

1 **Global Changes in Secondary Atmospheric Pollutants during the 2020 COVID-19** 2 **Pandemic**

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17 **Key Points:**

- 18
- 19 • During the Covid-19 lockdown, the atmospheric concentration of primary pollutants (NO_x,
VOCs, CO, SO₂) was considerably reduced.
 - 20 • The concentration of secondary pollutants increased in NO_x-saturated areas and decreased
21 in NO_x-limited areas.
 - 22 • The response of the chemical system depends strongly on weather variability and on the
23 relative changes in NO_x and VOCs emissions.

Abstract

We use the global Community Earth System Model to investigate the response of secondary pollutants (ozone O₃, secondary organic aerosols SOA) in different parts of the world in response to modified emissions of primary pollutants during the COVID-19 pandemic. We quantify the respective effects of the reductions in NO_x and in VOC emissions, which, in most cases, affect oxidants in opposite ways. Using model simulations, we show that the level of NO_x has been reduced by typically 40 % in China during February 2020 and by similar amounts in many areas of Europe and North America in mid-March to mid-April 2020, in good agreement with space and surface observations. We show that, relative to a situation in which the emission reductions are ignored and despite the calculated increase in hydroxyl and peroxy radicals, the ozone concentration increased only in a few NO_x-saturated regions (northern China, northern Europe and the US) during the winter months of the pandemic when the titration of this molecule by NO_x was reduced. In other regions, where ozone is NO_x-controlled, the concentration of ozone decreased. SOA concentrations decrease in response to the concurrent reduction in the NO_x and VOC emissions. The model also shows that atmospheric meteorological anomalies produced substantial variations in the concentrations of chemical species during the pandemic. In Europe, for example, a large fraction of the ozone increase in February 2020 was associated with meteorological anomalies, while in the North China Plain, enhanced ozone concentrations resulted primarily from reduced emissions of primary pollutants.

43

Plain Language Summary

With the reduction in economic activities following the COVID-19 pandemic outbreak in early 2020, most emissions of air pollutants (i.e., nitrogen oxides (NO_x), carbon monoxide (CO), sulfur dioxide (SO₂), volatile organic carbon (VOC), black carbon (BC), organic carbon(OC)) have decreased substantially during several months in different regions of the world. This unintended global experiment has given insight on some of the processes that control air quality and offered a glimpse into a potential future in which air quality would be improved. Here, a global atmospheric model is used to assess the changes in the chemical composition of the atmosphere during the pandemic period and in the related chemical processes that lead to the formation of ozone (O₃) and secondary organic aerosols (SOA). The study illustrates the nonlinearity of the air quality response to reduced NO_x and VOC emissions, which depends on the chemical environment including the background level of nitrogen oxides. Meteorological variability can lead to anomalies in the

56 concentration of chemical species with magnitudes that are as large or even larger than the
57 perturbations due to COVID-induced changes in the emissions.

58

59 **1. Introduction**

60

61 With the development of the COVID-19 pandemic and the resulting slowdown in economic
62 activity, first in China and then in the rest of the world, anthropogenic emissions of primary
63 pollutants were significantly altered for several months. This unanticipated planet-wide
64 experiment allows us to examine the response of the atmosphere's chemical system and in
65 particular, the formation of secondary compounds such as ozone (O₃) and the fraction of the
66 airborne particles including PM_{2.5} (particles with a diameter smaller than 2.5 micrometers) that is
67 produced in situ. It offers a glimpse into a potential future in which air quality would be improved
68 following structural regulations in the emissions of nitrogen oxides (NO_x), carbon monoxide (CO),
69 and volatile organic compounds (VOCs). A reduction in the emissions of the pollutants is expected
70 to modify the level of photo-oxidants present in the atmosphere and the formation of secondary
71 species including ozone (O₃) or secondary organic aerosols (SOA) (Miyazaki et al., 2020; Kroll et
72 al., 2020; Huang et al., 2020). In some polluted geographical areas, the large decrease observed in
73 NO_x concentrations might have shifted the chemical regimes from NO_x-saturated towards NO_x-
74 sensitive conditions. A better understanding of the chemical processes that determine the oxidative
75 potential of the atmosphere and their disruption during the pandemic is therefore useful in
76 developing adequate measures to improve air quality.

77

78 The pandemic manifested itself first in China where the first lockdown measures were adopted
79 from the end of January to the month of March. In Europe, North and South America as well as
80 India and the Middle East, lockdowns were imposed with varying degrees of stringency from
81 March onwards and lasted at least until June.

82

83 Observations by spaceborne and ground-based instruments during the first months of 2020
84 highlighted a substantial decrease in the atmospheric concentrations of NO₂ relative to
85 measurements performed during the same period in 2019 (e.g., Bauwens et al., 2020; Le et al.,
86 2020; Liu et al., 2020; Shi and Brasseur, 2020), relative to longer-term averaged data (e.g.,
87 Deroubaix et al., 2020) or relative to model-based weather benchmarks (Venter et al. 2020; Keller
88 et al., 2020). Numerous specific studies analyzing on air quality anomalies have focused on

89 specific regions or urban areas (e.g., Baldasano, 2020; Bedi et al., 2020; Chauhan and Singh, 2020;
90 Fu et al., 2020a; He et al., 2020; Krecl et al., 2020; Menut et al., 2020; Otmani et al., 2020;
91 Rodriguez-Urrego and Rodriguez-Urrego, 2020; Sicard et al., 2020; Siciliano et al., 2020; Zangari
92 et al., 2020 among many others). A large fraction of the observed reductions in air pollutant
93 emissions has been attributed to a drastic disruption in road traffic and in manufacturing
94 operations. In the city of Wuhan, where the pandemic started and very strict lockdown measures
95 were imposed to the entire population, NO₂ and PM_{2.5} concentrations were reduced by
96 approximately 50-60% and 30-40%, respectively, while a large positive anomaly was reported in
97 the concentration of surface ozone (Shi and Brasseur, 2020; Lian et al., 2020; Fu et al., 2020b).
98 For the North China Plain (NCP), the ozone increase was estimated to be larger than 40% (Huang
99 et al., 2020; Shi and Brasseur, 2020; Zhu et al., 2020). Compared to the first months of 2019,
100 measurements made by the spaceborne TROPOMI instrument onboard the Sentinel-5 Precursor
101 satellite in early 2020 showed a decrease in the NO₂ column of typically 40-50% during the
102 lockdown in northern China (Bauwens et al., 2020). Using TROPOMI data, Miyazaki et al. (2020)
103 estimated a reduction of Chinese NO_x emissions reaching 36% from early January to mid-
104 February 2020. Several studies assessing the impact of the COVID-19 pandemic on the emissions
105 of greenhouse gases showed for example that the emission of CO₂ decreased by about 11 to 25 %
106 in April 2020 relative to the mean 2019 levels (Le Quéré et al., 2020; Forster et al., 2020). To
107 analyze observational data during the pandemic, all the reported numbers must be disentangled
108 from the long-term changes in pollutant emissions associated, for example, with air quality and
109 climate policies, multi-scale meteorological variability and the occurrence of occasional societal
110 events such as the New Year festivals in China. The need to consider the influence of weather
111 variability (i.e., anomalies in temperature, humidity, circulation, cloudiness, boundary layer
112 stability) during the pandemic has been highlighted by Diamond and Wood (2020), Barré et al.
113 (2020), Deroubaix et al. (2020), Liu and Wang (2020), Ordóñez et al. (2020), Wang and Zhang
114 (2020) and several other authors. Models have the advantage that they can isolate these different
115 effects and derive the response of the atmosphere to the specific forcing mechanisms.

116

117 In this study, we use a global earth system model with a comprehensive representation of
118 atmospheric gas phase and aerosol chemistry to analyze the importance of the chemical and
119 meteorological processes that have led to a change in the surface concentrations of primary
120 pollutants (e.g., NO_x, CO, VOCs, SO₂, organic and black carbon), secondary photo-oxidants (e.g.,

121 ozone and radicals such OH, HO₂, RO₂, where R is an organic chain such as CH₃ or C₂H₅) and
122 aerosol particles in several regions of the world in response to the reduced emissions of volatile
123 organic compounds, carbon monoxide and nitrogen oxides during the period January to May 2020.
124 The subsequent situation, linked to the onset of a second wave of the pandemic in late 2020, is not
125 considered in this work.

126

127 To quantify the role of different processes that affected the level of pollutants during the COVID-
128 19 pandemic period, different components of the atmospheric system must be carefully examined
129 (Kroll et al., 2020):

- 130 (1) the changes in the emissions of primary pollutants resulting from the reduction in economic
131 activities; these include primarily an abrupt disruption in road, air and maritime traffic as
132 well as in industrial activities, but a possible increase in domestic activities;
- 133 (2) the changes in chemical regimes and specifically in the formation rate of secondary
134 pollutants associated, for example with a shift from VOC to NO_x controlled conditions,
135 and in the formation of ozone and secondary organic aerosols under lower NO_x levels;
- 136 (3) the changes in the concentration and chemical composition of particulate matter;
- 137 (4) the changes in meteorological factors including temperature, humidity, dynamical
138 variability, boundary layer physics, cloudiness, precipitation and the related multi-scale
139 transport processes.

140 The purpose of the paper is to assess the nonlinear relationship between the synergistic emission
141 reduction of atmospheric primary pollutants during the COVID-19 pandemic and the level of
142 atmospheric photo-oxidants and secondary species (e.g., ozone and secondary organic aerosols)
143 produced in different regions of the world during the early months of 2020.

144

145 Ozone is formed during daytime by nonlinear processes at a rate that is determined by the
146 atmospheric concentrations of VOCs and NO_x. Under low NO_x levels in remote or weakly
147 polluted areas, the ozone production is controlled (or limited) by the concentration of NO_x because
148 high NO_x concentrations enhance the production of peroxy radicals and subsequently ozone.
149 Ozone is primarily destroyed by reactions involving hydrogenated species leading eventually to
150 the formation of hydrogen peroxide ($\text{HO}_2 + \text{HO}_2 \rightarrow \text{H}_2\text{O}_2$), which is scavenged by wet and dry
151 deposition. In very high NO_x environments, i.e., in heavily polluted areas including industrial and

152 urban complexes, nitrogen oxides act as a sink for the OH radical, which slows down the oxidation
153 of VOCs and hence the formation of peroxy radicals. As a result, the ozone production is
154 considerably reduced. Rather, ozone is sequestered by NO to form NO₂ and is further titrated by
155 NO₂ and eventually converted to nitric acid (NO₂ + OH → HNO₃), which is removed from the
156 atmosphere by wet and dry scavenging. This situation is referred to as NO_x-saturated or VOC-
157 controlled conditions. The reduction in VOCs and NO_x during the pandemic is therefore expected
158 to have led to a reduction of ozone in NO_x-limited regions, but to have caused an increase in the
159 ozone concentration in the most polluted areas, especially during winter when the levels of NO_x
160 are the highest. An increased oxidation capacity in the eastern part of China has been reported in
161 Huang et al. (2020), whereas enhanced concentrations of ozone in the North China Plain were
162 reported by Liu and Wang (2020), Shi and Brasseur (2020) and Miyazaki et al. (2020). This
163 question will be further examined in subsequent sections.

164
165 The dominant source of secondary organic aerosols is provided by biogenic hydrocarbons
166 including isoprene and terpenes, and by anthropogenic VOCs (linear and aromatic hydrocarbons)
167 resulting from fossil fuel consumption, the industrial and domestic use of solvents and of other
168 products, and from biomass burning. The rate at which the degradation of primary hydrocarbons
169 proceeds, depends on the concentration of oxidants and on the level of nitrogen oxides (NO_x)
170 present in the atmosphere (Hallquist et al., 2009). SOA concentrations are expected to be reduced
171 as a result of the reduction in VOC emissions. However, the reduction in NO_x tends to increase
172 the SOA production (Ng et al., 2007) under high NO_x conditions by increasing the concentration
173 of the OH radical, and therefore to partially offset the SOA decrease caused by the reduced VOC
174 emissions.

175
176 The paper is organized as follows. Section 2 provides a short description of the emission reductions
177 that are considered here as a forcing factor to the anomalies in the concentrations of chemical
178 species during the pandemic period. Section 3.1 presents a description of the global earth system
179 model that is adopted to analyze the atmospheric response to this forcing during the pandemic.
180 Section 3.2 focuses on the evaluation of the atmospheric fields derived by the model and compares
181 selected results with surface measurements of key species at different locations in the world.
182 Section 4 provides a global view of the changes that have occurred in the chemical composition of
183 the atmosphere in response to the reduced emissions of primary pollutants and to meteorological

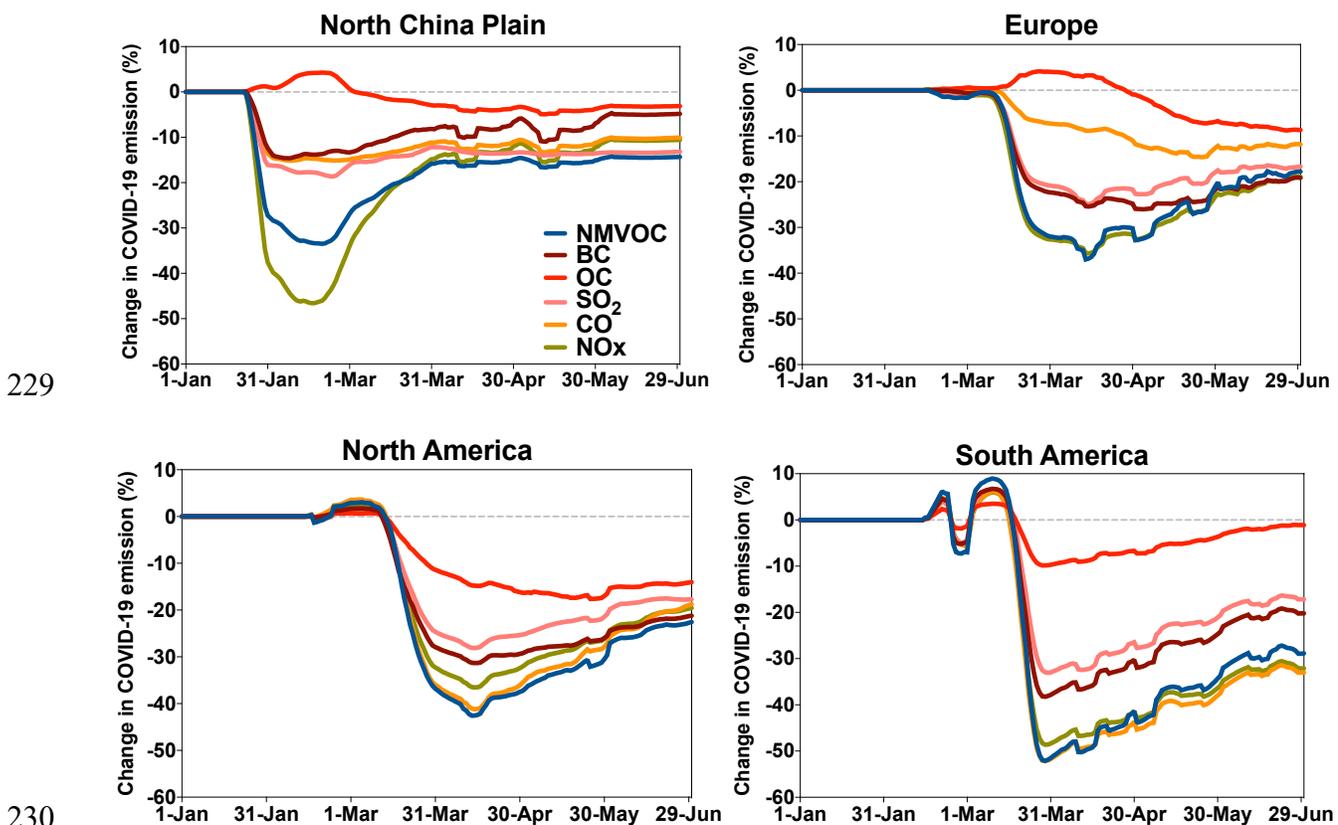
184 anomalies occurring during the pandemic. Section 5 assesses the calculated changes in the
185 concentration of chemical species in selected regions (China, Europe, North and South America)
186 where lockdowns were imposed during the first months of 2020. This section discusses in
187 particular the respective impact of the reduction in NO_x versus VOC emissions as well as the role
188 of meteorological variability on the calculated chemical fields. A summary of the findings and key
189 conclusions are provided in Section 6.

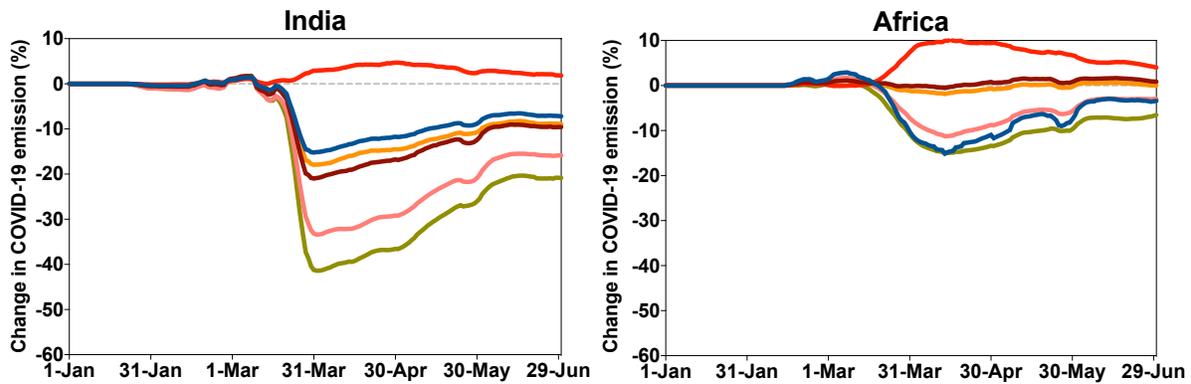
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191 **2. Adjustments in Emissions during the Pandemic**

192 The change in the emissions of primary pollutants associated with the COVID-19 pandemic has
193 considerably varied among different economic sectors and geographic areas. The time at which
194 the lockdowns were enforced and the severity of the measures taken to protect the population was
195 different from country to country and even from region to region. Several studies (Guevara et al.,
196 2020; Doumbia et al., 2020) have attempted to estimate these changes in emissions on the basis of
197 available economic information regarding different sectors: transportation (road, air and sea
198 traffic), industrial production, energy consumption and residential activity. Here we adopt the
199 global estimates provided by Doumbia et al. (2020) gridded at a spatial resolution of 0.1 x 0.1
200 degree (about 10 x 10 km). In this study, adjustment factors were derived for each economic sector
201 and geographic region based on activity data, and the resulting changes in the emissions of a given
202 primary chemical species were calculated at each grid point of the model based on the relative
203 contribution of each sector to the total emission. In some cases, the input data used to derive the
204 adjustment factors was available at the country level, but in some cases, more resolved sub-
205 regional-scale and even local-scale information was used. Some input data, for example the
206 reduction of road traffic intensity, was accessible on a day-to-day basis for major cities in many
207 (but not all) countries, and allowed Doumbia et al. (2020) to provide 10 km resolution emission
208 estimates on a daily basis from January to August 2020. The reduction in the emissions for the
209 shipping and aviation sectors adopted in the present study is also obtained from Doumbia et al.
210 (2020). Figure 1 shows the geographically averaged percentage surface emission adjustment
211 applied during the pandemic in different regions of the world and for different chemical species.
212 In the Northern China Plain, the change in the emissions attributed to the COVID-19 pandemic
213 occurred as early as 23 January 2020 and, as the lockdown was immediately implemented,
214 happened abruptly. The largest reduction in emissions occurred in mid-February 2020. At that

215 time, the reduction in NO_x reached 50% and is explained in large part by the nearly complete
 216 shutdown of road traffic. The second largest reduction factor, as estimated by Doumbia et al.
 217 (2020), is the decrease of 30% for the emissions of non-methane volatile organic compounds
 218 (NMVOC). For carbon monoxide (CO), black carbon (BC) and sulfur dioxide (SO₂), the maximum
 219 adjustment factor is close to 0.9 (10% reduction). In the case of organic carbon (OC), an increase
 220 of a few percent results from the enhanced domestic activity (stay-at-home policy), particularly
 221 related to more extensive cooking and heating during the pandemic period. In the other regions of
 222 the world, the COVID-related perturbations in the emissions are most pronounced in mid-March
 223 to mid-April at a time where China's emissions were already in a recovery phase. In the European
 224 Union and North America, the estimated change in the emissions was largest in April. In South
 225 America and India, a sharp decrease appeared in the second half of March followed by a slow
 226 recovery from April to June. In Africa, only a small reduction occurred for NO_x and VOCs with a
 227 maximum reduction in mid-April and a slow recovery afterwards. In all countries, except the
 228 Americas, a slight increase in organic carbon is derived during the pandemic.



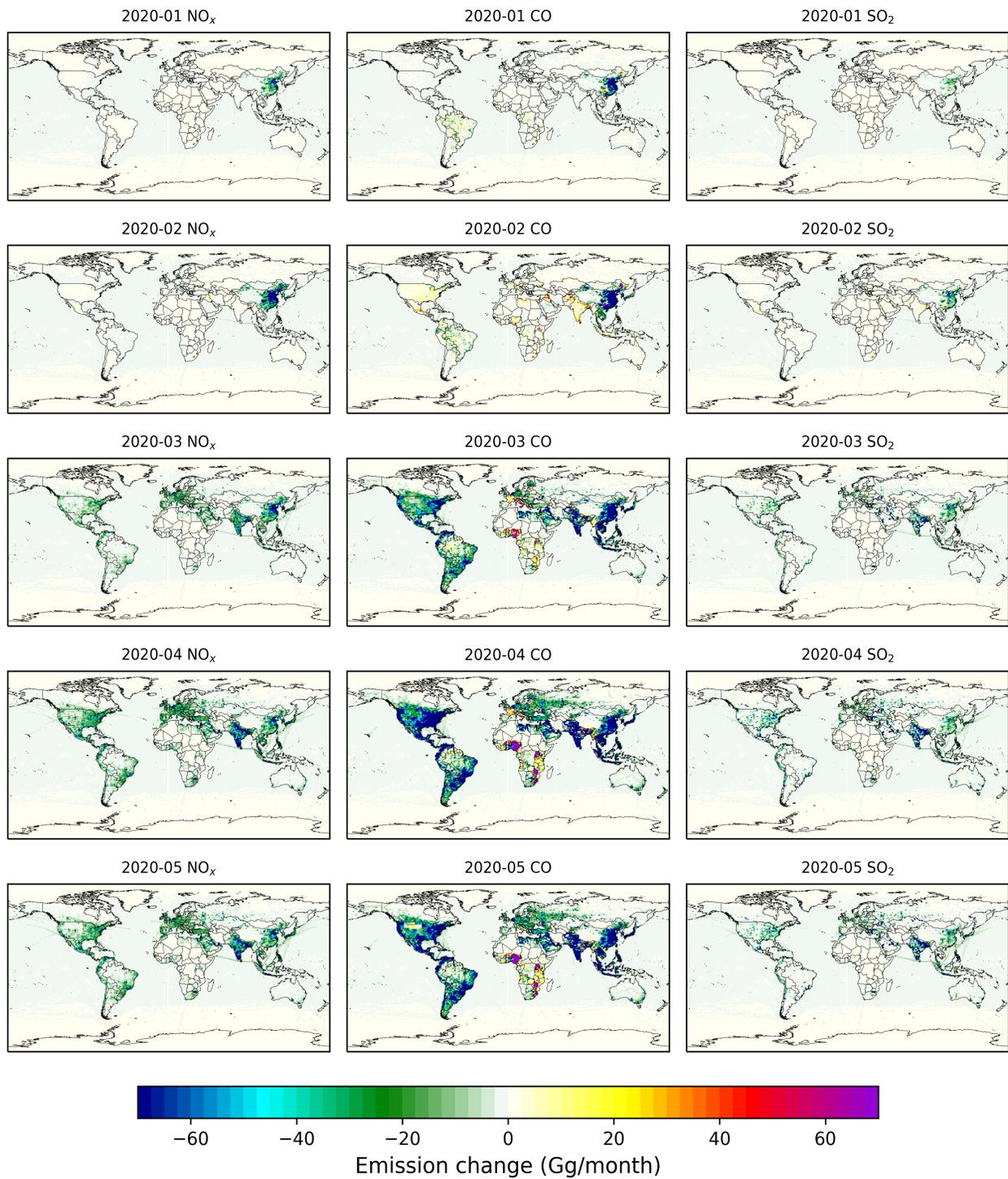


231

232 **Figure 1.** Evolution from 1 January to 30 June 2020 of the daily geographically averaged adjustment factors
 233 (percent) for the emissions for nitrogen oxides (NO_x), carbon monoxide (CO), sulfur dioxide (SO₂), organic
 234 carbon (OC), black carbon (BC) and non-methane volatile organic carbon (NMVOC) in the North China
 235 Plain, Europe, North America, South America, India and Africa. A weak filter has been applied to smooth
 236 out the high frequency variability in the curves. Based on Doumbia et al. (2020).

