237 stations. The CAMS data do not account for COVID-19 related emissions reductions in 2020. 238 Grey lines: individual years from 2000 to 2019. Thick red line: year 2020. Thick black lines: 239 240 average anomaly, ± 1 standard deviation over the years. Dark blue lines in panel a): Global CO₂ emission reductions in 2020 (arbitrary units) from Le Ouéré et al. (2020b), as in Fig. 1. 241

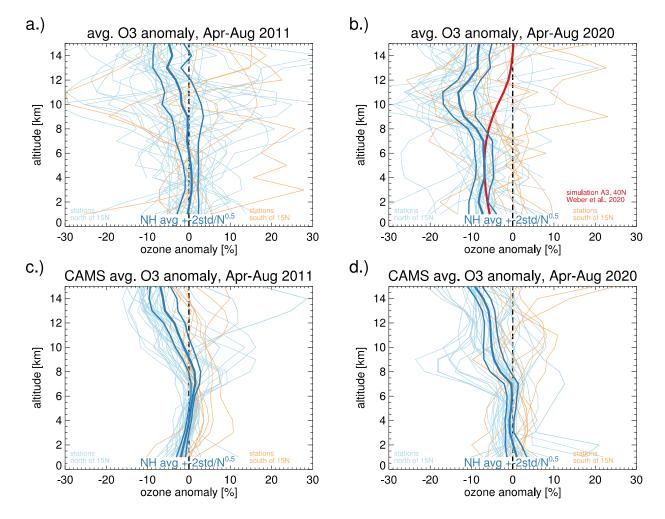
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Figs. 1 and 2 show large negative anomalies from April to August 2020. Fig. 3 compares 243 anomaly profiles averaged over those five calendar months, between the years 2011 and 2020. 244 Both years saw unusually large springtime ozone depletion in the Arctic stratosphere (Manney et 245 al., 2020; Wohltmann et al., 2020). In the stratosphere, above ≈ 10 km, the Arctic depletion 246 appears as low ozone, both in observations and CAMS results (particularly for stations north of 247 50°N). In both the stratosphere and the troposphere, the observed profiles show more variability 248 than the smoother CAMS profiles. In 2020, most observed single station anomaly profiles (Fig. 249 250 3b) are negative throughout the northern extratropical troposphere (between 1 and 10 km). This is not the case in 2011 (Fig. 3a, 3c), nor in the CAMS data in 2020 (Fig. 3d). 251

The 2020 anomaly is even clearer for the northern extratropical mean profile (dark blue lines in Fig. 3). The observed 2020 mean anomaly profile is large, -6% to -9%, and statistically significant at the 95% level (more than 99% in fact) from 1 to 8 km (Fig. 3b), whereas the corresponding CAMS profile is close to zero (Fig. 3d). Fig. 3 indicates that Arctic stratospheric springtime ozone depletion did not have a large effect on tropospheric ozone below 8 km in 2011 and 2020 (see also Fig. S1 in the supplement), and that the CAMS "business as usual" simulation does not account for the observed large negative tropospheric anomaly in 2020.

Fig. 3b also shows a simulated profile of tropospheric ozone reduction from a recent chemistry-climate modelling study of COVID-like emissions decreases by Weber et al. (2020). This simulated profile (red line in our Fig 3b) matches the observed northern extratropical ozone reduction (dark blue line), from the ground up to about 8 km. Above 8 km, the simulated profile deviates by $\approx 10\%$ from the observed profile, because it assumes fixed 2012 to 2014

- 264 meteorological conditions. The CAMS analyses (Fig. 3d) show that 2020 meteorological
- conditions and springtime Arctic stratospheric ozone depletion resulted in ozone reductions of
- 266 5% to 10% above 9 km, consistent with the observations.