237 Figure 2 shows a global view of the monthly mean percentage reduction in the emissions of NO_x
 238 CO and SO₂ for January to May 2020. In February, the reduction in the emissions is contained in
 239 China. In March and April with lockdown measures imposed in other parts of the world, substantial
 240 reductions are seen in the NO_x and VOC emissions of Europe, the Middle East, India and North
 241 America. In the southern hemisphere, emissions are substantially reduced on both coasts of South
 242 America and South Africa. The decrease over the global ocean accounts for the reduced shipping
 243 activity in response to the slowdown of the economy.

244 We should acknowledge here that large uncertainties might reside the aforementioned adjustment
 245 factors as discussed in Doumbia et al. (2020). As an example, Guevara et al. (2020) estimated for
 246 the period 23 March to 26 April 2020 in Europe an average emission reduction of 33% for NO_x,
 247 8% for VOCs and 15% for CO. The average reductions by Doumbia et al. (2020) for the similar
 248 period are 33 %, for NO_x, 30% for VOCs and 15% for CO. The difference in the VOC emission
 249 adjustment factors might be due to a different treatment of the reduction in solvent emissions
 250 during the pandemic, as solvents contribute a large share of the anthropogenic VOC emissions in
 251 Europe. In the model simulations presented in the following sections, we address this particular
 252 uncertainty by considering a case in which the high VOC emission reduction of Doumbia et al.
 253 (2020) is adopted (upper limit) and a case in which no reduction in VOC and CO emissions is
 254 applied (lower limit).



255

256

257 **Figure 2.** Change in the monthly mean emissions of NO_x (left) CO (middle) and SO₂ (right) from January
258 2020 (upper panels) to May 2020 (lower panels).

259

260 **3. Global model simulations**

261 **3.1 Community Earth System Model version 2.2 (CESM2.2)**

262 The chemical fields presented in our study are provided by the Community Earth System Model
263 (CESM) version 2.2 that accounts for interactive physical, chemical and dynamical processes
264 (Danabasoglu et al., 2020). The atmospheric component of CESM, the Community Atmosphere
265 Model (CAM-Chem), provides a comprehensive description of atmospheric chemistry and aerosol
266 processes (Gettelman et al., 2019; Tilmes et al., 2019; Emmons et al., 2020; Gaubert et al., 2020)
267 with a spatial resolution of 1.25° in longitude by 0.95° in latitude (about $100 \times 100 \text{ km}^2$ at mid-
268 latitude) and with 32 vertical layers from the surface to the pressure height of 3.6 hPa (about 40
269 km altitude). The MOZART Troposphere Stratosphere (TS1) chemistry mechanism includes 221
270 gas phase and aerosol species and 528 chemical and photochemical reactions, and provides
271 therefore a rather comprehensive explicit and interactive representation of tropospheric and
272 stratospheric chemical processes (Emmons et al., 2020). Aerosols are represented by the four-
273 mode Modal Aerosol Model (MAM4, Liu et al., 2016; Mills et al., 2016). The updated Secondary
274 Organic Aerosol parameterization includes the Volatility Basis Set (VBS) approach discussed by
275 Tilmes et al. (2019).

276 In order to accurately represent the meteorological conditions, the wind velocity components and
277 the temperature are nudged towards the Global Modeling and Assimilation Office (GMAO) GEOS
278 Forward Processing (GEOS-FP) meteorological analysis. The GEOS-FP assimilation is performed
279 on the cubed-sphere grid at C720 resolution or 12 km. The data from the three-hourly
280 meteorological horizontal wind and temperature analysis are regridded to the CAM-chem
281 horizontal and vertical resolution. They are nudged with weight calculated at every CAM-Chem
282 physical step (30 min) and with a Newtonian relaxation of about 5 hours. The ocean and sea-ice
283 interfaces are prescribed from a daily analysis of sea surface temperatures (SST, Reynolds et al.,
284 2007).

285
286 The model includes an active coupling between the atmosphere and the Community Land Model
287 version 5 (CLM5; Lawrence et al., 2019), where the deposition of gases and aerosols is calculated.
288 Biogenic emissions are calculated online in CLM5 with the Model of Emissions of Gases and
289 Aerosols from Nature (MEGAN v2.1; Guenther et al., 2012). Daily biomass burning emissions
290 are based on the biomass burning CO_2 emission inventory available from the Quick-Fire Emissions

291 Dataset (QFED; Darmenov and Da Silva, 2014). The chemical speciation is derived using the Fire
 292 INventory from NCAR (FINN) emissions ratios.

293
 294 The anthropogenic emissions are specified according to the CAMS-GLOB-ANT_v4.2-R1.1 global
 295 inventory described by Granier et al. (2019) and Elguindi et al. (2020). This inventory provides
 296 monthly-averaged emissions of the main chemical compounds and 25 speciated volatile organic
 297 compounds for the 2000-2020 period at a spatial resolution of 0.1x0.1 degree. It is based on the
 298 EDGARv4.3.2 inventory developed by the European Joint Research Center (Crippa et al., 2018)
 299 and the CEDS emissions (Hoesly et al., 2018), which provide historical emissions for the 6th IPCC
 300 Assessment Report (AR6). EDGARv4.3.2 emissions are available until 2012: the emissions are
 301 linearly extrapolated to 2020 according to the trends derived from the CEDS emissions for the
 302 years 2011-2014. For China, the emissions from MEIC1.3 (Zheng et al. 2018) are used to account
 303 for the recent decrease in the emissions of most pollutants in this region. Daily averaged emissions
 304 obtained from an interpolation of the monthly averaged emissions are used for the baseline
 305 simulations and are adjusted for COVID-related runs by applying the daily adjustment factors
 306 discussed in Section 2 (Dolumbia et al., 2020).

307
 308 To evaluate the relative influence of different forcing processes responsible for the variations in
 309 the surface concentrations of reactive species, we perform different model simulations that are
 310 summarized in Table 1. In cases 1 and 2, referred to as “control” and “climato” cases respectively,
 311 the COVID-related reductions are ignored, but, in cases 3 to 5, the long-term trend in surface
 312 emissions associated with the evolving economy and air pollution mitigation measures are taken
 313 into account. The meteorology varies from year to year with the model being nudged to assimilated
 314 observations for the period 2016-2020. In case 2, the same 2020 emissions are used for each year,
 315 with, however, the meteorology evolving as in case 1. Case 3 is a repeat of case 1 for the year
 316 2020, but with an adjustment in the emissions for the COVID-19 pandemic period. Cases 4 and 5
 317 are similar, but with adjustments for only NO_x and for only VOCs and CO, respectively. Global
 318 distributions of simulated chemical species obtained with the controlled case are displayed in
 319 Figure S1 of the Supplementary Information.

320

321 **Table 1.** Description of the different model simulations used in the present study

Case	Name	Reduction in	Meteorology	Notes
------	------	--------------	-------------	-------

Emissions				
1	Control (2016 to 2020)	None	GEOS-FP evolving from 2016 to 2020	Long-term emission trends taken into account
2	Climato (2016-2020)	None	GEOS-FP evolving from 2016 to 2020	Simulation from 2016 to 2020 with annually repeated 2020 emissions
3	COVID-All (2020)	All emitted species	GEOS-FP for 2020	Combined emission reduction
4	COVID-NO _x (2020)	NO _x only	GEOS-FP for 2020	Impact of NO _x reduction
5	COVID-VOC (2020)	VOCs and CO only	GEOS-FP for 2020	Impact of VOC and CO reduction

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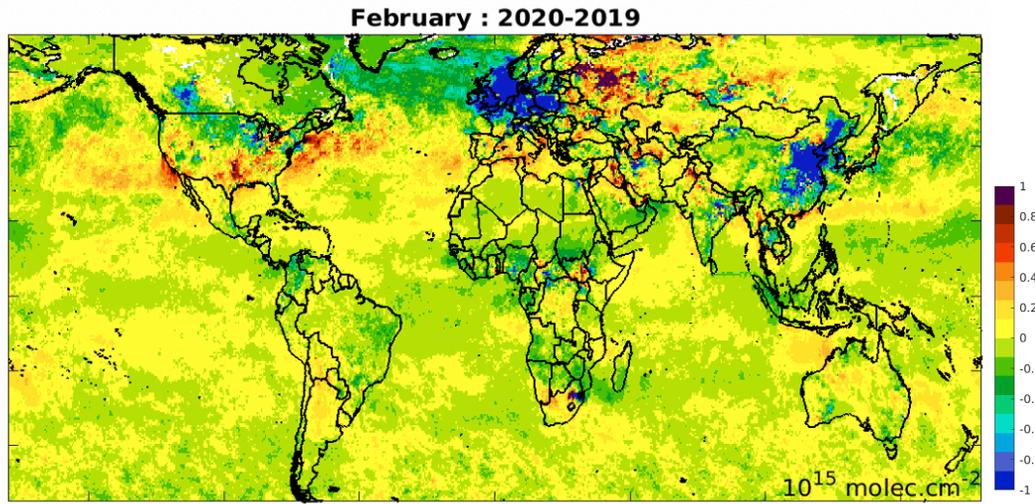
3.2 Model evaluation

324

325 We first provide some insight about the performance of the CESM model regarding its ability to
 326 reproduce the observed concentration of reactive species at the surface. Since a detailed evaluation
 327 of the model is beyond the scope of this paper and has been performed in earlier studies (Emmons
 328 et al., 2020), we only compare the calculated time series of ozone and other key atmospheric
 329 species (NO₂, CO, SO₂, PM_{2.5}) with measurements made at selected locations in different parts of
 330 the world. We first show in Figure 3 the global distribution of the monthly mean difference
 331 between 2019 and 2020 in the monthly mean NO₂ column derived from TROPOMI measurements
 332 in February and compare it with the change in the column as calculated by the model. The
 333 comparison between the two figures should be viewed as qualitative since the TROPOMI data are
 334 representative of early afternoon measurements while the model results refer to 24-hour average
 335 values. Further, both 2019 and 2020 simulations with the meteorology specific for each year have
 336 been performed with the same background emissions. In the 2020 case, however, the COVID-
 337 adjusted emissions are accounted for. What needs to be stressed here is that, in addition to the NO₂
 338 decrease seen in China during the month of February in response to the economic slowdown, a
 339 significant reduction in the NO₂ column is found in northern Europe, in the North Atlantic and in
 340 the northern part of the US. Since no lockdown was imposed at that time in Europe and in North
 341 America, these changes should be viewed as a fingerprint of meteorological variability. Barré et
 342 al. (2020) and Goldberg et al. (2020) stress that meteorological variability complicates the analysis
 343 of observed data, and that such natural variations have a large effect when comparing, for example,
 344 observations between two consecutive years. The reduction observed in NO_x observed in northern
 345 Europe during February has generated an increase in ozone (see Figure 8). Interestingly, both the

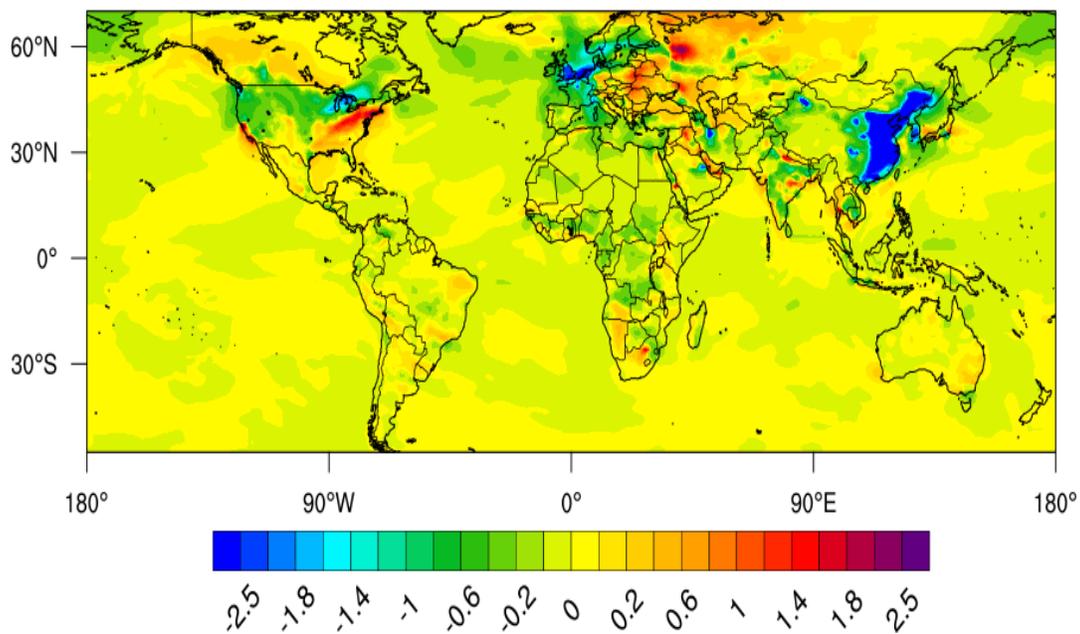
346 model and the space observations also show a small NO₂ increase in the southern and eastern part
 347 of the US and in the western part of Russia.

348
 349
 350



COVID-ALL - Control_2019 NO2 Column February

1.e15 molec.cm-2



351
 352

353 **Figure 3.** Difference between February 2020 and February 2019 in the global distribution of the monthly
 354 mean NO₂ column abundance (10^{15} cm^{-2}) Upper Panel: Observed values derived from TROPOMI satellite
 355 instrument. Lower Panel: Model values based on 2019 and 2020 simulations accounting for the reduction
 356 in surface emission and meteorological variability.

357
 358

359 We now provide comparisons of calculated and observed time series for surface concentrations of

360 different chemical species in selected regions where measurements are available. We show in
361 Figure 4a-g the evolution of the species concentration between 1 January and 31 May 2020 for
362 two model cases: (1) a baseline case with no correction for COVID-19 effects (case 1 or control
363 case; green curves in the figures) and (2) a perturbed case with emissions adjusted as described
364 above to account for the COVID-19 effect (case 2 or COVID-All case; red curves in the figures).
365 The differences between these two cases are further addressed in Section 3.3. We also show as
366 supplemental information (Figure S2) the evolution of the ozone mixing ratio averaged over areas
367 of about 100 x 100 km² around 6 European cities. The observations represent an average of the
368 measurements made at monitoring stations in these areas. The outputs of the simulations are first
369 interpolated to the location of these stations and, as for the observational data, are averaged over
370 the chosen area.

371
372 In most regions, the model reproduces relatively well the chemical composition of the lower layers
373 of the atmosphere. The long-term average evolution of the concentrations calculated by the model
374 are influenced mainly by emissions from primary species and by chemical processes that occur in
375 the atmosphere, while day-to-day variability is directly dependent on atmospheric dynamics and
376 other meteorological processes. Data from monitoring stations are influenced by local emissions
377 and by small-scale dynamic processes that cannot be captured by a model with a relatively coarse
378 spatial resolution. Further, monitoring stations are often located in areas prone to high air pollution.
379 Polluted areas may therefore be oversampled relative to the spatial averages provided by the
380 model. In Europe and in the Sao Paulo region, we address this issue by removing from our analysis
381 the data from stations located in areas with a population density larger than 1500 inhabitants per
382 km², which is the threshold adopted by the European Union to define “high-density areas”.

383
384 We evaluate the model results by comparing the surface concentrations averaged over relatively
385 large areas with the average of measurements made in these areas (Table 1). Model outputs are bi-
386 linearly interpolated to each selected station. We then calculate the mean concentration and the
387 standard-deviation of all selected stations to construct the daily time series. In Europe, we use four
388 criteria to calculate the daily averages and select the stations of the European network (AQ e-
389 Reporting database) according to the following criteria: (i) 70% of hourly data must be available
390 to calculate the daily average, (ii) a maximum of 10% of missing values on daily averages, (iv) the
391 urban stations are not taken into account (threshold of 1500 inhabitants per km²), (iv) the altitude

392 of the station is lower than 300 m. These selection criteria are especially important over
 393 Southwestern Europe because of the large variability in the environment (urban, rural, mountain)
 394 compared to Northern Europe. For the São Paulo region (CETESB network), we use the three
 395 criteria (i, ii and iii). For the three domains of North America (Air Quality System Data Mart
 396 database of US Environmental Protection Agency), we use two criteria (i and ii). For the North
 397 China Plain (China Environmental Observation Network), only one criterion (i) is used because
 398 this area is a continuously urbanized and represents a flat area.

399

400 **Table 2.** Domain names, acronyms, coordinates of the south-western and north-eastern corners of
 401 the domain's frames, and the number of stations per species retained in the present study.

Domain Name	Domain Acronym	Coordinates	Species (Number of Stations)
North China Plain	NCP	(112°E; 34°N) to (119°E; 42°N)	NO ₂ (220), CO (220), O ₃ (220), SO ₂ (220), PM _{2.5} (220)
Northern Europe	N-Eu	(1°W; 47°N) to (11°E; 54°N)	NO ₂ (151), CO (16), O ₃ (156), SO ₂ (35), Ox (115), PM _{2.5} (53)
Southwestern Europe	SW-Eu	(9°W; 39°N) to (3°E; 45°N)	NO ₂ (32), CO (9), O ₃ (42), SO ₂ (19), Ox (29), PM _{2.5} (10)
Northeastern USA and Southern Canada	NE-US	(126°W; 36.5°N) to (94°W; 52°N)	NO ₂ (122), CO (81), O ₃ (233), PM _{2.5} (257)
Southern USA	S-US	(94°W; 24°N) to (66°W; 36.5°N)	NO ₂ (43), CO (27), O ₃ (113), PM _{2.5} (86)
Western USA	W-US	(94°W; 24°N) to (66°W; 52°N)	NO ₂ (243), CO (120), O ₃ (417), PM _{2.5} (291)
Sao-Paulo	Sao-P	(47.5°W; 24°S) to (45.5°W; 22°S)	NO ₂ (6), CO (1), O ₃ (47), SO ₂ (3), Ox (4), PM _{2.5} (3)

402

403 The model results obtained for the North China Plain (32° – 40° N, 112.5° – 120° E) show that the
 404 model satisfactorily reproduces the concentrations of NO₂, ozone and Ox (= O₃ + NO₂) as well as
 405 their temporal variability (Figure 4a). The model also underestimates the CO concentration by a
 406 factor of 1.5-2 during and after April 2020. In the case of SO₂, the agreement of the model with
 407 the observation is satisfactory, except during the months of January and February when the
 408 modeled concentrations and their temporal variability are considerably higher than in the
 409 observations. During this period, the model also underestimates the concentrations and temporal
 410 variability of atmospheric particles (PM_{2.5}) and CO. The repeated high peaks in PM_{2.5}, which are
 411 observed, but whose amplitude is underestimated by the model, show acute haze episodes as those
 412 reported for example in Beijing (Li et al., 2020). The high correlation of the model overestimation
 413 for SO₂ with the observed PM_{2.5} concentration suggests that the aerosols might be a significant

414 SO₂ sink in winter in this region, as recently proposed by Cheng et al. (2016) to explain the inability
415 of the models to reproduce the observed sulfate production during haze events.

416

417 In northern Europe (Figure 4b), the calculated concentrations of NO₂, ozone and Ox are generally
418 in good agreement with observations. However, the calculated concentrations of CO are about
419 40% lower than the observations. This discrepancy, also noted in Northern China in April and
420 May, repeats itself in most regions of the northern hemisphere and remains an open scientific
421 question since it appears in most models unless their emissions have been artificially increased
422 (Stein et al., 2014) or observationally constrained using data assimilation (Gaubert et al., 2016,
423 2020). The calculated SO₂ variability is larger than observed and the concentration values
424 generally higher than observed.

425

426 In southwestern Europe (Figure 4c), the concentrations of CO, NO₂, SO₂ and PM_{2.5} are
427 underestimated by the model, while the concentrations of ozone and Ox are overestimated. The
428 concentration ratio O₃/NO₂ is too high, perhaps due to an overestimation of the NO₂ photolysis
429 rate during this period that was unusually cloudy.

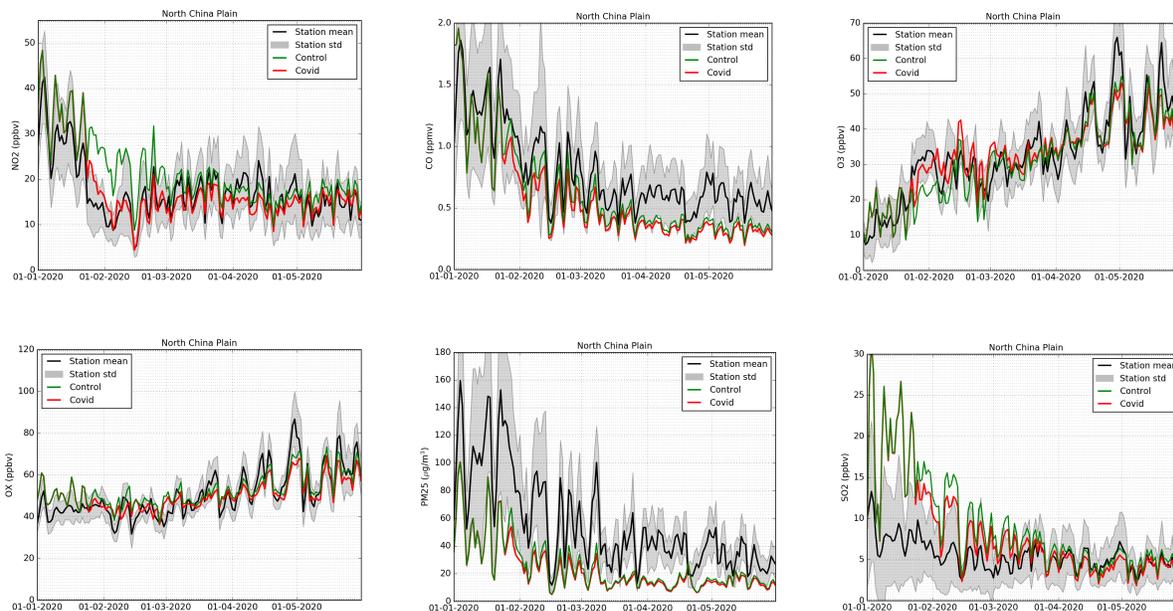
430

431 In the USA (Figures 4d-f), the concentrations of NO₂ are in fair agreement with measurements in
432 the northeastern (NE) US and southern Canada; there are, however, lower than observations in the
433 southern and western regions of the US. Ozone is in fair agreement with measurements in the NE
434 US, but overestimated in the southern and western parts of the country. CO is underestimated in
435 all regions. PM_{2.5} are underestimated in the NE and western US and in good agreement in the
436 southern region.

437

438 In Sao Paulo, Brazil (Figure 4g), the temporal evolutions of ozone, Ox and PM_{2.5} are correctly
439 simulated despite a large underestimation of NO₂ (factor 3-4). SO₂ is underestimated by a similar
440 factor. It should be noted, however, that, in this very heterogeneous megapolis, the NO₂ and SO₂
441 measurements vary greatly from one location to another and that these differences cannot be taken
442 into account by a model whose mesh size is of the same order of dimension of the city. CO
443 concentrations are in fair agreement with observed values.

444

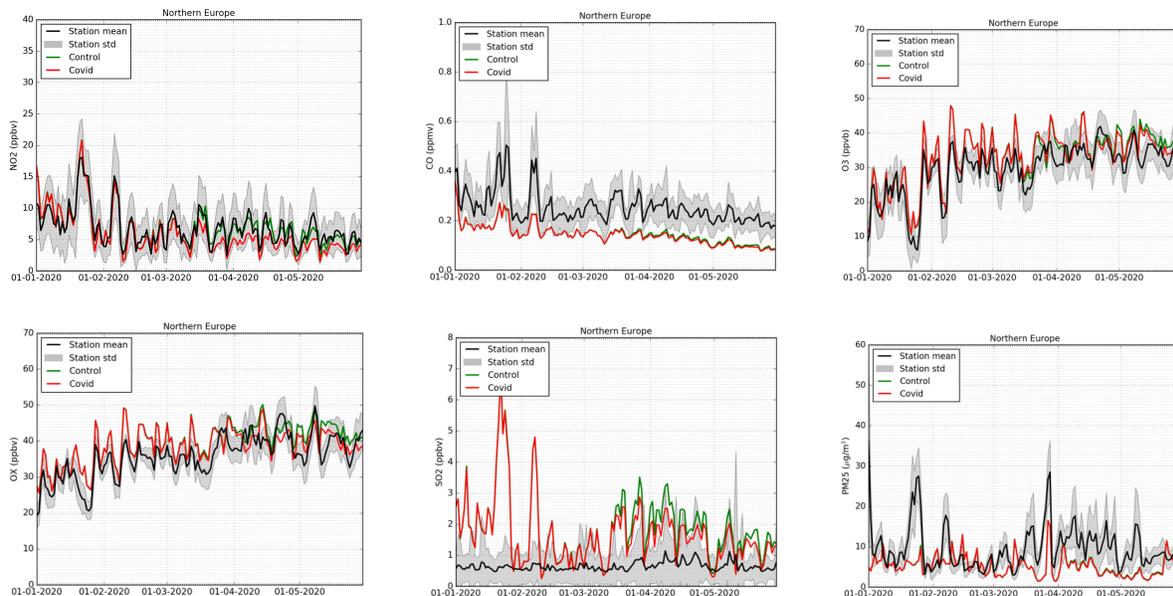


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446

447 **Figure 4a.** Evolution for the period 1 January – 31 May 2020 of the surface concentration of NO₂, CO, O₃,
 448 Ox, SO₂ and PM_{2.5} in the North China Plain. Black curve: measurements from monitoring stations. Green curve:
 449 model control case. Red curve: model case with emissions modified to account for the effect of the
 450 COVID-19 pandemic.
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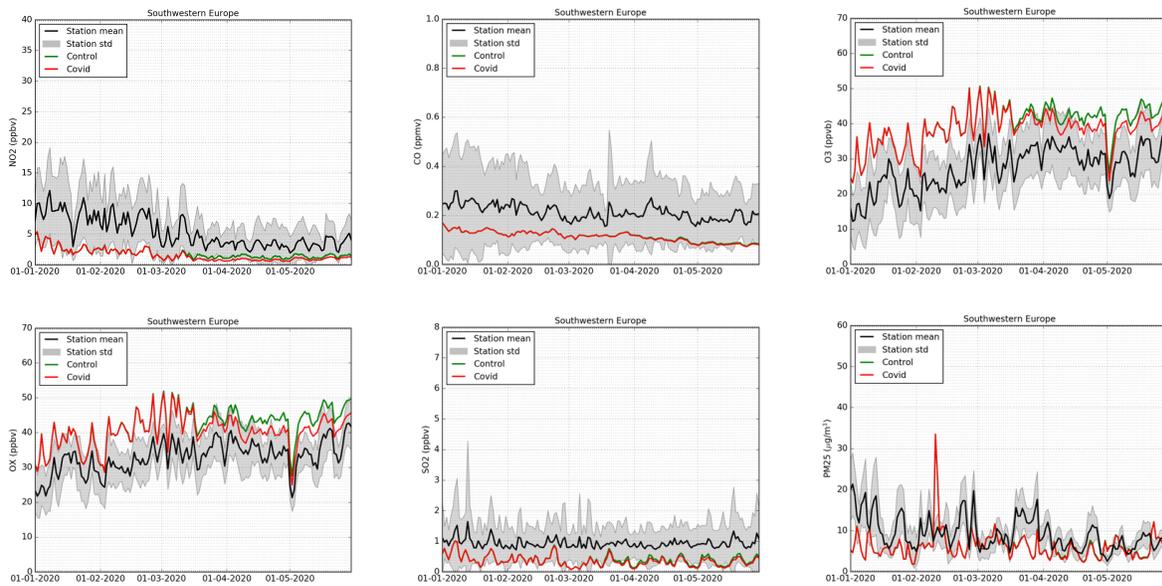


453

454

455 **Figure 4b.** Evolution for the period 1 January – 31 May 2020 of the surface concentration of NO₂, CO, O₃,
 456 Ox, SO₂ and PM_{2.5} in northern Europe. Black curve: measurements from monitoring stations. Green curve:
 457 model control case. Red curve: model case with emissions modified to account for the effect of the COVID-
 458 19 pandemic.

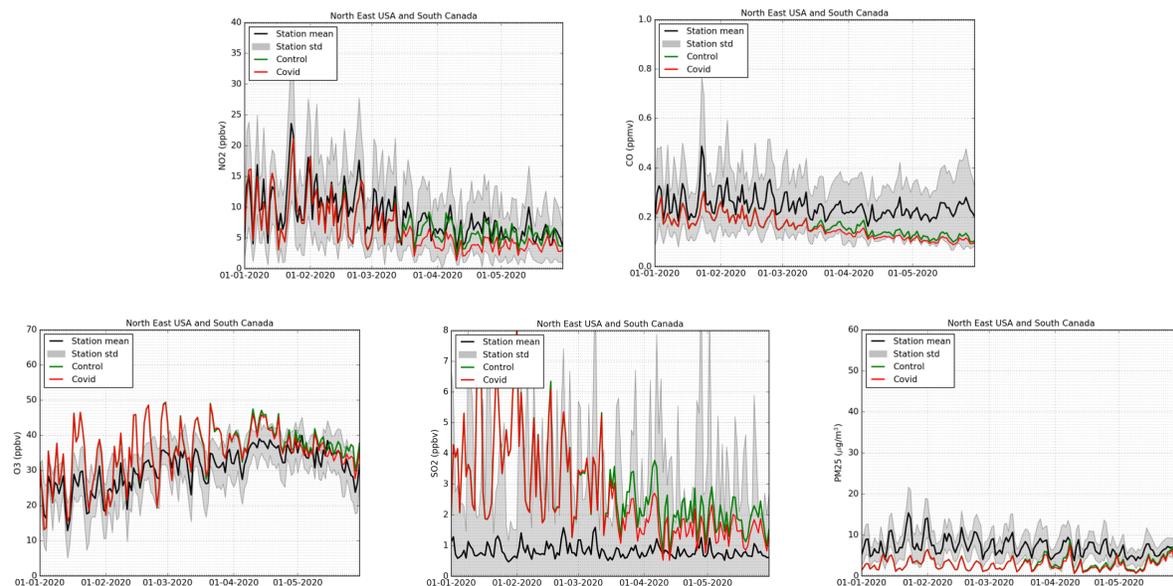
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Figure 4c. Evolution for the period 1 January – 31 May 2020 of the surface concentration of NO₂, CO, O₃, Ox, SO₂ and PM_{2.5} in southwestern Europe. Black curve: measurements from monitoring stations. Green curve: model control case. Red curve: model case with emissions modified to account for the effect of the COVID-19 pandemic.

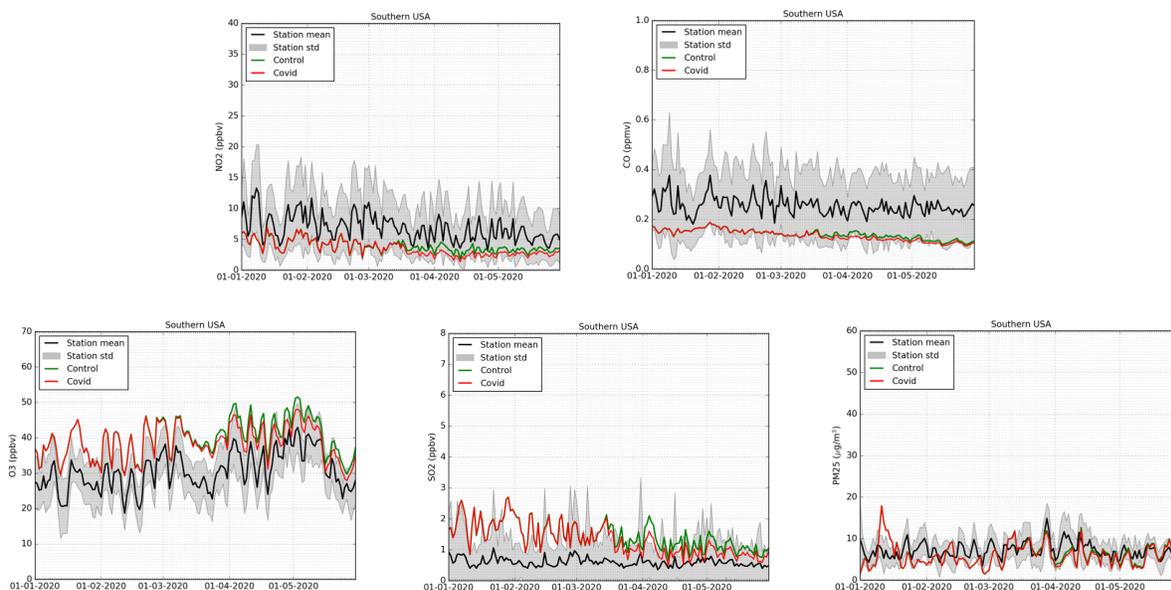
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Figure 4d. Evolution for the period 1 January – 31 May 2020 of the surface concentration of NO₂, CO, O₃, SO₂ and PM_{2.5} in Northeastern USA and South Canada. Black curve: measurements from monitoring stations. Green curve: model control case. Red curve: model case with emissions modified to account for the effect of the COVID-19 pandemic.

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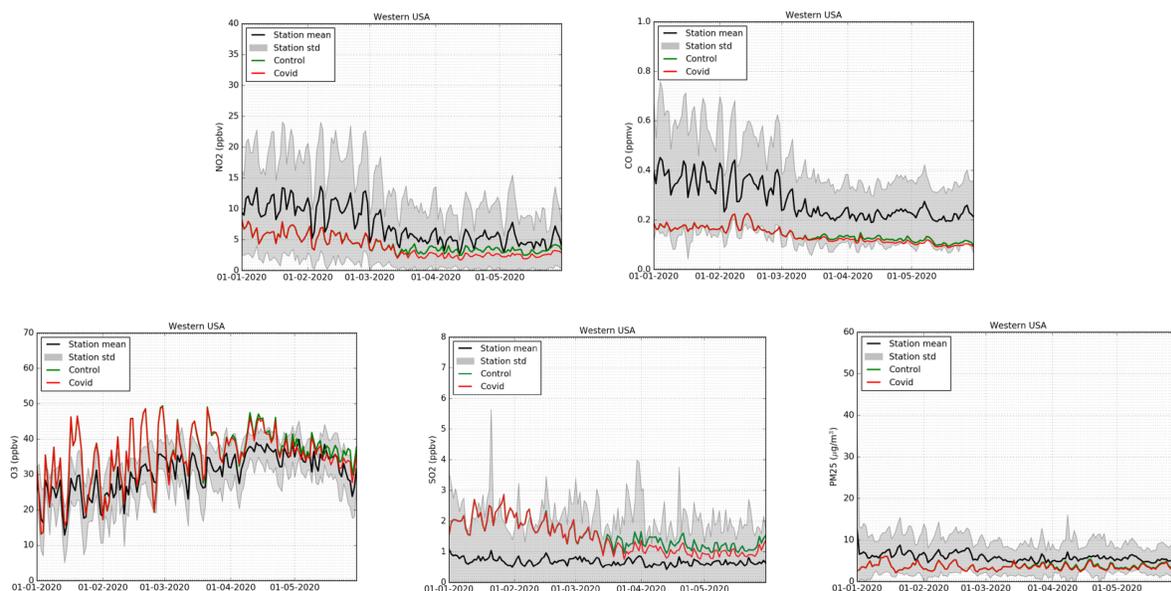


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Figure 4e. Evolution for the period 1 January – 31 May 2020 of the surface concentration of NO₂, CO, O₃, SO₂ and PM_{2.5} in Southern USA. Black curve: measurements from monitoring stations. Green curve: model control case. Red curve: model case with emissions modified to account for the effect of the COVID-19 pandemic.

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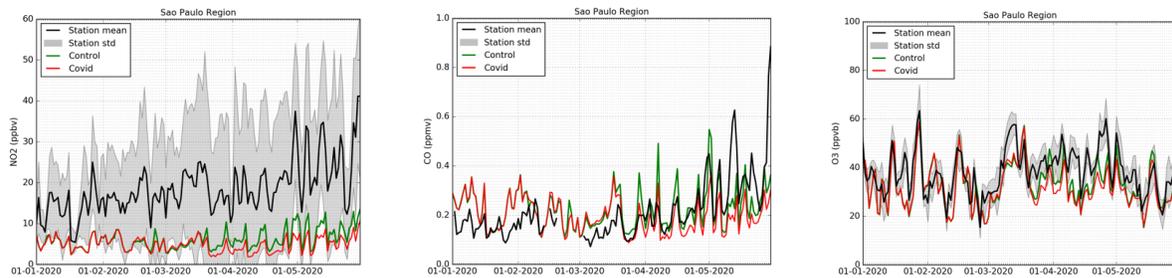


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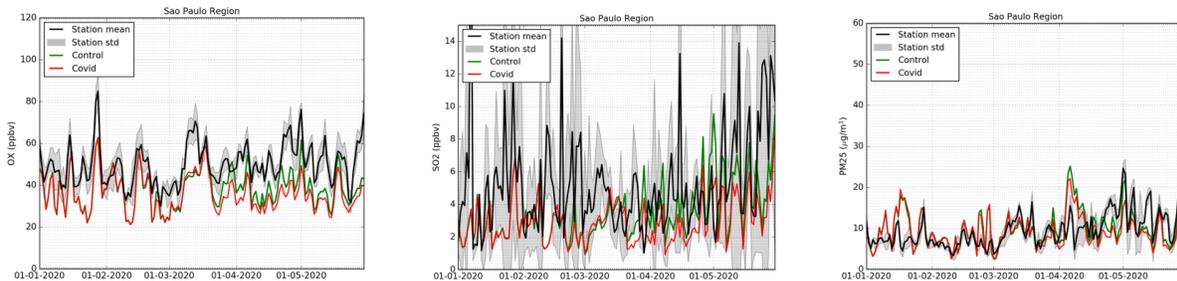
Figure 4f. Evolution for the period 1 January – 31 May 2020 of the surface concentration of NO₂, CO, O₃, SO₂ and PM_{2.5} in Western USA. Black curve: measurements from monitoring stations. Green curve: model control case. Red curve: model case with emissions modified to account for the effect of the COVID-19 pandemic.

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489



490



491 **Figure 4g.** Evolution for the period 1 January – 31 May 2020 of the surface concentration of NO₂, CO, O₃,
 492 Ox, SO₂, and PM_{2.5} in the region of Sao Paulo. Black curve: measurements from monitoring stations. Green
 493 curve: model control case. Red curve: model case with emissions modified to account for the effect of the
 494 COVID-19 pandemic.

495

496

497 Finally, an examination of the ozone concentration in several densely populated areas of Europe
 498 (Figure S2), shows a good agreement between the model and the ground measurements,
 499 particularly near Berlin, Hamburg and Paris. In the area around London, Milan and Madrid,
 500 however, the model slightly overestimates the concentration of this gas in January, but the
 501 difference is reduced in the following months.

502 4. Changes in surface air quality during the COVID-19 pandemic

503 We now examine the global response of the chemical composition to the changes in the surface
 504 emissions during the pandemic. For this purpose, we compare the model results obtained with and
 505 without the modified emissions (COVID-all minus Baseline) as described above. In order to isolate
 506 the impact of the changes in emissions, we constrain the model in both cases by the same
 507 meteorological input corresponding to the year 2020. We also show how the particular dynamical
 508 situation in 2020 has produced anomalies relative to a multi-year averaged meteorology. We
 509 present panels that provide the percentage change in the monthly mean surface concentration of
 510 key chemical species for two different months: February and April, which correspond to the peak

511 time of the lockdown episode in China and in the rest of the world, respectively. Model results for
512 other months are provided as Supplementary Information (Figure S3). Note that the patterns
513 presenting relative changes may be very different from patterns of absolute changes. In this section,
514 we focus on global aspects. A more detailed analysis of the chemical processes that are responsible
515 for the calculated changes in selected regions of the world is provided in Section 4.

516 **4.1 Response to changes in surface emissions of primary pollutants**

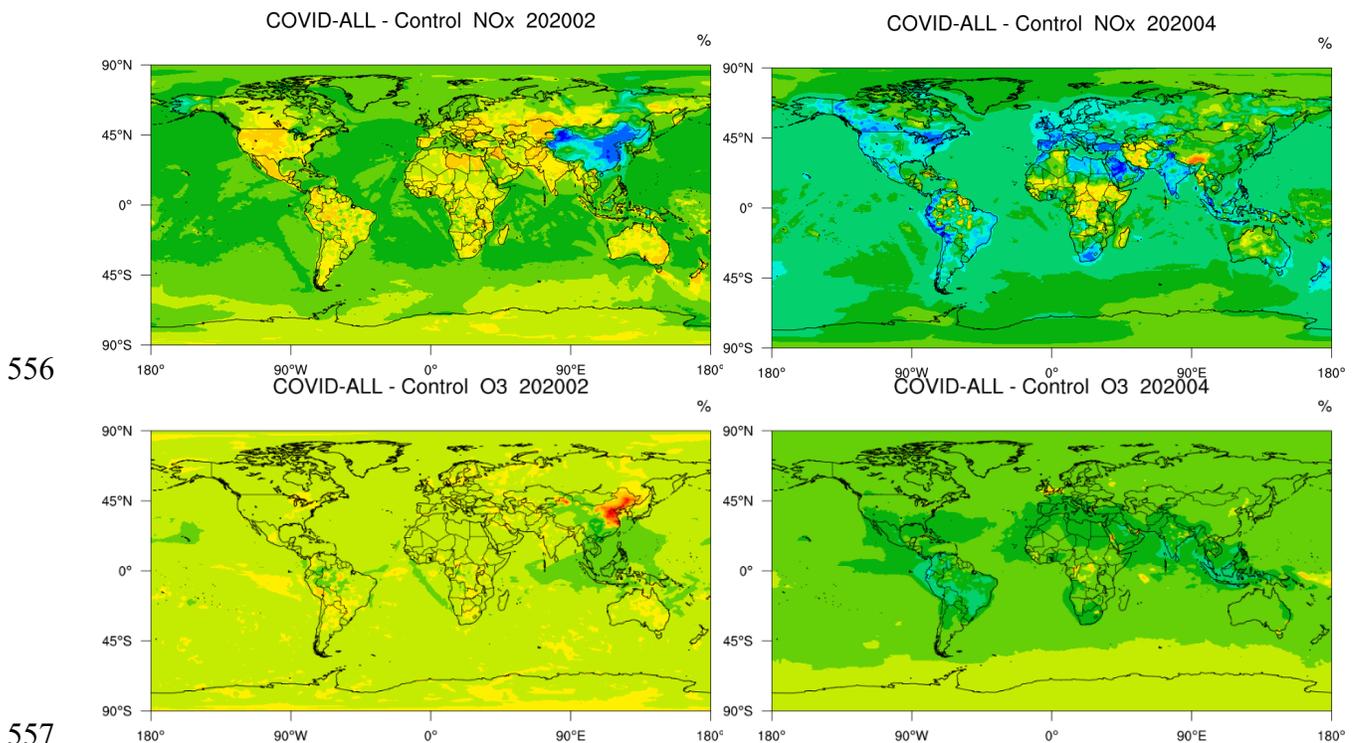
517 When examining the changes in the surface abundance of nitrogen oxides resulting from the
518 synergic emissions of NO_x and VOCs (Figure 5), we note a reduction in the concentration during
519 February that amounts to 30-50% in China, particularly in the Northern China Plain (i.e., north of
520 the Yangtze River) and in the western province of Xinjiang. No significant reduction is yet detected
521 in other parts of the world. Only a small decrease of a few percent is found over the oceans,
522 particularly along the ship tracks and accounts for the assumed slowdown in international shipping
523 activities. In April, the calculated NO_x reduction in China is a factor of two smaller than two
524 months earlier, but the impact of the pandemic has now reached most regions of the world.
525 Reductions of typically 40% are derived by the model in India, Western Europe, Saudi Arabia,
526 Canada, South Africa and in the southern hemisphere, Bolivia, Peru and Ecuador. In Eastern
527 Europe, New Zealand, the east coast of Australia and most regions of the United States and Brazil,
528 the surface concentration of NO_x is reduced by 20-30%. Very small changes are calculated for
529 Central Africa, the center and western coast of Australia, the Asian regions of Russia and Iran. The
530 absence of reduction calculated for Iran is consistent with the observations derived by the
531 TROPOMI instrument (Bauwens et al., 2020).

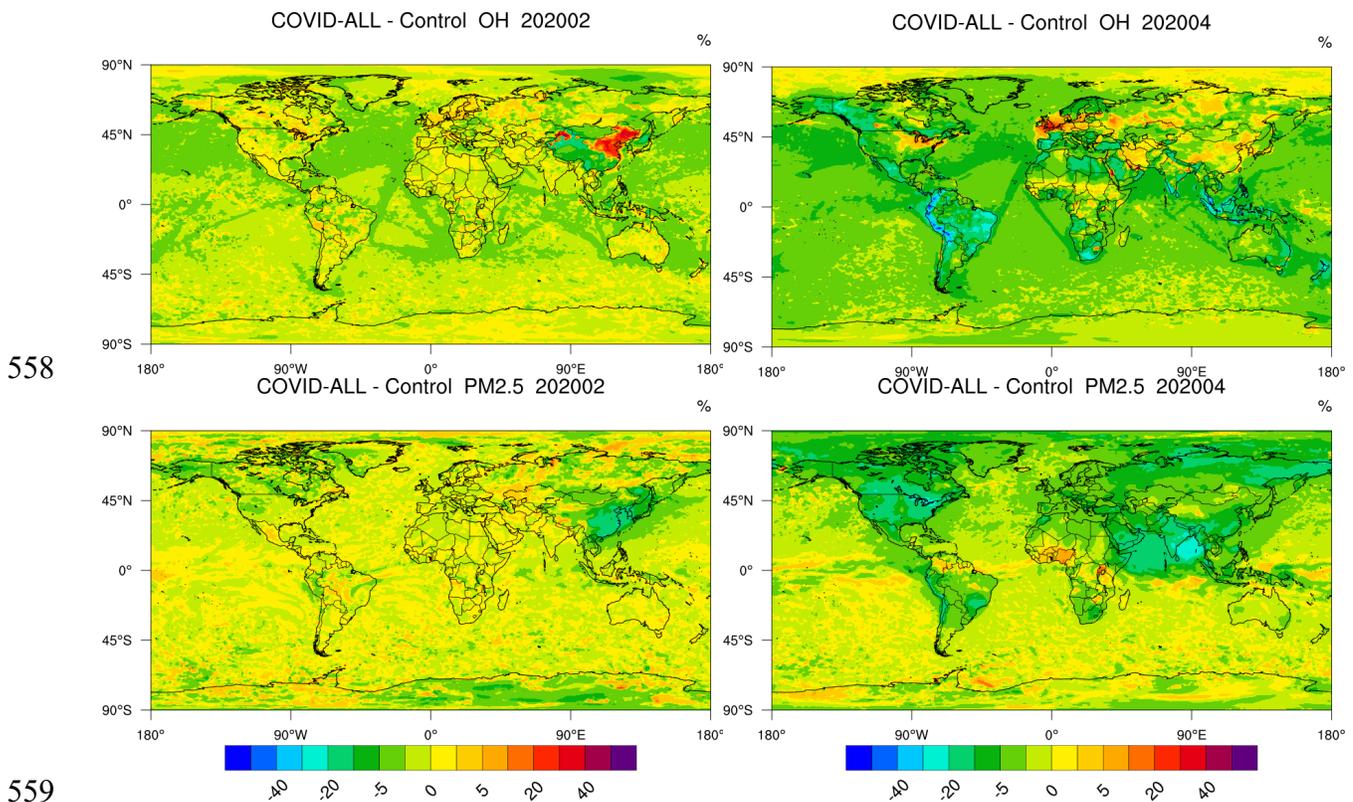
532 The response of ozone during the lockdown of February is characterized by a concentration
533 increase of typically 20-40% in the northeastern part of China. A small spot with a similar increase
534 is found in the province of Xinjiang. In April, the ozone increase in China is vanishing, but changes
535 in the concentration of this gas have now spread in other regions of the world. The largest relative
536 surface ozone reductions are found in the tropics (5 – 20%), specifically in northern Peru and
537 Ecuador as well as along the Indian coasts, in Indonesia and in Malaysia. Some increases are noted
538 in a few regions including northern Europe, eastern Canada (Québec) and northeastern United
539 States (East Coast, Chicago). Such specific situations will be further discussed in Section 4.