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Figure 3. Ozone anomaly profiles (in percent), averaged over April to August. Stations are 268 excluded in years where their data cover less than three of these five months. Panel a) for the 269 year 2011. Panel b) for the year 2020. Light blue lines: northern extratropical stations (north of 270 15°N). Light orange lines: remaining stations, south of 15°N. Thick dark blue line: mean of the 271 northern extratropical stations. Thin dark blue lines: 95% confidence interval of the mean of the 272 273 northern extratropical stations. Red line in panel b): simulated ozone change at 40°N from Weber et al. (2020; Fig. S4, scenario A3). Panels c), d): Same as a), b), but for CAMS (re-)analyses at 274 the grid-points closest to the stations. 275

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Time series of the tropospheric anomaly (averaged from April to August, and from 1 to 8 km altitude) are shown in Fig. 4. In the observations (left panel), the year 2020 stands out with 279 large negative anomalies, not seen in the CAMS data. Across the twenty previous years, ozone 280 anomalies at individual stations (thin lines) are scattered around zero. The northern extratropical 281 average anomaly (dark blue line) is usually smaller than $\pm 3\%$. The only other observed exception

is the positive anomaly related to the (European) heat-wave summer of 2003 (Vautard et al.,

283 2007). The large negative northern extratropical anomaly in the observations in 2020, \approx -7%, is

clearly outside of the $\pm 2\sigma$ range of the previous 20 years (thin dark blue lines). It is not

reproduced by the CAMS "emissions as usual" analysis.

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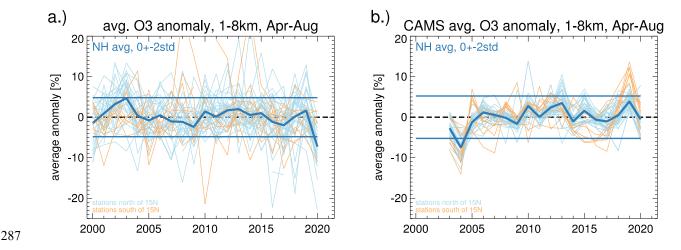


Figure 4. Tropospheric ozone anomaly, averaged over April to August and from 1 to 8 km, for
the years 2000 to 2020. Panel a) Observations. Panel b) CAMS atmospheric composition (re)analyses. Light blue lines: northern extratropical stations (north of 15°N). Light orange lines:
stations south of 15°N. Thick dark blue line: Average over all stations north of 15°N. Thin dark
blue lines: ±2 standard deviations over all years of this average.

293

The geographic distribution of the average tropospheric ozone anomalies is shown for 294 2011 and 2020 in Fig. 5. 2020 stands out in the observations with large negative anomalies at 295 nearly all northern extratropical stations, and a fairly uniform geographical distribution (see 296 Table S1 of the supplement for the numerical values). CAMS does show negative anomalies in 297 2020, but only north of 50°N, and not as large as the observations. In the Southern Hemisphere 298 in 2020, agreement between observations and CAMS is quite good, typically within 2.5% or 299 better (see also Table S1 in supplement). In 2011, some stations show positive anomalies, 300 negative anomalies are not as large as in 2020, and the geographical distribution is less uniform. 301 Agreement between observations and CAMS is reasonable in 2011, usually within a few percent. 302

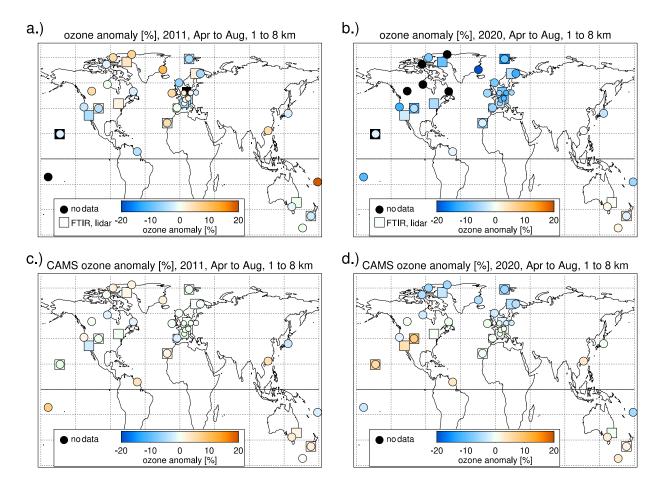


Figure 5. Geographic distribution of observed tropospheric ozone anomalies (averaged over the months April to August, and over altitudes from 1 to 8 km) for the years **a**) 2011 and **b**) 2020. Panels **c**) and **d**): same, but for CAMS results at the station locations. Colored circles give the anomaly at the ozonesonde stations. Squares are for FTIR and lidar stations. See Table S1 of the supplement for the numerical values. Black filling indicates insufficient data in the given year.