540 The changes in the monthly mean surface concentration of the hydroxyl radical (OH), which

541 provides indications about the change in the oxidation potential of the atmosphere, is characterized
 542 during the month of February by an increase to typically 30-40% in northeastern and northwestern
 543 China. At the same time, the model highlights a decrease in the southern and southwestern regions
 544 of the country. During the month of March and April, an increase of OH concentration has become
 545 apparent in Northern Europe. The level of OH, however, decreases in the southern part of Europe.
 546 In the populated regions of Canada and the northeastern United States, OH concentration
 547 anomalies are positive. In the southern hemisphere, the OH concentration usually decreases, except
 548 in urban areas such as Santiago, Buenos Aires, Sydney and Melbourne, where positive anomalies
 549 are derived. The level of OH is also reduced during the pandemic, along the ship tracks.

550 Formaldehyde, which is directly emitted from combustion and industry, is also produced as an
 551 intermediate species in the photooxidation of primary hydrocarbons, a process that is influenced
 552 by the presence of nitrogen oxides. For this oxygenated VOC, we note a reduction in the surface
 553 concentration of 10-30% in China during February 2020. In April, reductions of the same order of
 554 magnitude are found in Canada, southern Europe, South Africa as well as along the Pacific and
 555 Atlantic coasts of South America.



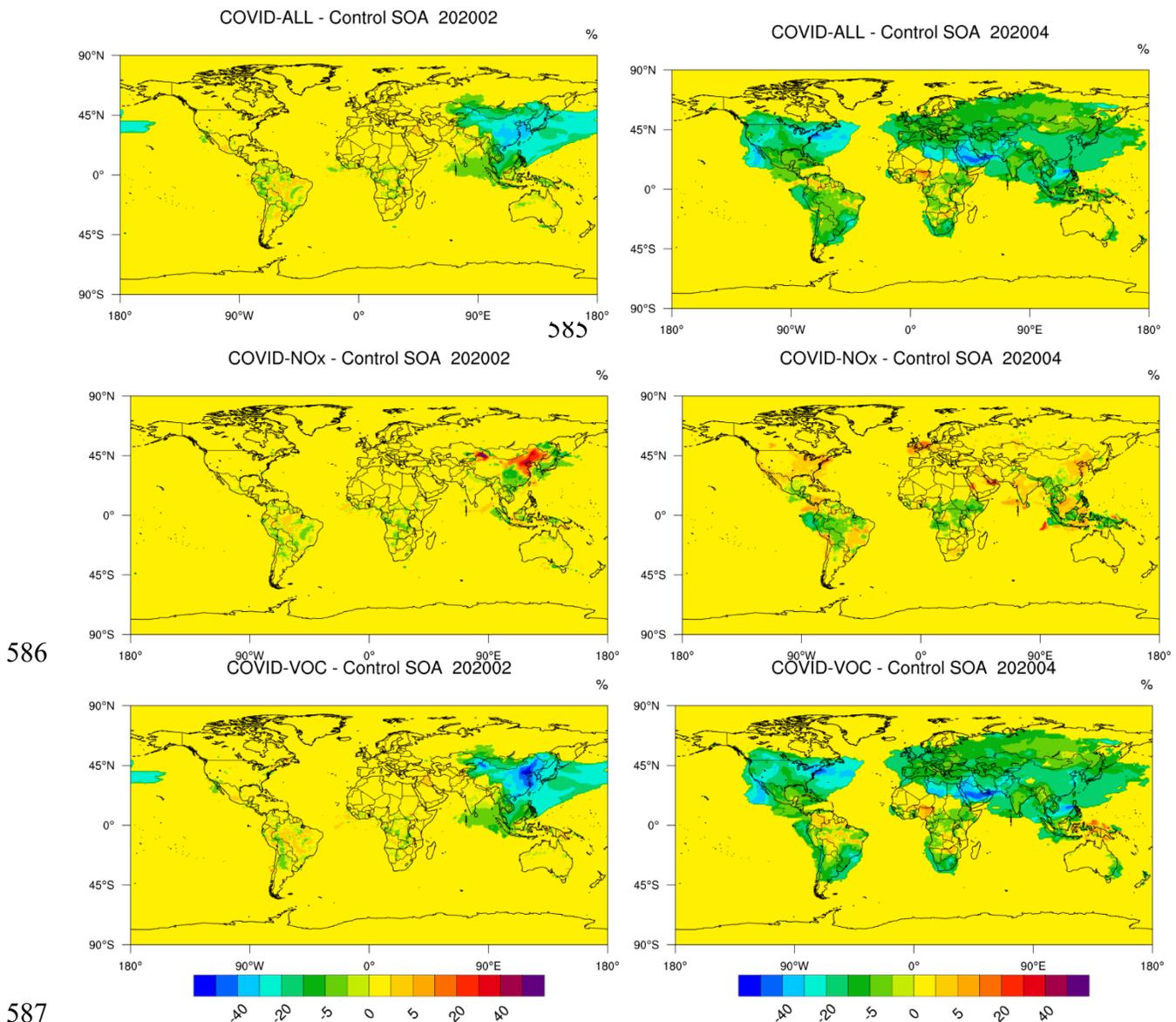


560 **Figure 5.** Relative change (percent) in February and April 2020 in the global monthly mean concentration
 561 of (from top to bottom) of NO_x, ozone, OH and PM_{2.5} resulting from the change to the adopted surface
 562 emissions of primary pollutants during the COVID-19 pandemic period.

563

564 When considering the changes in particulate matter as calculated by the model, we note (Figure 5)
 565 that the concentration of PM_{2.5} first decreased by nearly 20% in China during the month of
 566 February. Later in April, it was reduced by 10 – 20% in India, the US and Canada and by about
 567 10% in Europe. A fraction of this reduction is attributed to the decrease of the direct emission of
 568 particulate matter during the pandemic. However, the change in the emission of gas-phase
 569 precursors and in their photo-oxidation processes under reduced NO_x concentrations must also be
 570 considered. We address this issue by examining the changes affecting the quantity of SOA as
 571 derived by the model, based on the oxidation scheme described by Tilmes et al. (2020). Figure 6
 572 shows that, according to the model (COVID-All case), the SOA concentration was substantially
 573 reduced, during February 2020 in China (up to 40 %) and later during April in other parts of the
 574 world including the eastern US and a large area of South America. A fingerprint of the reduced
 575 SOA concentration extends in a plume over the northern Pacific Ocean. Interestingly, if only the
 576 NO_x emissions had been reduced (case 4 or COVID-NO_x), the concentration of SOA would have
 577 increased in northern part of China (February and April) as well as in the region surrounding the

578 English Channel (April) where the oxidation capacity increased after the pandemic outbreak. A
 579 smaller increase in SOA is seen in India, eastern Brazil and the eastern US. If only the VOC and
 580 CO emissions had been reduced (case 5 or COVID-VOC), the SOA concentration would have
 581 decreased everywhere. The patterns of the SOA in response to the combined decrease in NO_x and
 582 VOCs/CO emissions is similar to the patterns derived for the COVID-VOC case, but with smaller
 583 concentration reductions in high NO_x regions such as northern China during wintertime.
 584



587
 588 **Figure 6.** Relative change in the concentration of secondary organic aerosols (SOA) resulting from reduced
 589 emissions of primary pollutants during the COVID-19 pandemic for February (left panels) and April 2020
 590 (right panels). Upper panels: all emissions reduced; middle panels: reduction of NO_x emissions only; lower
 591 panels: reduction in VOC and CO emissions only.

592

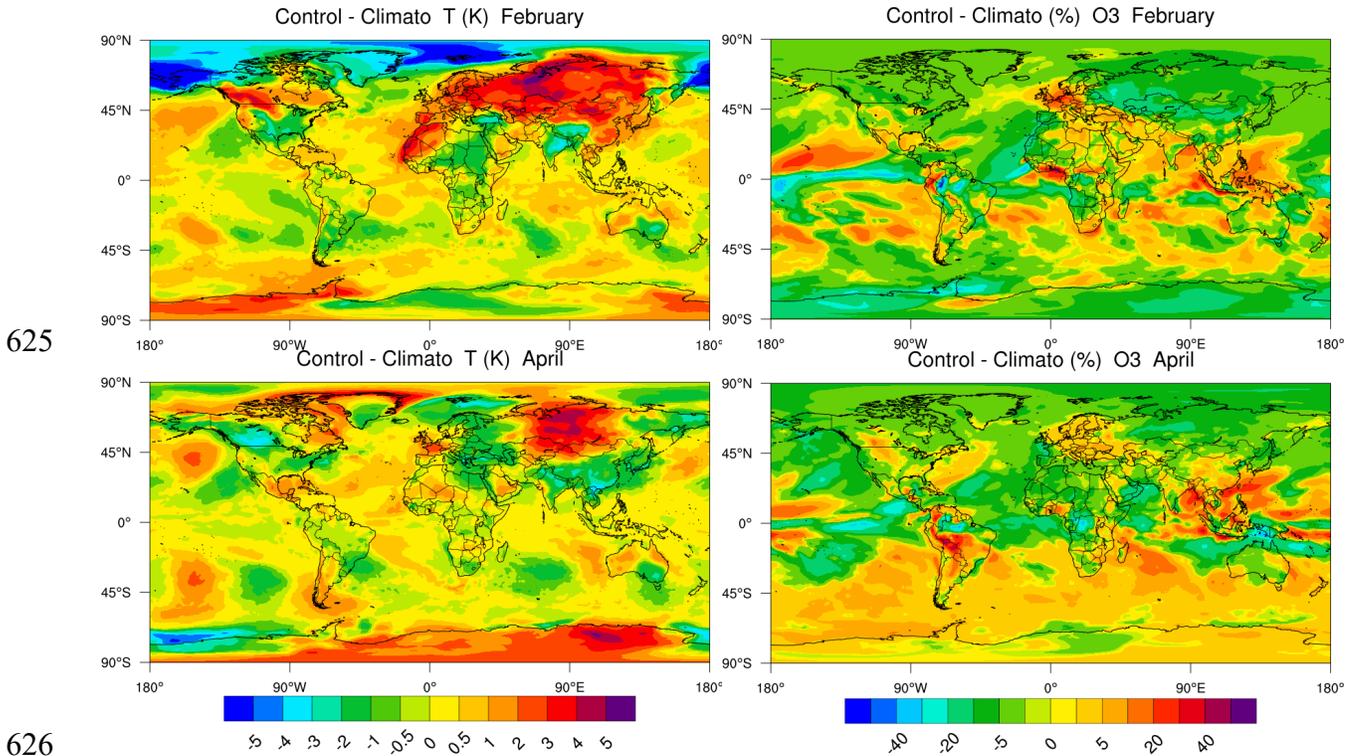
593 **4.2 Effect of meteorological anomalies**

594 The analysis of observed changes in the chemical composition during the pandemic must carefully
595 assess the influence of meteorological variability and, when examining monthly mean values, of
596 weather anomalies for the month under consideration. The early months of 2020 were strongly
597 affected by weather events, for example by the passage of two storms (Ciara and Dennis) in
598 northern Europe during the month of February and the influence of two other storms (Karine and
599 Myriam) in southern Europe (Petetin et al., 2020; Barré et al., 2020). We assess to what extent
600 meteorological variability during the pandemic has generated variations in the calculated chemical
601 fields. This information should help in the analysis of observations that are affected by both the
602 COVID-related changes in the emissions and by weather anomalies (combined atmospheric
603 dynamics, temperature, cloudiness, precipitation, atmospheric stability, etc.). For this purpose, we
604 derive the difference in the surface temperature and monthly mean ozone concentrations in 2020
605 (COVID-All case) relative to the average concentrations derived from a model simulation
606 conducted for five consecutive years (called here “pseudo-climatology” for the period 2016 to
607 2020). In this last case (referred to as the Climato-case or Case 2), the surface emissions are subject
608 to their usual seasonal variations, but their values are repeated from one year to the other. The
609 corresponding global fields of surface temperature and percentage ozone anomaly, calculated for
610 February and April 2020, are shown in Figure 7.

611 In February, besides variations occurring over the oceans, we note a small impact of the mesoscale
612 weather situation on the monthly mean ozone fields in China during the COVID pandemic
613 (February 2020). A positive ozone anomaly of 5 – 10%, however, is seen along a line that stretches
614 from northern India to Europe. This anomaly reaches about 10% in northern Europe including the
615 north of France, the Benelux countries, the UK and Germany. In Spain, the ozone anomaly is
616 negative. A negative anomaly of up to 20% is derived in northern China, Mongolia and Russia. In
617 the US, a positive anomaly of a few percent is seen in the vicinity of Chicago and along the Rocky
618 Mountains, while there is a small negative anomaly elsewhere. In South America, the largest ozone
619 anomaly is found along the Andes in Peru and Ecuador.

620 In April, the patterns of variations relative to our 5-year pseudo climatology are characterized as
621 follows: positive anomalies in southern China (5 – 10%) and in South Asia (20 – 30%), in northern

622 and eastern Europe (5 – 10%) as well along the Rockies in the US and Canada (10 – 20%) and
 623 along the Andes in Argentina, Bolivia, Peru, Ecuador and Colombia (20 – 30%); negative anomaly
 624 in Russia (10 – 20%), in Spain and the southwest of France (5 – 15%).

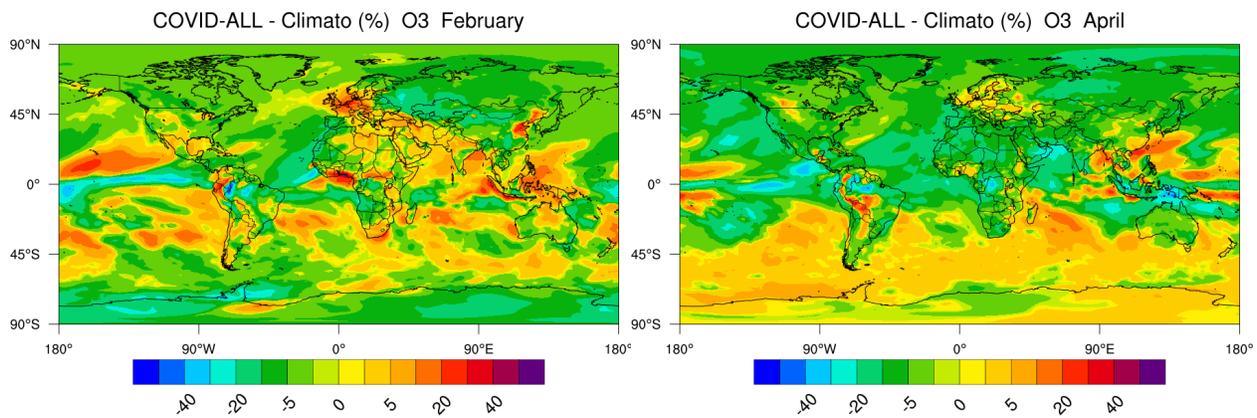


627 **Figure 7.** Anomalies in monthly mean surface temperature (K) and ozone concentration (percent) in 2020
 628 relative to a 5-year monthly mean (“pseudo-climatology” for 2016 -2020) highlighting the perturbation
 629 effects of the meteorological situation during the year of the pandemic (2020).

630

631 **4.3 Combined effects**

632 Finally, we show in Figure 8 the response in February and April 2020 of the surface ozone
 633 concentration to the *combined* effects of the entire COVID-related emission adjustments and of
 634 the meteorological anomalies. This purpose is to reproduce as closely as possible the real changes
 635 in ozone relative to the monthly mean values averaged over 5 years (2016-2020) without
 636 accounting for long-term trends in emissions (COVID-All minus Climato cases). In February, the
 637 model produces positive anomalies for ozone in northern China, northwestern Europe, in the center
 638 of the US, in the region of the Great Lakes and in the Middle-East. In April, ozone is higher than
 639 the pseudo-climatological values in northern Europe, in southern China, along the Rockies near
 640 the US-Canadian border, along the Andes in South America and in the eastern Pacific.



641

642 **Figure 8.** Relative change (percent) in February and April 2020 in the global monthly mean concentration
 643 of ozone resulting from the combined changes in surface emissions of primary pollutants during the
 644 COVID-19 pandemic period and the meteorological anomalies during the same period.

645 We summarize the results of our model simulations (Table 3) by providing values (orders of
 646 magnitude) that characterize the ozone changes in different populated regions of the world during
 647 the middle of the pandemic (monthly mean values for February in China and for April in the rest
 648 of the world). We compare the relative importance of the contributions of chemistry (reduced
 649 emissions) and meteorological anomalies in 2020; we also show the response to the two combined
 650 forcing processes.

651 **Table 3.** Relative changes (orders of magnitude in percent) in the monthly mean values of the
 652 surface concentrations of ozone as calculated for different regions (non-urban conditions).
 653 Changes due to modified emissions during the pandemic, to specific meteorological anomalies of
 654 2020 (relative to the average from a “pseudo-climatology of years 2016-2020) and to the combined
 655 effects.

Region	Emission Adaptation	Meteorological Anomaly	Combined Effects
February 2020			
North China Plain	0 to +30	0 to +5	+20 to +30
Southern China	-10 to -5	-5 to +5	-20 to -5
April 2020			
India	-15 to -5	-5 to +5	-20 to -10
Northern Europe (UK, Benelux,	+2 to +5	+2 to +5	0 to +5

Germany, northern France)			
Southwestern Europe (south of France, Spain)	-10 to -5	-20 to -5	-20 to -10
Northeastern US and southern Canada	+2 to +5	-5 to +2	-2 to +20
Eastern Brazil	-25 to -10	-5 to +15	-20 to -10
Peru Ecuador	-35 to -25	+5 to +25	-20 to -10
South Africa	-10 to -5	-5 to +2	-15 to -5

656

657

5. Process analysis and discussion

658 To identify the chemical processes that explain the changes in the concentrations of secondary
659 pollutants (e.g., ozone, SOA), we now examine, in more detail than in Section 3, the response of
660 a set of chemical species, which contribute to the formation and destruction of these secondary
661 pollutants. We focus on several regions of the world, which are differentiated by the intensity of
662 incident solar radiation and by environmental conditions such as, for example, the level of nitrogen
663 oxides in the boundary layer. We take advantage of the fact that the season that corresponds to the
664 lockdowns was different in different regions of the world. To quantify the respective role of
665 nitrogen oxides and carbon compounds, we consider in addition to the simulations (COVID-All)
666 considered in Section 3 two additional cases: in one of them (COVID-NO_x), only the reduction of
667 nitrogen oxide emissions are taken into account, while in the second case (COVID-VOC), only
668 VOC and CO emissions are reduced.

669

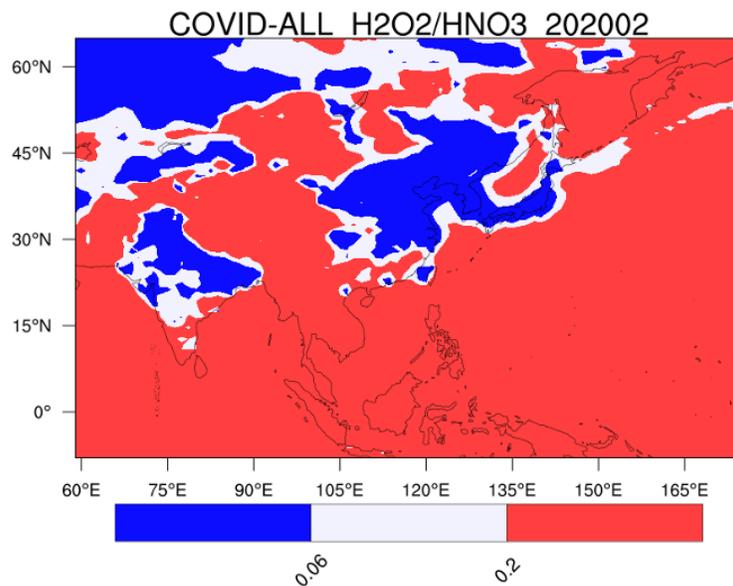
670

5.1. Air quality in China during the pandemic

671

672 Our first case focuses on the significant changes that took place in China during the 2020 lockdown
673 (Zhang et al., 2020). To analyze the response of secondary species, it is first useful to determine
674 the distribution of chemical regimes (VOC/NO_x control of ozone) during the winter period
675 (February). To estimate if a region is NO_x-limited or NO_x-saturated (VOC-limited), we represent
676 in Figure 9 the ratio R of the production of H₂O₂ relative to the production of HNO₃. When R is

677 greater than 0.2 (red zone), the ozone production is controlled by the level of nitrogen oxides,
 678 while if it is less than 0.06 (blue zone), the region is NO_x-saturated and the ozone formation is
 679 controlled by the atmospheric level of VOCs (Tonnesen et al., 2000; Zhang et al., 2009; Fu et al.,
 680 2020b). The white zone shown in the figure corresponds to an intermediate situation. We note that,
 681 in continental areas where population density and the economic activity are low or moderate and
 682 over the oceans, ozone is, as expected, NO_x-limited. In the north of China, in India, Korea, Japan,
 683 Kyrgyzstan, Kazakhstan and certain highly urbanized zones (e. g., Hong Kong and Guangzhou,
 684 Taipei), ozone is NO_x-saturated and therefore VOC-limited. This condition corresponds to a
 685 winter and early spring situation. In summer, however, when the concentration of NO_x is lower,
 686 the area with VOC-limited conditions is reduced. In China during the lockdown period, the limit
 687 between the VOC and NO_x controlled regions is located along a line extending from
 688 approximately Lanzhou in the center of China to Xiamen along the ocean in the vicinity of Taiwan.
 689 Inside the NO_x-limited regions, urban centers are often VOC-limited areas.



690

691 **Figure 9.** Ratio between the production rate of hydrogen peroxide and nitric acid, a measure of the chemical
 692 conditions governing the formation of ozone. Geographical areas in which ozone is NO_x controlled (red)
 693 and NO_x-saturated or VOC controlled (blue). The white area represents an intermediate situation.

694 Figure 10 shows that, during the lockdown of February, the surface concentration of NO_x was
 695 severely reduced (40-50%) in most areas of eastern China and in the northwest of the country. At
 696 the same time, the concentration of ozone increased in the northeastern part of China and locally
 697 in several large urban areas of other regions as evidenced by surface observations (e.g., Shi and
 698 Brasseur, 2020; Huang et al., 2020; Liu and Wang, 2020). Further, a reduction in ozone occurred

699 in the southern part of the country. This result is consistent with the results of the regional modeling
700 study of Liu and Wang (2020) and with surface observations (e.g., Lian et al., 2020; Fu et al.,
701 2020b). To further address this question, we show how surface NO_x and ozone would have
702 responded according to the model if only the emissions of VOC or of NO_x had been reduced.

703 **5.1.1 COVID-VOC case: Reduction only in the VOC and CO emissions**

704 If *only* VOC and CO emissions are reduced, while the emissions of other species including NO_x
705 remain unchanged in China during February 2020, the ozone and peroxyacetyl nitrate (PAN)
706 concentrations as calculated by the model decrease in the North China Plain by 20-30% and 30-
707 50%, respectively. A substantial reduction in the concentration levels of hydrogen radicals (HO_x
708 = OH + HO₂) also occurs, but the concentration of NO_x slightly increases due to the reduced loss
709 rate via HNO₃ formation, the reduced formation of organic nitrates and peroxy nitrates and the
710 reduced uptake of NO_x by SOA. With the adjustment factors adopted for VOCs, the model derives
711 a reduction of around 30% for OH, 50% or higher for HO₂, 50% for CH₃O₂, 20-30%, for
712 formaldehyde, 20% for hydrogen peroxide and 20% for nitric acid in the North China Plain (see
713 also Figure S4). The concentration of OH, however, is slightly enhanced (5 to 10%) outside this
714 particular region. The concentration of the NO₃ radical, which is a major oxidant during nighttime,
715 slightly increases (typically 2-5 %) in most regions of China except in the North China plain, where
716 it decreases by as much as 30 %. The decrease in the level of HO_x directly results from the
717 reduction in the sources of these radicals, including the reactions of alkenes with ozone, the
718 photolysis of formaldehyde and of other carbonyls and the photolysis of nitrous acid (HONO)
719 since the heterogeneous formation of this last compound on the surface of aerosol particles is
720 reduced as the aerosol concentration (including the concentration of SOA and sulfates) has
721 decreased. Under the assumptions adopted here, the HO₂ to OH ratio declines since the conversion
722 of OH to HO₂ by CO and VOCs is slowed down, while the conversion of HO₂ back to OH slightly
723 accelerates due to slightly enhanced NO_x concentrations. We note that the relatively large
724 reduction in HO₂ and CH₃O₂ (more than 50%) together with a smaller increase in NO_x (10-30%)
725 leads to a decrease in the photochemical production of ozone.

726 **5.1.2 COVID-NO_x case: Reduction only in the NO_x emissions**

727 If we make a simulation in which *only* the NO_x emissions are reduced during the pandemic, the
728 response to the chemical system is very different (opposite sign) than in the previous case. Under

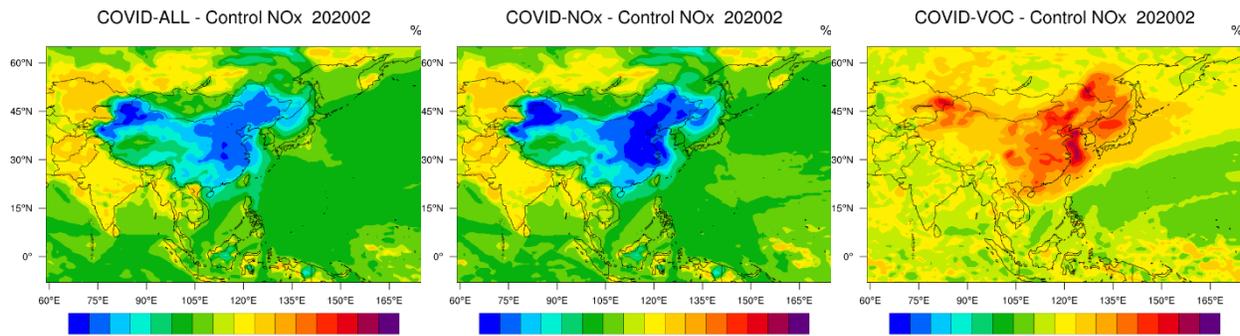
729 our assumption, the concentrations of OH and HO₂ increase by 50% or more, mostly in the
730 northeastern part of China. The concentrations of methyl peroxy (CH₃O₂) and formaldehyde
731 (HCHO) increase by 10-20%. The increase in HO_x is attributed primarily to a reduced
732 recombination of OH with NO₂, which leads to the reduction in the HNO₃ concentration derived
733 by the model. Since the photolysis of HCHO is a significant source of HO_x radicals, the increase
734 in the concentrations of OH and HO₂ radicals also results from the enhanced concentration of
735 formaldehyde (Li et al., 2020). The reduction in the NO concentration tends to shift the balance
736 between HO₂ to OH towards HO₂. The concentration levels of the NO₃ radical and of PAN are
737 enhanced in northern China and particularly in the region of Beijing (up to 50% for both species),
738 but are reduced in southern China. The response of ozone (increase in northern China and decrease
739 in southern China) results from synergetic changes in both the production and destruction rates of
740 the molecule. First, the simultaneous reduction in NO_x, and enhancement in HO₂ and CH₃O₂
741 concentrations result, according to the model, in a *reduced* photochemical ozone production rate
742 of 20-30%. Second, the titration of ozone by NO₂, a major loss for ozone in the highly polluted
743 areas of northern China, is reduced, while the direct ozone loss due to the enhanced levels of OH
744 and HO₂ increased. Taking into consideration these two processes acting in different directions,
745 we find a resulting ozone loss that is *reduced*. This suggests that the most important factor
746 explaining the ozone increase in northern China is the reduction of the ozone titration by NO₂. In
747 southern China, where the background levels of nitrogen oxides are lower and solar radiation
748 intensity is higher, the reduction in NO_x has led to an *enhanced* net ozone destruction and hence
749 a reduction in the surface concentration of this molecule except in cities where the ozone
750 concentration increases. As expected, the net ozone production rate calculated for February 2020
751 (Figure 8) is positive in northeastern China and negative in other regions.

752 **5.1.3 COVID-All case: Reduction in the NO_x, VOC, CO and aerosol emissions**

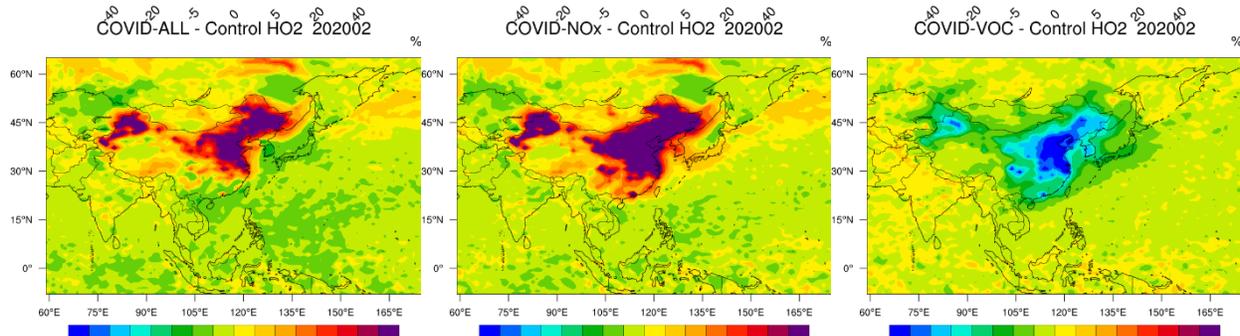
753 The response of the surface composition, when all emission adjustments for the emissions are
754 taken into consideration, leads to an intermediate situation between the two cases described above.
755 In fact, the response of most chemical species to the NO_x and VOC emission reduction generally
756 happens in opposite directions. When the two effects are combined, the reduction in NO_x is of the
757 order of 40% in the North China Plain and in the northwest of the country. The increase in HO₂
758 and CH₃O₂ is of the order of 50% and that of OH around 30%. The mean concentration of the NO₃
759 radical increases by up to 50% in the urbanized regions of Beijing and Shanghai. The decrease in

760 HCHO concentration is limited to 10-20% (Figure S4). PAN and SOA concentrations decrease
 761 only by a few percent in the northern China, but decrease more substantially (20-30%) in the
 762 central and southern parts of the country. The exact quantitative response of PAN and SOA
 763 depends critically on the relative amplitude of the VOC and NO_x emission reduction. It would
 764 have been positive in the North China Plain during February if the adopted VOC emission
 765 reduction had been somewhat smaller. In the case of SOA, a plume with decreased concentration
 766 values is noticeable over Korea, Japan and the Western Pacific Ocean. The change in ozone is
 767 positive in the northeastern part of China (about 30%) and negative in the southern part of the
 768 country (about 10%). As seen in the figures, the most pronounced changes in the concentration
 769 level of most chemical species are located in the North China Plain and further north. Additional
 770 model results are provided in Figure S4 of the Supplementary Information.

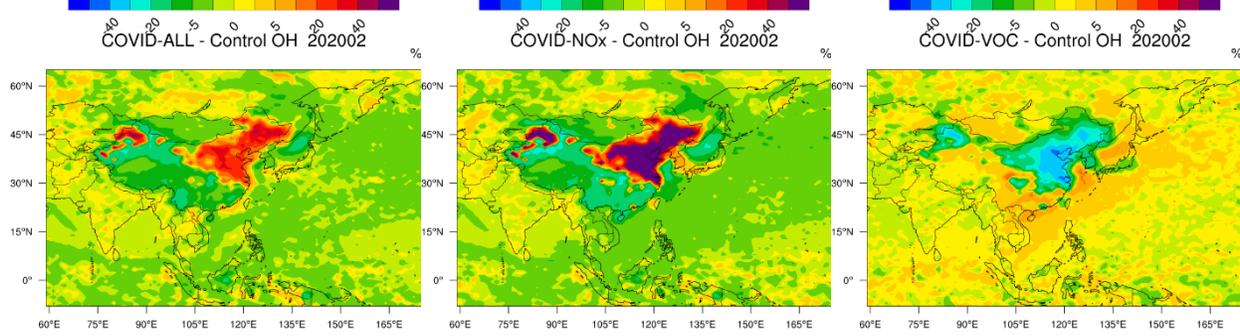
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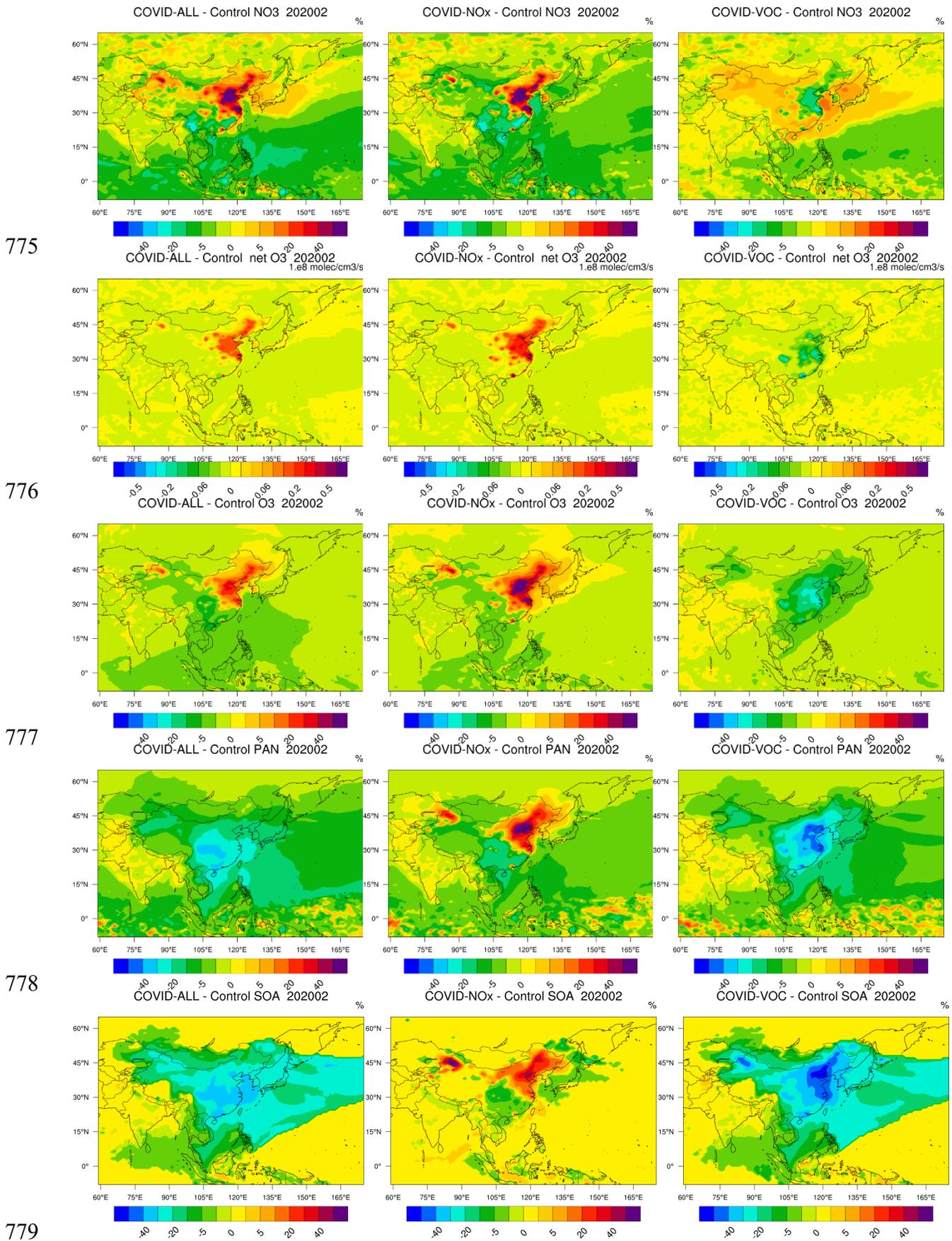
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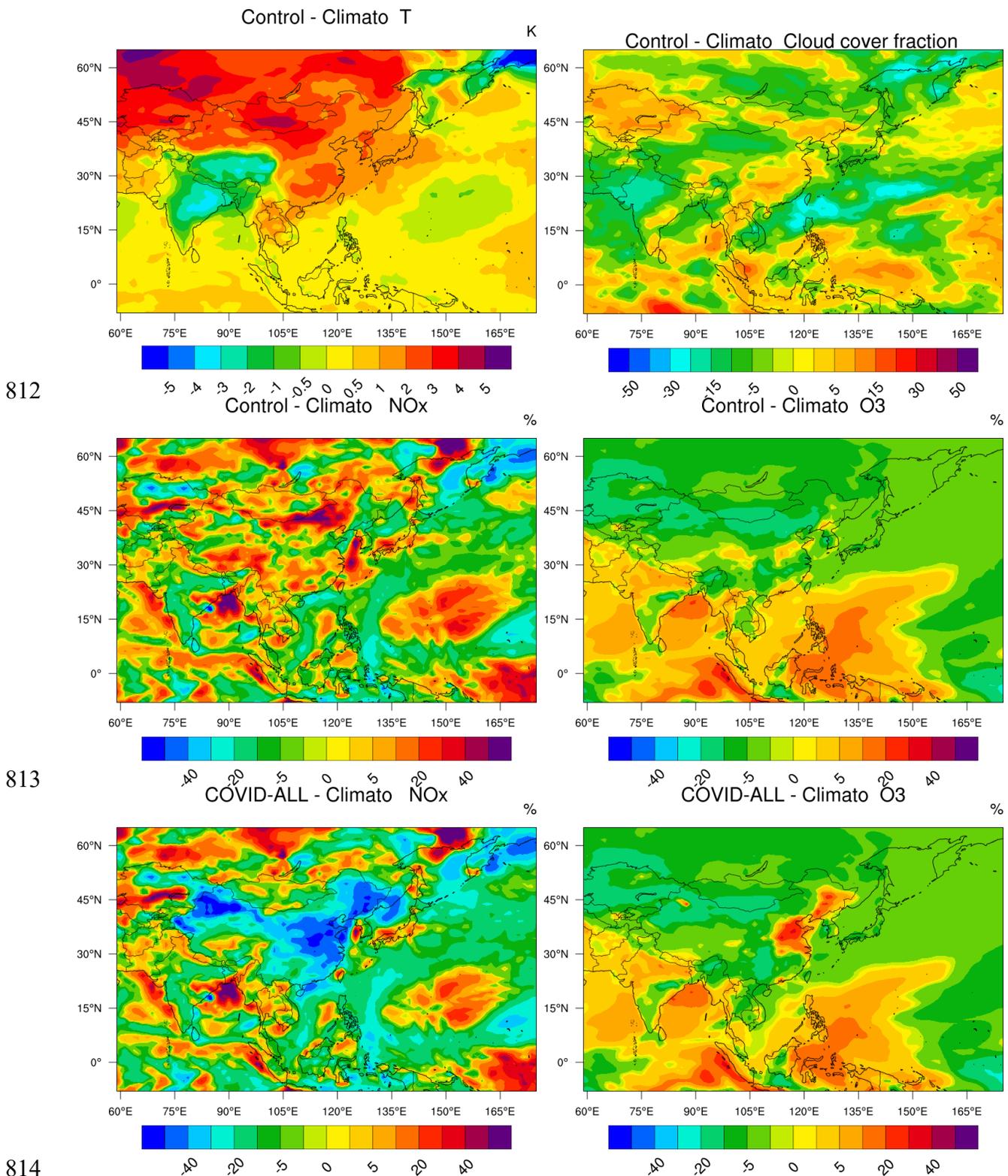
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780 **Figure 10.** Percentage change in several chemical variables in China in response to reduced emissions of
781 primary pollutants in February 2020 during the COVID-19 pandemic. Panels from the top to the bottom:
782 NO_x, HO₂, OH and NO₃; net ozone production (cm⁻³ s⁻¹), ozone, PAN and SOA. Left column: reduction in
783 all emission; center panel: reduction in NO_x emissions only; right panel: reduction in VOC and CO
784 emissions only.

785
786 In order to provide some insight on the relative forcing effects of the emission reduction during
787 the pandemic and of the meteorological variability, we provide in Figure 11 an estimate of the
788 ozone anomaly generated by weather dynamics and by the combined effects of the two forcing
789 factors. Wang and Zhang (2020) provide a detailed assessment of the effects of meteorological
790 elements during the pandemic period. Our model simulations as nudged towards the GEOS-FP
791 meteorology show that during February 2020 and relative to our 5-year “pseudo-climatology”,
792 Eastern China was abnormally warm by 1.5 to 2.5 K and subject to high cloud fraction; northern
793 China was 2-4 K warmer with cloud fraction lower relative to the previous 5 year average. During
794 this month, ozone anomalies associated with meteorological variability were dominant in the
795 tropical regions south of China, but were relatively weak on the Chinese mainland. Abnormally
796 low ozone was found along the border between China and Mongolia related to the abnormally high
797 NO₂ concentration calculated during February 2020. The increase in the monthly mean ozone
798 concentration in the North China Plain (up to 5%) predicted by the model in response to
799 meteorological anomalies adds to the ozone perturbation caused by the reduction in emissions.
800 Our simulations suggest that chemical disturbances rather than meteorological anomalies explain
801 the ozone concentration increase in the North China Plain during February 2020. Shorter time
802 fluctuations linked to specific weather conditions should be considered in a finer analysis to
803 explain, for example, the acute air pollution episodes reported in several urban areas during
804 January and February 2020 (Wang et al., 2020). In southern China, where the perturbed chemistry
805 tended to reduce ozone, a small positive anomaly is visible along the South China Sea. The change
806 resulting from the two simultaneous effects is however negative except in the urban zone of
807 Guangzhou/Hong Kong/Macao. In short, the enhancement in the level of oxidants in the North
808 China Plain appears to be primarily a direct consequence of the reduction on chemical emissions
809 triggered by the pandemic, but could have been facilitated by unfavorable weather conditions.

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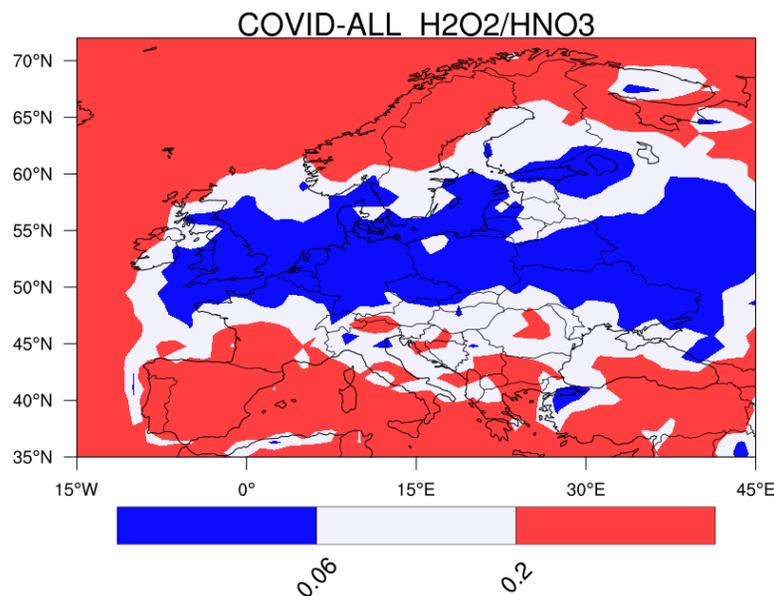
815 **Figure 11.** Percentage change in the monthly mean ozone surface concentration in Asia for February 2020
 816 relative to the value averaged over 5 years (2016 to 2020) for the same period of the year. Left Panel:
 817 Response taking into account the adjustment of the emissions associated with the pandemic and the
 818 meteorological anomaly. Right panel: ozone response only to the meteorological anomaly.

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5.2. Air quality in Europe during the pandemic

822 The situation in Europe during the pandemic is somewhat similar to what was derived in China
823 including a small increase of the ozone concentration in highly polluted areas. We first show in
824 Figure 12 that, during the period of the lockdowns (15 March to 15 April), the ozone production
825 in most regions of Europe was controlled by NO_x except in the most densely populated areas
826 where the influence of VOC was significant.

827



828

829 **Figure 12.** Ratio between the production rate of hydrogen peroxide and nitric acid, a measure of the
830 chemical conditions governing the formation of ozone. The geographical area in which ozone is NO_x
831 controlled is shaded in red and VOC controlled in shaded in blue. The white area represents an intermediate
832 situation between fully NO_x and VOC controlled situations.

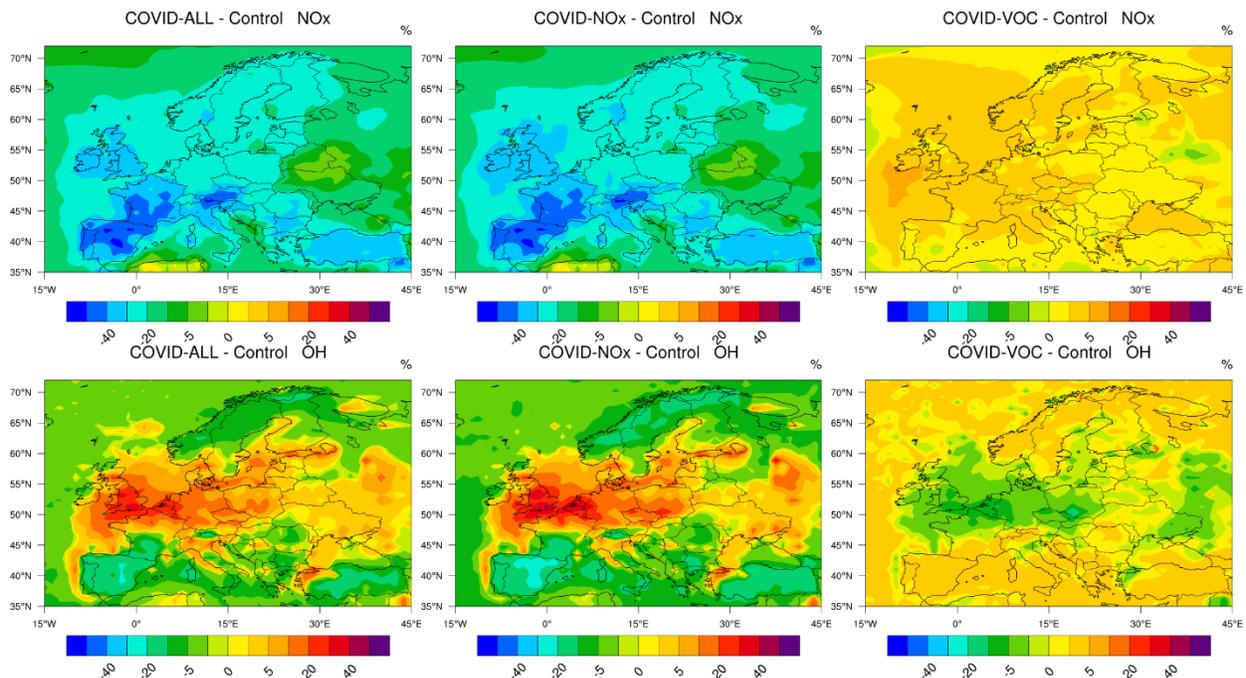
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834 In Figure 13, we show that, in March-April 2020, the relative reduction in NO_x concentrations
835 associated with the reduced emissions covers the entire European continent, but with the most
836 pronounced effects occurring in the western and southern part of the continent (30-50% in areas
837 of Spain, France and Switzerland; 20-30% in Germany Eastern Europe and Scandinavia). This
838 reduction is accompanied by an increase in the level of photo-oxidants (OH, ozone) that is most
839 pronounced in the UK, Belgium, the Netherlands, northern France and in the western part of
840 Germany. The ozone increase in this area is typically 5 to 10%, while the OH increase reaches
841 30%. In southern Europe, ozone concentrations are reduced by 5 to 10%. The net ozone production

842 slightly increases over most of the continent with a notable exception in Spain. The largest values
 843 are found again in the region extending from the UK to western Germany with hot spots in several
 844 urban or industrial areas. These patterns of ozone change are consistent with the regional model
 845 simulations performed by Menut et al. (2020) for western Europe. They derive on 28 March 2020
 846 ozone anomalies (relative to a “business as usual” reference case) that are positive in the
 847 geographical area extending from in northern France and the UK to Germany and Poland.
 848 Negative anomalies are found in southern France and Spain. Ozone concentrations are also
 849 abnormally high in the Po valley (northern Italy) and in several large European cities, including in
 850 southern Europe (Madrid, Barcelona, Rome, Naples Marseille, Toulouse).