309

310 4 Discussion and Conclusions

Ozone stations in the northern extratropics indicate exceptionally low ozone in the free 311 troposphere (1 to 8 km) in spring and summer 2020. Compared to the 2000-2020 climatology, 312 ozone was reduced by 7% (\approx 4 nmol/mol). Such widespread low tropospheric ozone, across so 313 314 many stations and over several months has not been observed in any previous year since 2000. The observed 7% ozone reduction in the free troposphere stands in contrast to increases of 315 surface ozone by 10% to 30%, reported for many polluted urban areas after the COVID-19 316 related emissions reductions in 2020 (e.g., Collivignarelli et al., 2020; Lee et al., 2020; Shi & 317 Brasseur, 2020; Siciliano et al., 2020; Venter et al., 2020). However, the chemical regime for 318 ozone in the free troposphere is different (e.g., Kroll et al., 2020; Sillman, 1999; Thornton et al., 319 2002), and free tropospheric ozone reductions are expected after the substantial decrease of 320 precursor emissions due to the COVID-19 pandemic (e.g. Guevara et al., 2020; Zhang et al., 321 2020). 322

Recent model simulations of COVID-like emissions decreases (Weber et al., 2020) find tropospheric ozone reductions very similar to our observational results. From our results, and the simulations by Weber et al., 2020, it appears that the total tropospheric ozone burden of the northern extratropics decreased by about 7% for April to August 2020. The contribution from ozone increases in polluted urban areas to the total burden is opposite, but very small.

The Weber et al. (2020) simulations indicate that the major causes of tropospheric ozone reduction come from reduced surface transportation (ozone decrease throughout most of the northern extratropical troposphere), and from reduced aviation (ozone decrease mostly between 10 and 12 km altitude and north of 30°N, see also Grewe et al., 2017). While the simulations are qualitatively consistent with the observations, they consider only March to May. New simulations using more recent and extended emissions estimates (Le Quéré et al., 2020b; Liu at al., 2020), and further comparison with our station observations would be worthwhile.

The observed large and fairly uniform 7% reduction of ozone in the northern extratropical troposphere in spring and summer 2020 provides a far reaching test case for the response of tropospheric ozone to emission changes. Further quantification of this anomaly will

be possible, when observations from commercial aircraft (IAGOS), and satellite instruments

become available. Additional modelling studies will improve our understanding of the

contributions from different sectors such as air traffic, and surface transportation.

341

342

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385 Data Sources

- 386 Most of the ozonesonde data used in this study are freely available from the World Ozone and
- 387 UV Data Centre (<u>https://woudc.org</u>) at Environment Canada (<u>https://exp-studies.tor.ec.gc.ca/</u>),
- and are downloadable at <u>https://woudc.org/archive/Archive-NewFormat/OzoneSonde_1.0_1/</u>).
- 389 Some ozonesonde data for 2020 were not yet available at the WOUDC. Instead, rapid delivery
- data were obtained from <u>ftp://zardoz.nilu.no/nadir/projects/vintersol/data/o3sondes</u> (requires
- registration), at the Nadir database of the Norwegian Institute for Air Quality (NILU,
- 392 <u>https://projects.nilu.no/nadir/obs.html</u>). Registration information, and the same data in a
- different format, are available from the European Space Agency Validation Data Center
- 394 (<u>https://evdc.esa.int/</u>).
- ³⁹⁵ For Boulder, Trinidad Head, Hilo, Fiji, and Samoa, stations operated by the US National Oceanic
- 396 and Atmospheric Administration, Global Monitoring Laboratory
- 397 (<u>https://www.esrl.noaa.gov/gmd/ozwv/</u>), data can be obtained freely from
- 398 <u>ftp://aftp.cmdl.noaa.gov/data/ozwv/Ozonesonde/</u>.
- FTIR and lidar data, as well as some ozonesonde data, are from the Network for the Detection of
- 400 Atmospheric Composition Change (<u>https://ndacc.org</u>), and are freely available at
- 401 <u>ftp://ftp.cpc.ncep.noaa.gov/ndacc/station/</u> and <u>ftp://ftp.cpc.ncep.noaa.gov/ndacc/RD/</u>.

- 402 Copernicus Atmosphere Monitoring Service (CAMS) global chemical weather EAC4 re-
- 403 analyses are available at <u>https://atmosphere.copernicus.eu/data</u> . CAMS operational global
- 404 analyses and forecasts are available at <u>https://apps.ecmwf.int/datasets/data/cams-nrealtime/</u>.

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