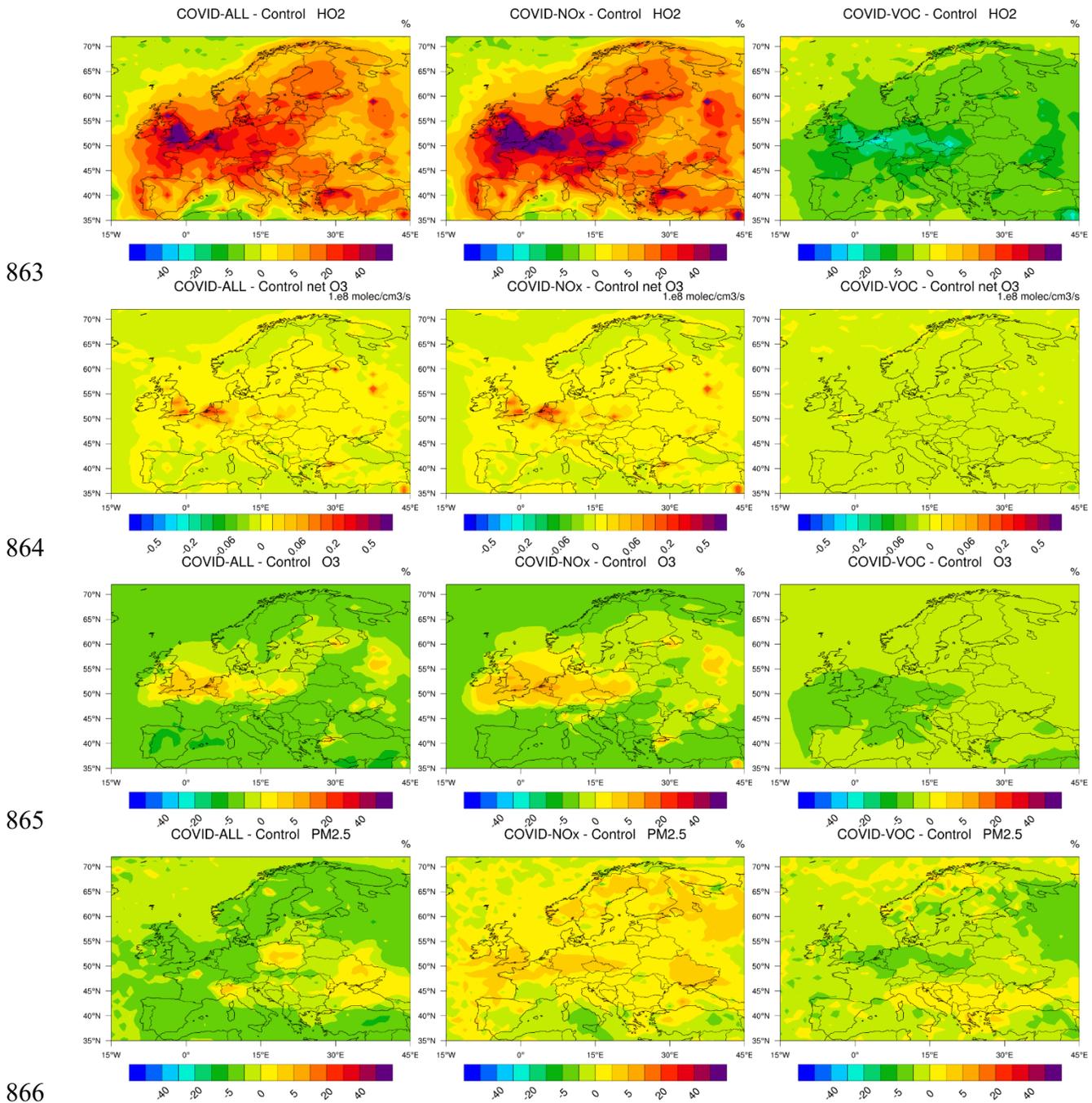
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 852 The same type of behavior is found in our global model when only NO_x emissions are reduced,
 853 but with reinforced changes in secondary products. When only VOC emissions are reduced, ozone
 854 decreases by 2 to 5 percent with the largest response located in an area extending from the Atlantic
 855 to Germany in the vicinity of the English Channel. In this area, OH concentrations are 5 to 10%
 856 lower than in the baseline case. Finally, we note again, in this particular case, a slight increase in
 857 the concentration of NO_x (2 percent with higher values of 5 percent over the sea east and west of
 858 the UK) resulting from a reduced conversion of nitrogen species to nitro-organic compounds.

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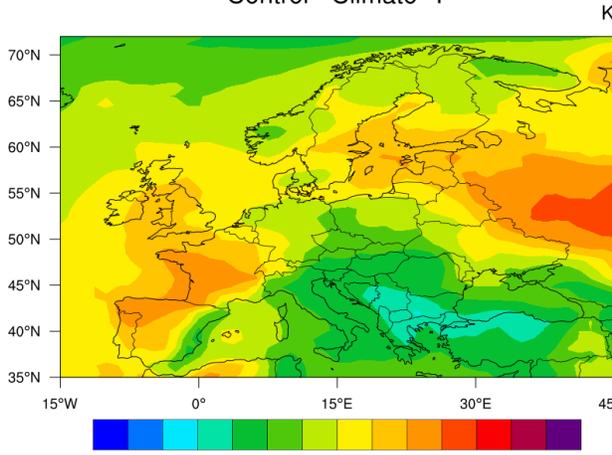
867 **Figure 13.** Percentage change in the surface concentration and in the net ozone production across Europe
 868 in response to the emissions of primary pollutants adjusted for the COVID-19 pandemic period of 15
 869 March-14 April 2020. From top to bottom: NO_x, OH, HO₂, net ozone production ($\text{cm}^{-3} \text{s}^{-1}$), ozone
 870 concentrations. Left column: reduction in all emissions; center panel: reduction in NO_x emissions only;
 871 right panel: reduction in VOC and CO emissions only.

872 We now examine the effects of meteorological anomalies on the calculated changes in the surface
 873 concentrations of NO_x, HO₂ and ozone. Figure 14 shows the changes in the concentration of
 874 several chemical species in response to the anomalies in the meteorology in March-April 2020.

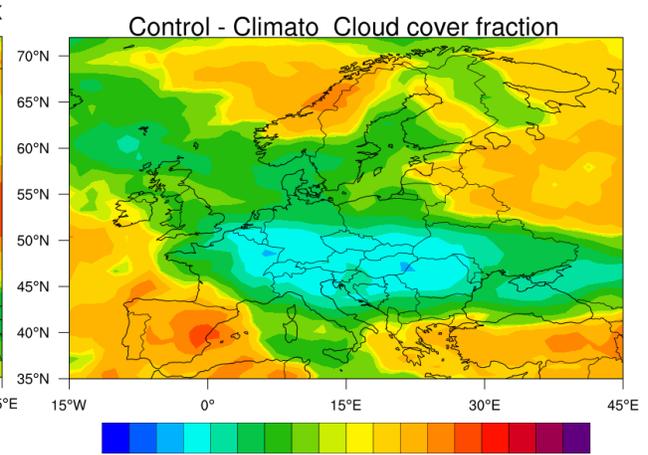
875 Meteorological analyses (Deroubaix et al., 2020; Ordóñez et al., 2020) show that this period was
876 characterized by unusual clear sky periods in central and northern Europe and cloudy skies in
877 southwestern Europe. Figure 14 also shows the anomaly in temperature and in cloud cover
878 calculated by the model for the month of April 2020 relative to a 5-year “pseudo-climatology”.
879 During this particular month, the temperature is higher than the mean value in France, in Spain,
880 near the Baltic Sea and in Eastern Europe. Abnormal low cloudiness is predicted in Central Europe
881 extending from France to the Black Sea and from Italy to Denmark. Cloudiness, however, is higher
882 than normal in Spain, Turkey and part of Norway.

883 When considering only the effect of meteorological variability and ignoring the adjustments in the
884 emissions, we see that, during the 15 March – 14 April period, the level of nitrogen oxides is
885 abnormally high at the western edge of the European continent, as well as in France and in large
886 parts of Italy and Central Europe. It is low along the eastern coast of Spain and in the southeastern
887 part of Scandinavia. The change in NO_x concentrations, when combining the emission reductions
888 and the meteorological effects (COVID-All – Climato) is more pronounced than in the “emission
889 reduction” case (see Figure 13). Meteorological anomalies play therefore a substantial role. In the
890 case of HO₂, meteorological perturbations reinforce the disturbances resulting from the changes
891 adopted for the emissions. The same reinforcement is also found in the case of ozone. In fact, for
892 this particularly species, meteorological anomalies are responsible for most of the changes in the
893 surface concentrations. The ozone increase attributed to the combined reductions in NO_x and VOC
894 emissions is visible only in the region that covers the southern UK, the Benelux, and parts of
895 Germany, as well as the eastern coast of Spain and areas in the Mediterranean. In summary,
896 contrary to what has been found for China, a large fraction of the ozone increase noted in Europe
897 during the pandemic must be attributed to meteorological anomalies (Ordóñez et al., 2020); the
898 reduction in pollutant emissions has substantially affected only a few specific regions of the
899 continent.

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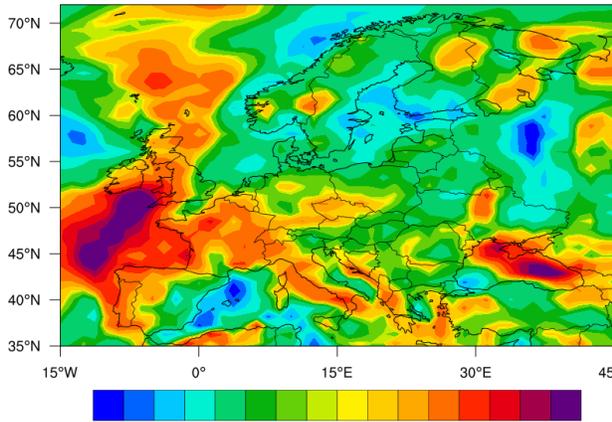


Control - Climato Cloud cover fraction

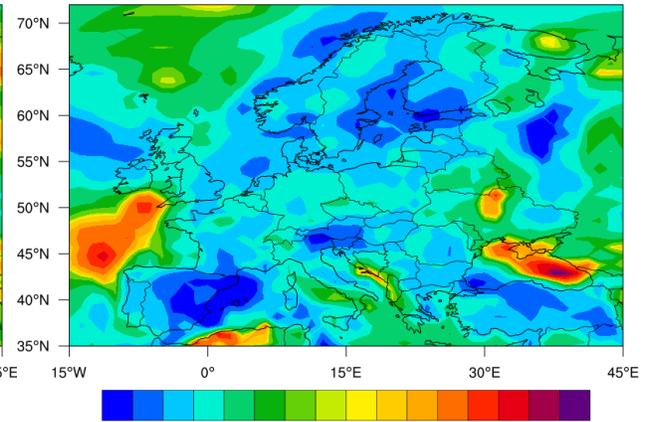


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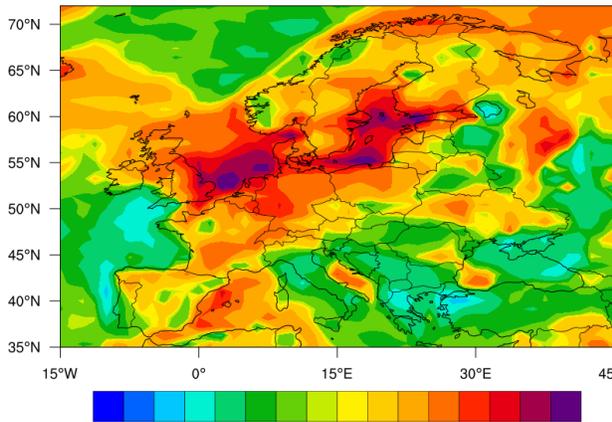


COVID-ALL - Climato NOx

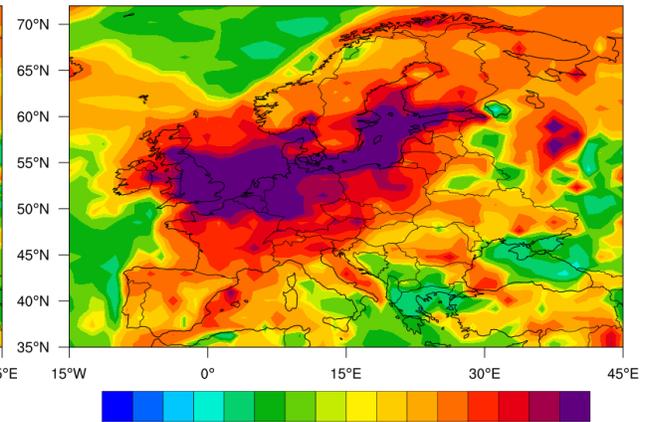


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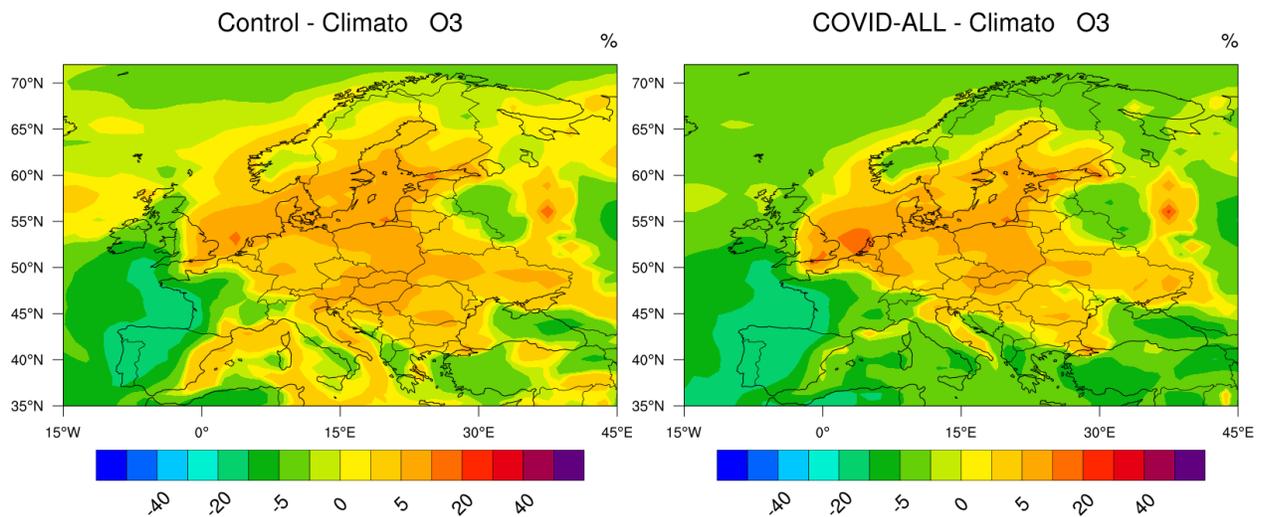
Control - Climato HO2



COVID-ALL - Climato HO2



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904 **Figure 14.** Percentage change in the surface concentration of ozone across Europe during the period 15
 905 March-14 April 2020 relative to the value averaged over 5 years (2016 to 2020) for the same period of the
 906 year. Left Panel: Response taking into account the adjustment of the emissions associated with the
 907 pandemic and the meteorological anomaly. Right panel: ozone response only to the meteorological
 908 anomaly.

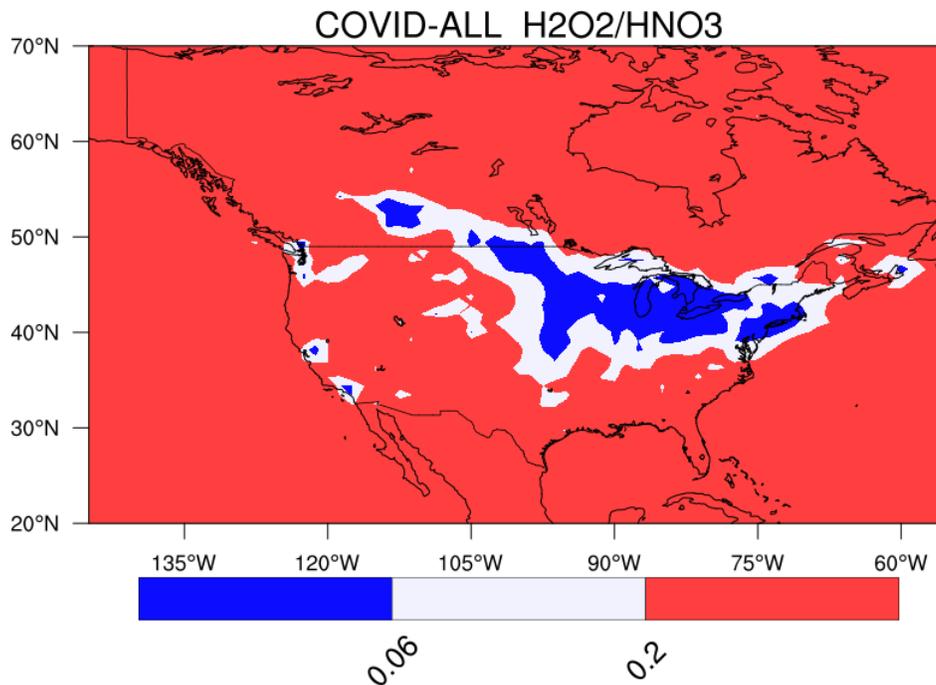
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910 **5.3. Air quality in North America during the pandemic**

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912 We now examine the results provided by the model in North America (COVID-All case) and focus
 913 again on the period 15 March to 14 April 2020. In most of the regions, particularly in rural areas,
 914 ozone is NO_x-limited during the spring conditions (Figure 15). However, in a region extending
 915 from the US East Coast to Alberta in Canada including the region of the Great lakes and part of
 916 the Middle West, ozone is VOC controlled.

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919 **Figure 15.** Ratio between the production rate of hydrogen peroxide and nitric acid, a measure of the
 920 chemical conditions governing the formation of ozone. The geographical area in which ozone is NO_x
 921 controlled is shaded in red and VOC controlled in shaded in blue. The white area represents an intermediate
 922 situation between fully NO_x and VOC controlled situations.

923

924 In relative terms, the largest decrease in NO_x concentrations is found in southern Canada (30-
 925 40%) as well as in the northeastern US (40-50%), notably near the Great Lakes and along the St
 926 Lawrence River (Figure 16). Substantial reductions in NO_x are also noticeable along the west coast
 927 (20-30%) and in the western and southern states of the US and in Mexico (20-30%).

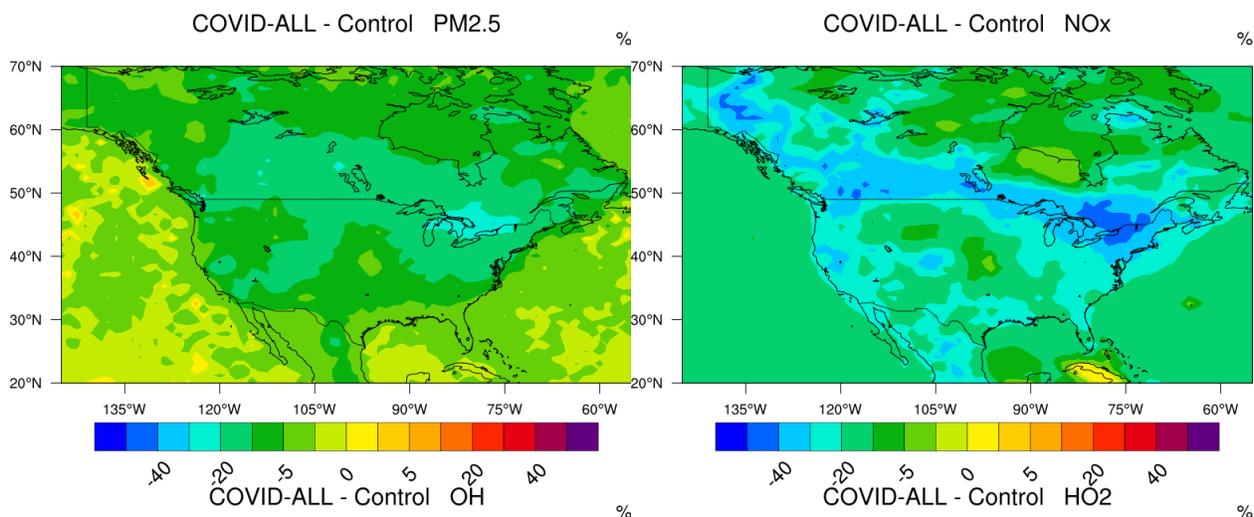
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929 The surface concentration of the hydroxyl and peroxy radicals has increased most in the region of
 930 the Great Lakes, along the US-Canadian Border (including the region of Calgary), in the central
 931 plain of the US, as well as in urban areas of the west coast including Los Angeles, San Francisco
 932 and Seattle (5-15% for OH, 30-50% for HO₂ and CH₃O₂). The reduction in formaldehyde is
 933 relatively small (less than 10%) except in southern Canada and the region of the St Lawrence,
 934 where it reaches 10 to 20% (Figure S5). The change in the net ozone production rate during March-
 935 April (see Figure S5) is limited to a few percent and so is the change in the surface ozone
 936 concentration. Since ozone is NO_x-controlled in rural areas, the reduction in NO_x leads to a small
 937 ozone decrease, mostly in the central and southern parts of the US. Only small ozone increases (2-
 938 10%) in response to the changes in emissions are noticeable in the model results for the period 15
 939 March – 14 April, and are located around the Great Lakes, particularly near densely populated

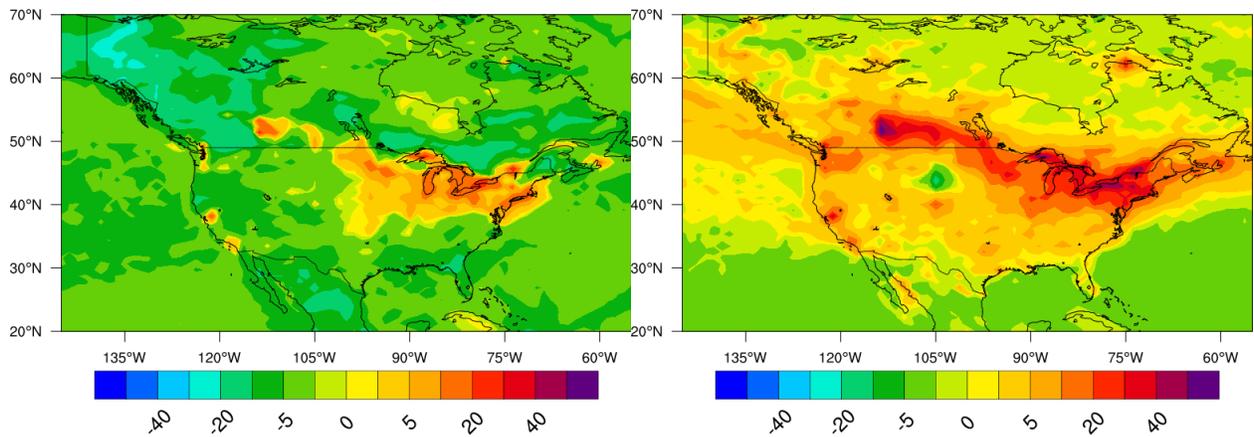
940 urban areas like New York, Boston, Toronto, Chicago, Calgary, Los Angeles and San Francisco.
 941 The bottom right panel in Figure 15 provides the response of ozone resulting from a reduction in
 942 the NO_x emissions only (no VOC and CO emission reduction, COVID-NO_x case). The patterns
 943 are the same as those discussed for the COVID-All simulations with, however, more pronounced
 944 ozone increases along the US-Canadian border and in the urban areas of the west coast. Chen et
 945 al. (2020) analyzed data acquired from 28 urban and sub-urban air quality stations across the
 946 United States that showed widespread nonuniform NO_x reductions relative to a pre-lockdown
 947 reference as well as mixed and relatively minor changes (less than 20%) in ozone. Additional
 948 model results are provided in Figure S5 of the Supplementary Information.

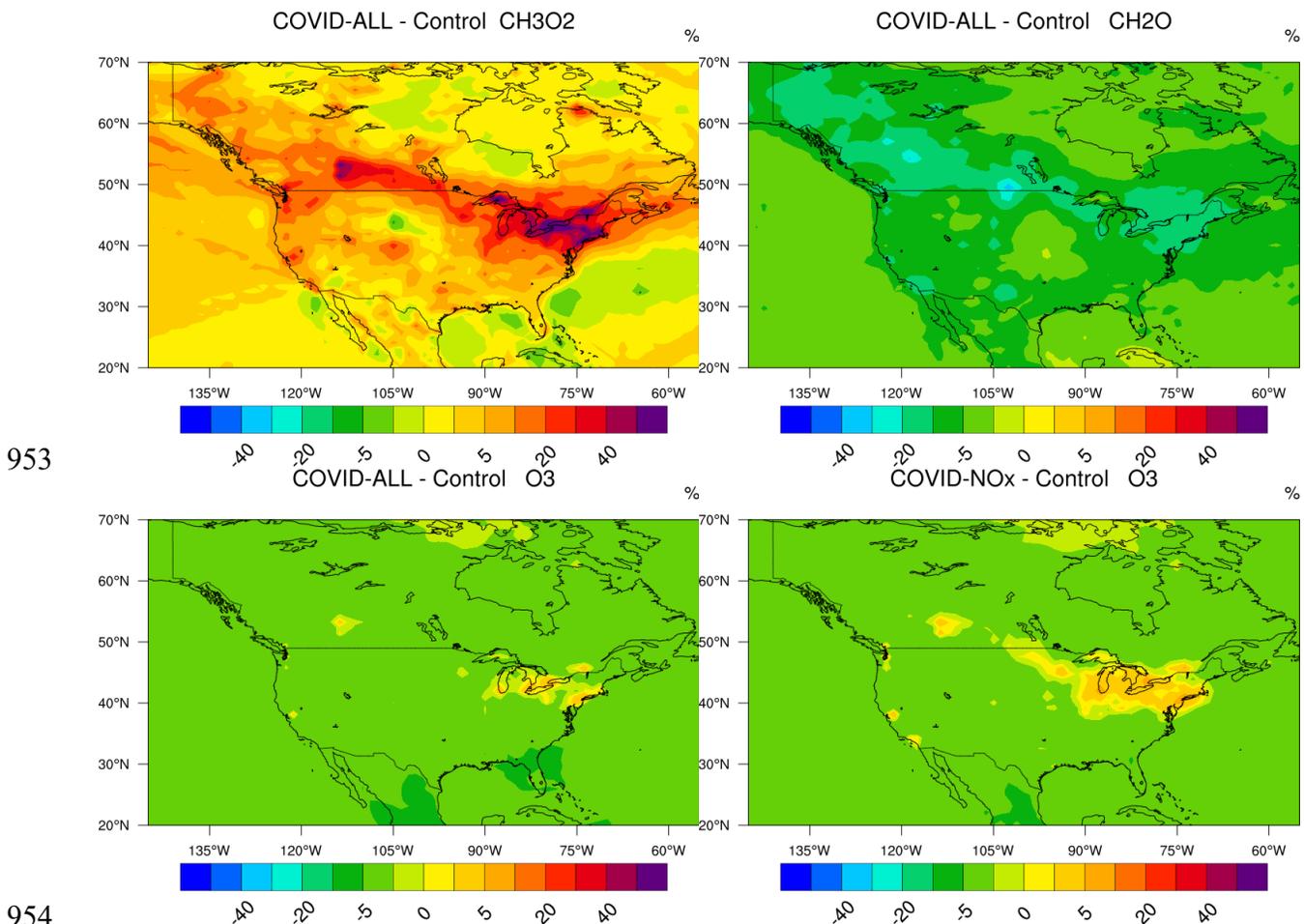
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956 **Figure 16.** Percentage change in the surface concentration of (from top left to bottom right) $\text{PM}_{2.5}$, NO_x ,
 957 OH , HO_2 , CH_3O_2 , HCHO , and ozone several chemical species across North America in response to adjusted
 958 emissions of primary pollutants during the COVID-19 period of 15 March-14 April 2020. All calculated
 959 fields result from a COVID-All simulation except the bottom right panel which is obtained from a COVID-
 960 NO_x simulation (no reduction in VOC and CO emissions).

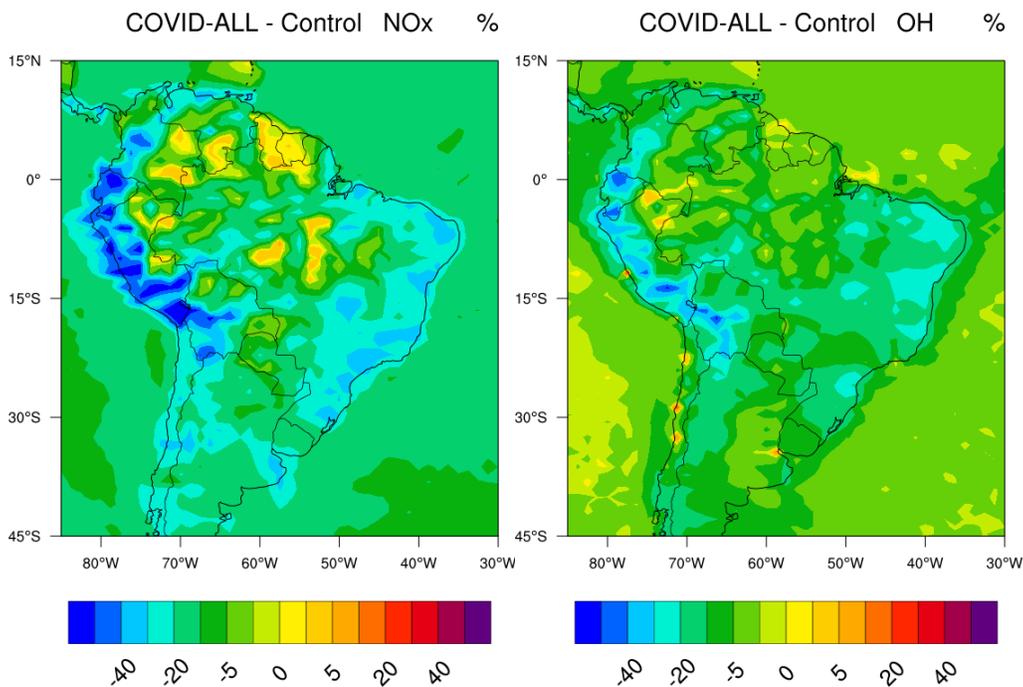
963 5.4. Air quality in South America during the pandemic

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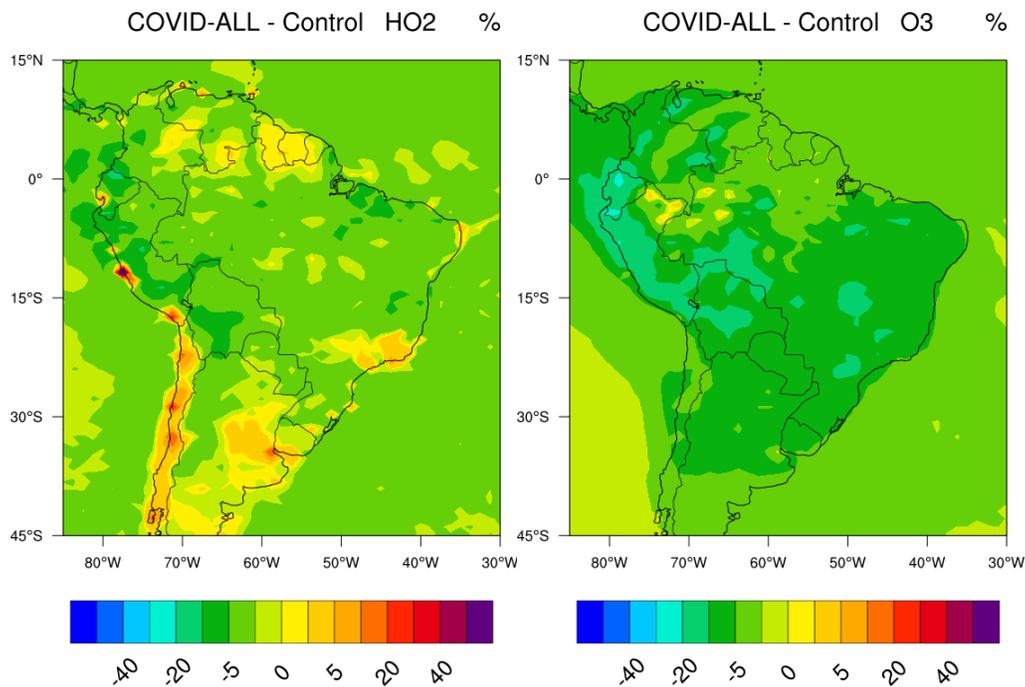
965 In South America (Figure 17), a significant reduction in the surface concentration of nitrogen
 966 oxides is derived for the period 15 March – 14 April, specifically along the Atlantic coast in Brazil
 967 (25 - 35%) and the Pacific coast in Peru and Ecuador (30 – 40%). Reductions of 30 to 40% are
 968 also found in the region of Buenos Aires, Argentina and Santiago, Chile. The reduction in
 969 formaldehyde is generally limited to a few percent across the continent since a large source of this
 970 compound is due to biogenic emissions, which is unchanged in this simulation. Except in urban
 971 areas, the level of OH decreases (20 – 25% in eastern Brazil; 30 – 40% in Peru and Ecuador).
 972 However, the concentration of HO_2 increases by 5 – 10% in Chile, eastern Brazil and eastern

973 Argentina, specifically in and near large South American metropolitan areas (Sao Paulo-Rio de
 974 Janeiro region, Buenos Aires, Santiago, Lima, Guayaquil). The concentration of nitric acid
 975 decreases along both coasts (30% in Brazil; 40 - 50% in Peru and Ecuador) and that of hydrogen
 976 peroxide slightly increases (up to 5 %) in Chile, in the region of Sao Paulo and Rio de Janeiro as
 977 well as in the northern part of the South American continent. A small reduction in the surface
 978 ozone concentration (5 – 10%) is derived in Brazil and a larger decrease (15 – 20%) is calculated
 979 in Peru and Ecuador. Cazorla et al. (2020) note that in the city of Quito, Ecuador, the average
 980 ozone level during the lockdown in April was not significantly different from the ozone level in
 981 January, which they attribute to unusually high cloudiness and to frequent precipitation during the
 982 month of April.

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Figure 17. Percentage change in the surface concentration of several chemical species across South America in response to adjusted emissions of primary pollutants during the COVID-19 period of 15 March-14 April 2020.

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6. Summary and conclusions

992 The world-wide disruption in the economic activities during the COVID-19 pandemic in early
993 2020 has generated large perturbations in the emissions of air pollutants. These perturbations have
994 been prominent first in China where the pandemic outbreak was reported, and later in other
995 countries of both hemispheres. The response of photo-oxidants to the simultaneous reductions in
996 NO_x, VOC and CO emission has varied according to the geographic location and the time of the
997 year. In the NO_x-saturated region of northeastern China, which was hit by the pandemic under
998 winter conditions, an increase in the concentrations of ozone, OH, HO₂, and RO₂ radicals was
999 derived by the model. The concentration of the NO₃ radical, a powerful nighttime oxidant, and of
1000 PAN, a secondary pollutant was also increased in the North China Plain. The reduced NO_x
1001 emissions also led to less titration of ozone, a reduced conversion of OH by NO₂ and an increased
1002 HO₂/OH concentration ratio. Further, even though the intensity of solar radiation is low during
1003 February, the photochemical production of ozone and OH was not suppressed. However, the strong
1004 decrease in NO resulting from reduced activities during the pandemic was not compensated by a
1005 sufficiently large increase in peroxy radicals, so that the overall ozone production by the limiting
1006 HO₂ + NO and RO₂ + NO reactions was reduced during the month of February. The ozone

1007 concentration increase was therefore due primarily to a relatively larger reduction in the ozone
1008 loss. In the NO_x-limited region of southern China, the concentration of ozone and other
1009 photooxidants decreased because their formation rate, favored by NO_x, was reduced, except in
1010 VOC-limited urban areas like Guangzhou or Hong Kong, where the model predicted ozone
1011 enhancements.

1012 In the other regions of the world during the peak of the lockdown period (corresponding to
1013 Northern Hemisphere spring and Southern Hemisphere fall), the oxidation level was also disturbed
1014 by the reduced emissions of ozone precursors. During April 2020, for example, the level of
1015 oxidants including ozone was enhanced in the regions of Europe where the background level of
1016 NO_x is relatively high. In response to the perturbed emissions of pollutants, ozone concentrations
1017 increased in a region extending from the UK to Germany, and OH levels increased in most of
1018 western Europe except in Spain. In North America, the reduced emissions led to enhanced
1019 concentrations of oxidants along the US-Canadian border and ozone concentrations increased
1020 slightly in the region of the Great Lakes. In South America, during this period of late summer and
1021 early fall, the level of photo-oxidants decreased except in metropolitan areas where elevated
1022 concentrations of OH and HO₂ were calculated by the model.

1023 The level at which the oxidizing capacity of the atmosphere changed in Northern China and to a
1024 lesser extent in Europe and North America, as well as the related increase in the concentration of
1025 secondary products such as ozone, OH, HO₂, NO₃ and PAN depends on the relative amplitude in
1026 the change in VOC and in NO_x emissions. Both forcing processes act in different directions.
1027 Therefore, if the VOC emission reduction adopted here was overestimated, the formation of the
1028 secondary species would be somewhat underestimated. In this case, a more likely description of
1029 the response of the atmosphere during the pandemic should be intermediate between the fields
1030 provided by the COVID-All and COVID-NO_x simulations.

1031 These results are obtained by model simulations that isolate the changes in surface emissions and
1032 consider them as the only forcing mechanism. However, meteorological variability provides an
1033 additional forcing mechanism that produces substantial changes in the monthly mean
1034 concentrations of chemical species; these changes can be comparable and, in some cases, larger
1035 than the chemical response to emission reductions. In China, although large-scale meteorological
1036 anomalies as derived by the model during the month of February may have contributed to the
1037 ozone increase in the North China Plain, the largest effect should be attributed to chemical

1038 perturbations related to the reduction in emissions. In most areas of Europe, however, the situation
1039 was different: during the acute period of the pandemic between 15 March and 14 April 2020, most
1040 of the ozone increase calculated by the model was associated primarily with weather anomalies
1041 rather than the emission reduction. Chemical perturbations contributed significantly to the ozone
1042 increase, but only in a limited region extending from the UK to Germany and including the
1043 Benelux countries.

1044 In summary, the simulations performed by the global atmospheric model (CESM v.2.2) with a
1045 detailed chemical scheme (MOZART TS1 mechanism) driven by emission changes of primary
1046 pollutants and forced by realistic weather conditions reproduce reasonably well the changes
1047 observed in the chemical composition of the atmosphere, and specifically in the perturbations of
1048 surface ozone and other oxidants during the COVID-19 pandemic. At least qualitatively, the
1049 response of the atmosphere to the gigantic chemical experiment that took place in the atmosphere
1050 during the first half of 2020 is found to be explained to a satisfactory degree by our current
1051 understanding of the photochemical theory, in particular in what concerns ozone formation. This
1052 unexpected global event allows us, however, to address unresolved questions related to the
1053 nonlinear atmospheric system with its complex chemical regimes including the mechanisms that
1054 control the formation of secondary pollutants under different chemical environments. More
1055 detailed and specific studies that investigate regional responses to emission reductions together
1056 with mesoscale and local weather variability should be conducted with higher resolution models.

1057

1058 **Data availability**

1059 CESM2.2.0 is a publicly released version of the Community Earth System Model and freely
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1063 [maps/data/aqereporting-8](https://www.eea.europa.eu/data-and-maps/data/aqereporting-8) (permanent link: [b21a537e763e4ad9ac8ccffe987d6f77](https://www.eea.europa.eu/data-and-maps/data/aqereporting-8)), last access:
1064 November 4, 2020. For São Paulo region, the observational dataset is provided by the CETESB
1065 Network of the environmental state agency of São Paulo, available at
1066 <https://qualar.cetesb.sp.gov.br/qualar/home.do>, last access: November 4, 2020. For the North
1067 China Plain region, the observational dataset is provided by the China Environmental Observation
1068 Network operated by the China National Environmental Monitoring Center, available at
1069 <http://www.cnemc.cn/en/>, last access: November 4, 2020. For the USA, the observational dataset
1070 is provided by the US Environmental Protection Agency, Air Quality System Data Mart [internet
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1072 This publication contains modified Copernicus Sentinel-5 TROPOMI data for 2019–2020.

1073 TROPOMI data versions 1.2.2 and 1.3.0 used here are available at <https://s5phub.copernicus.eu>.

1074

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1089

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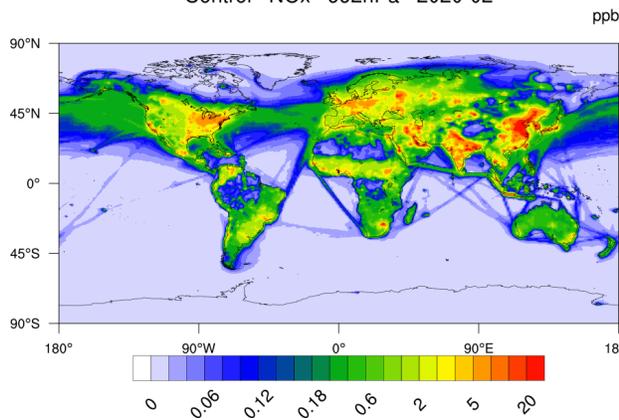
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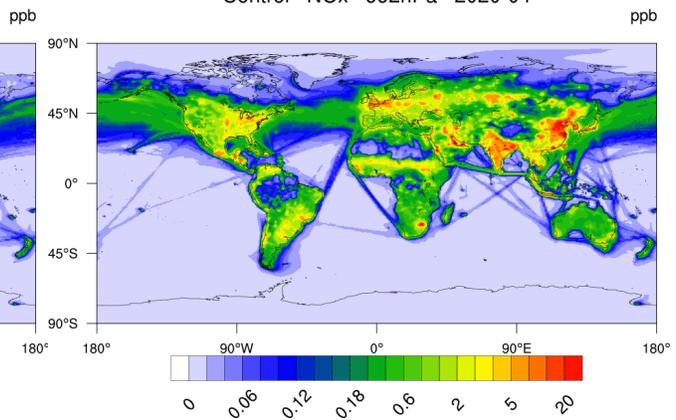
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Supplementary Information

Control NO_x 992hPa 2020-02

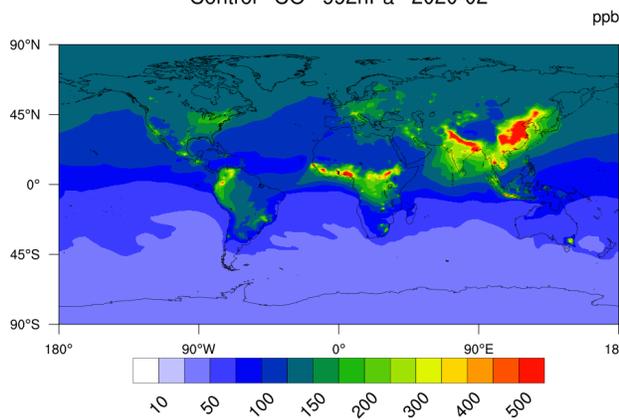


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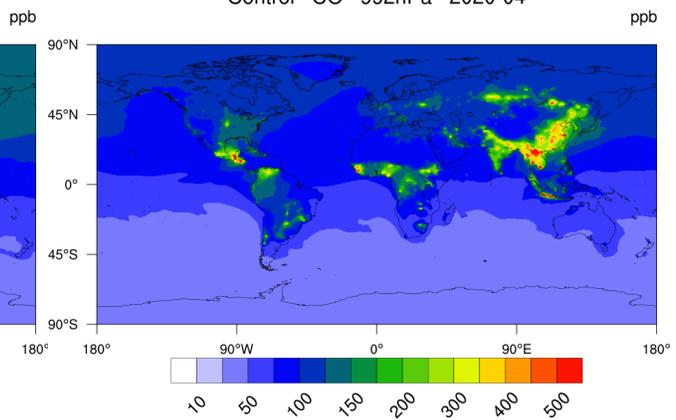


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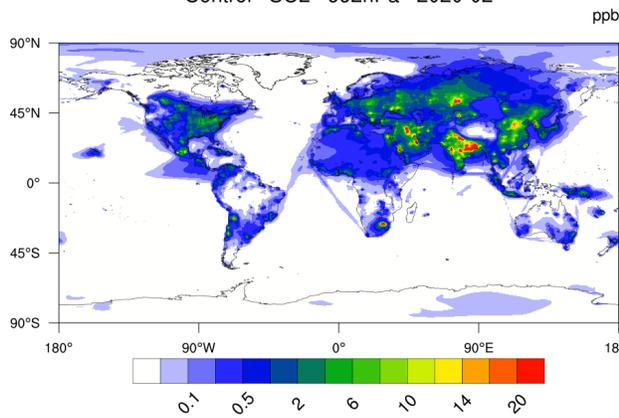


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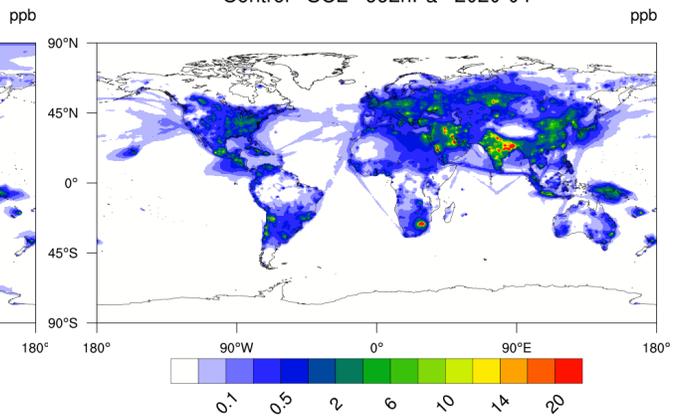


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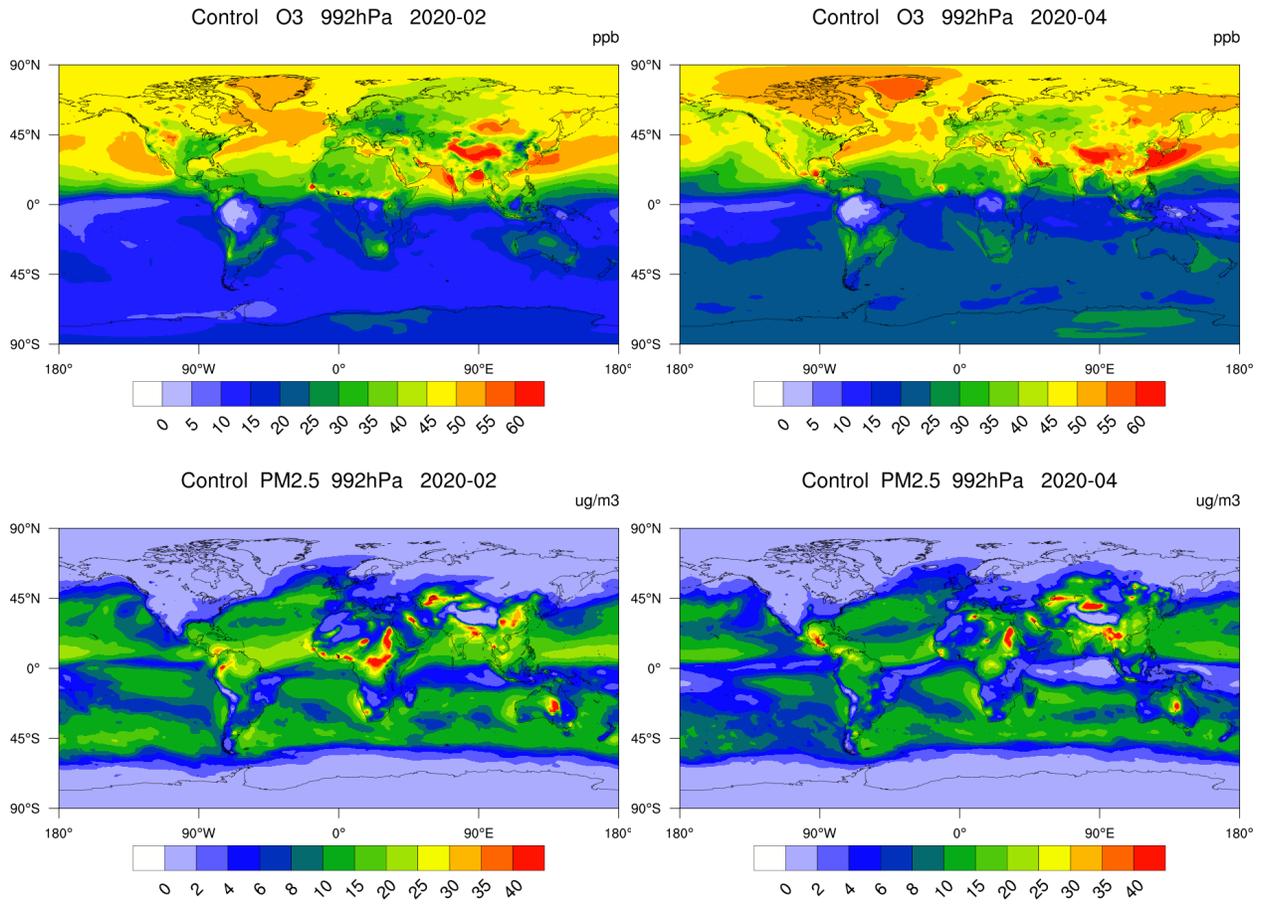
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Control SO₂ 992hPa 2020-04



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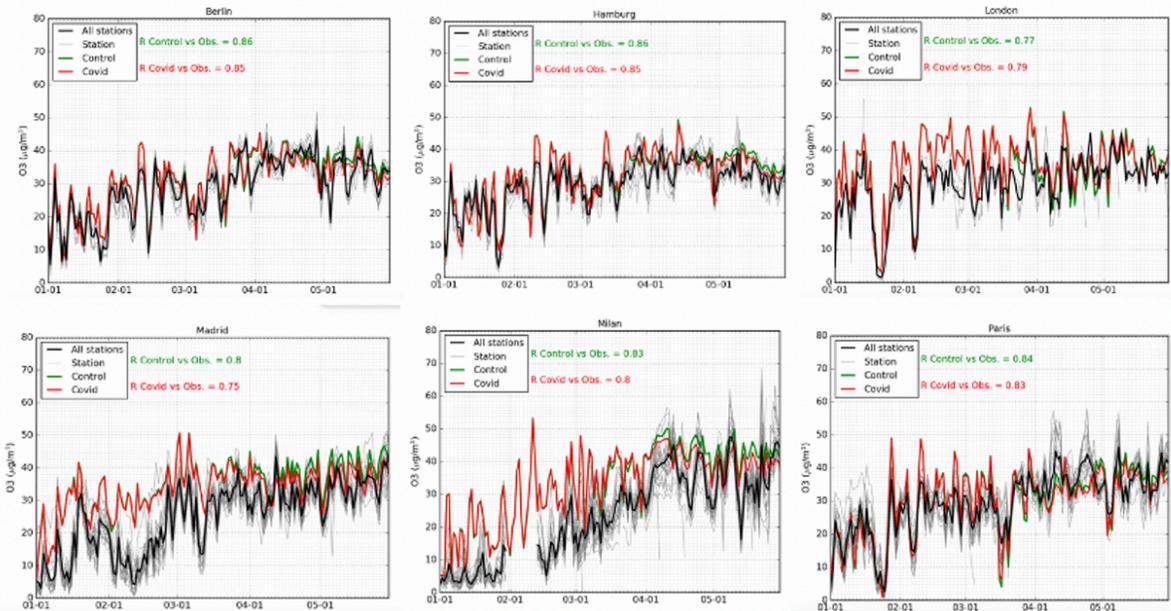


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1348 **Figure S1.** Global distribution of the surface mixing ratio of (from top to bottom) NO₂, CO, SO₂, O₃ and
 1349 PM_{2.5} (ppbv) for the month of February (left column) and April (right column) resulting from the control
 1350 simulation of the CESM model.

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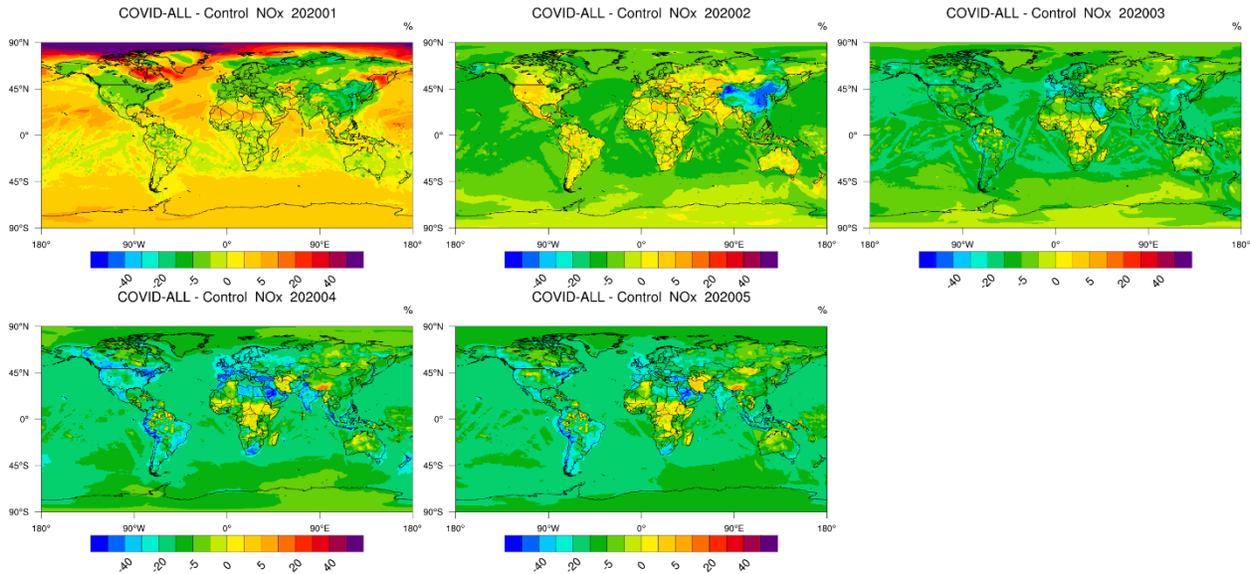
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1353 **Figure S2.** Evolution for the period 1 January – 31 May 2020 of the surface concentration of ozone in the
 1354 urban areas of Berlin, Hamburg, London, Madrid, Milan and Paris. Black curve: measurements from
 1355 monitoring stations. Green curve: model baseline case. Red curve: model case with emissions modified to
 1356 account for the effect of the COVID-19 pandemic.

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Reduction in all anthropogenic emissions (COVID-ALL)

NO_x



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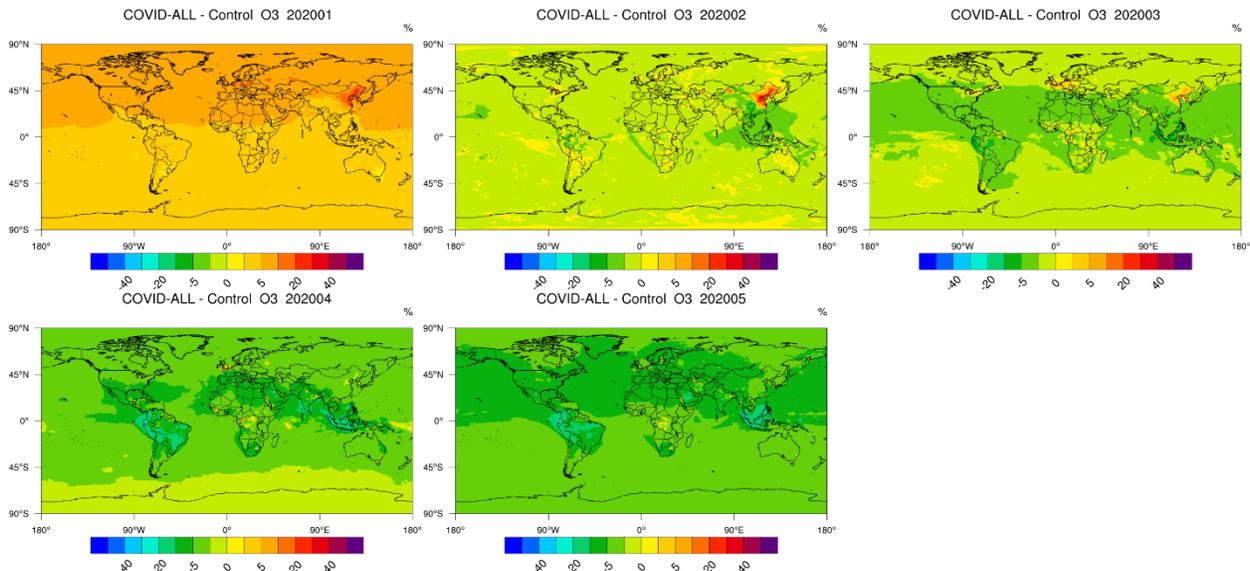
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Reduction in all anthropogenic emissions (COVID-ALL)

O₃



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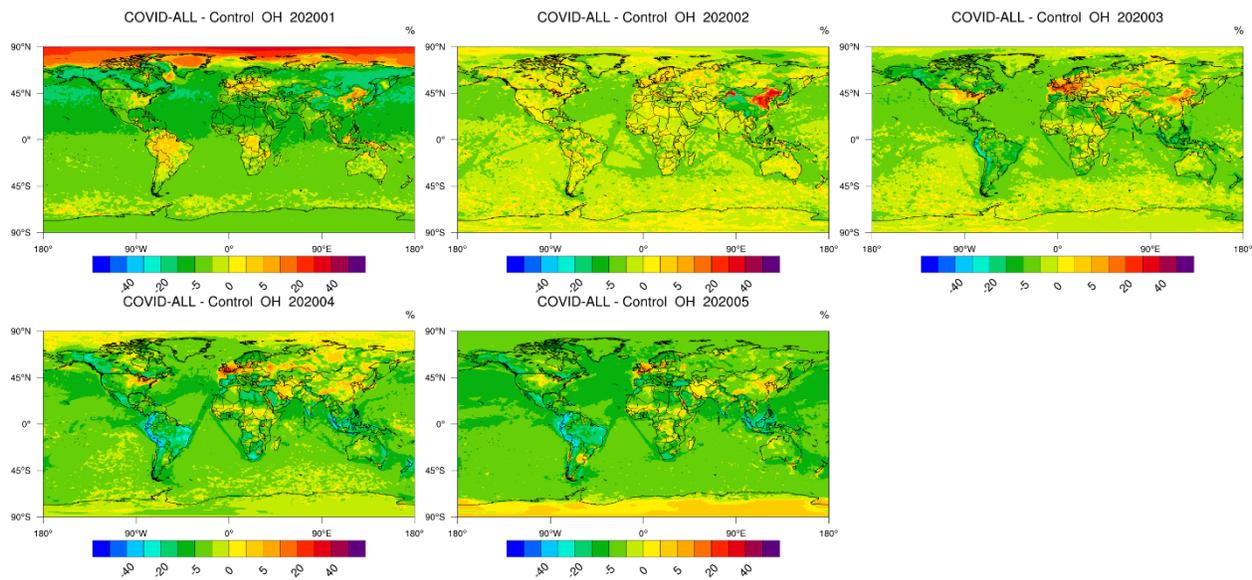
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Reduction in all anthropogenic emissions (COVID-ALL)

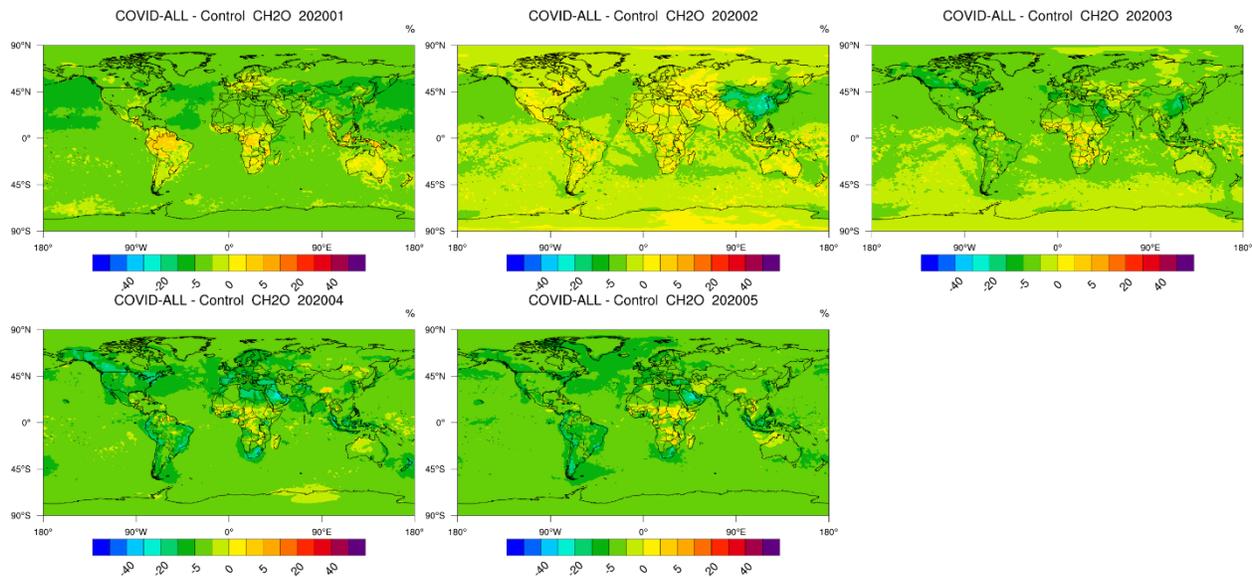
OH



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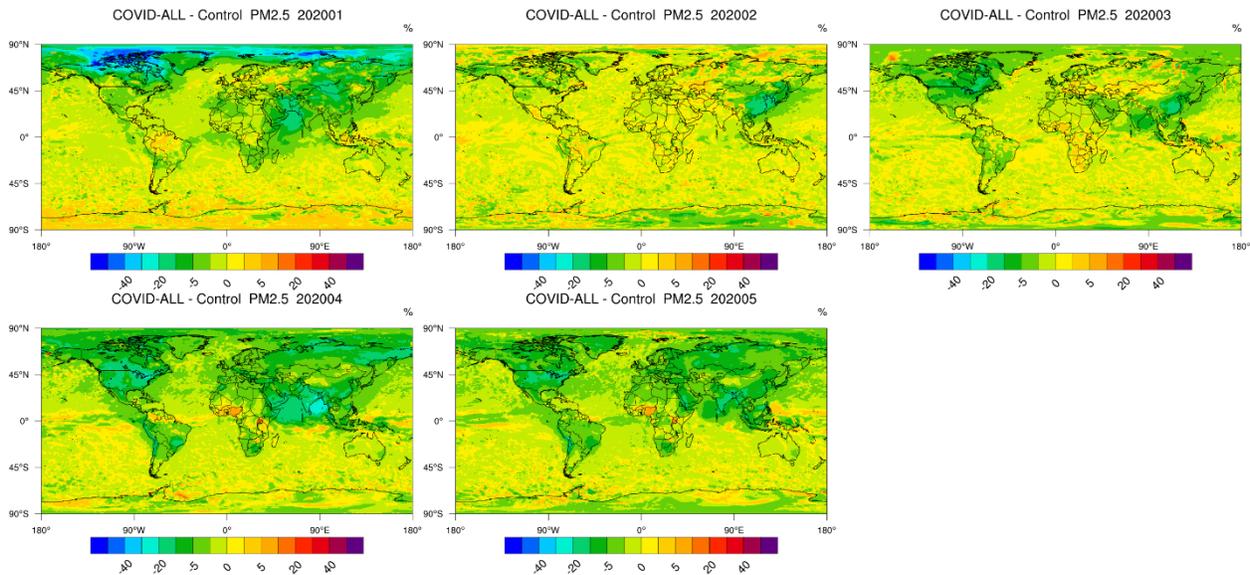
Reduction in all anthropogenic emissions (COVID-ALL)

HCHO



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1371

Reduction in all anthropogenic emissions (COVID-ALL) PM2.5



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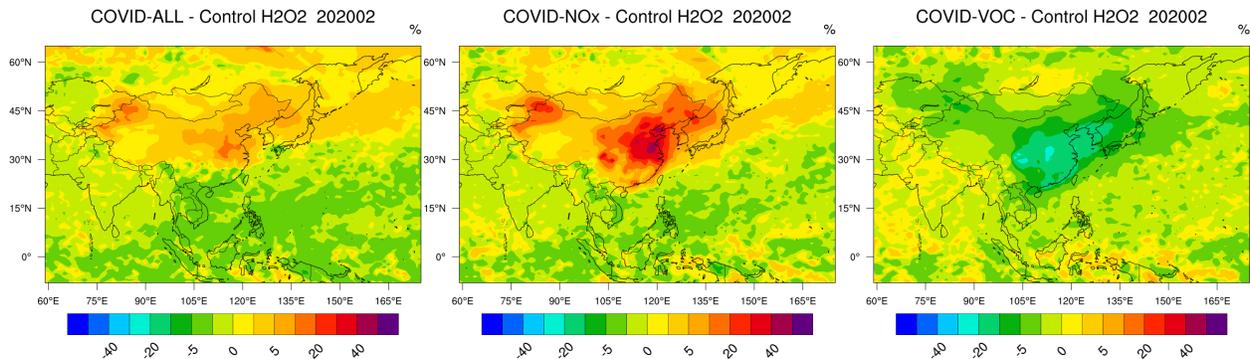
1374 **Figure S3.** Evolution of the monthly mean change (percent) in the surface mixing ratio of NO_x,
 1375 ozone, OH, HCHO, and PM_{2.5} from January to May 2020 in response to the reduction in the
 1376 anthropogenic emissions of primary pollutants resulting from the COVID-19 pandemic.

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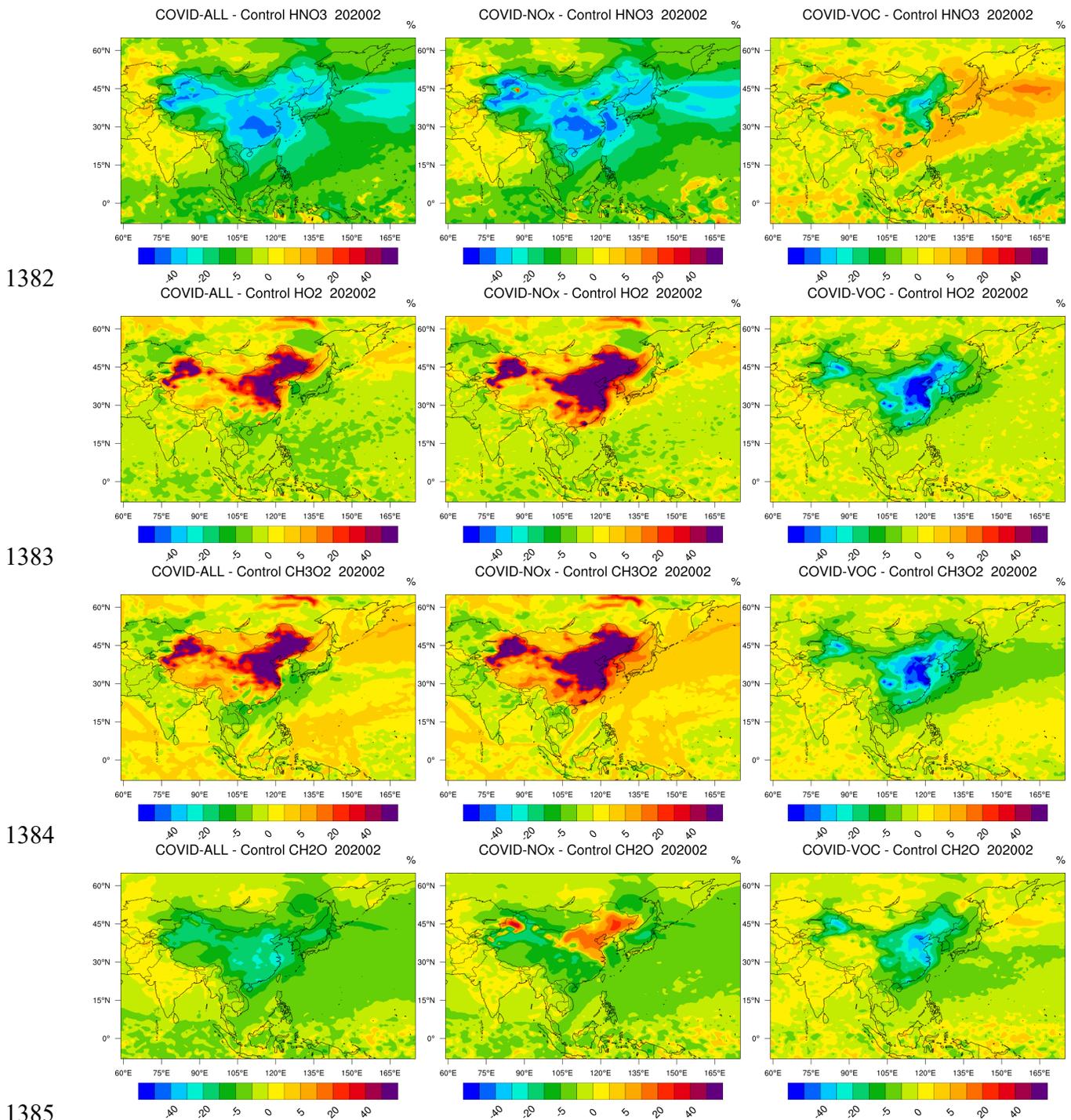
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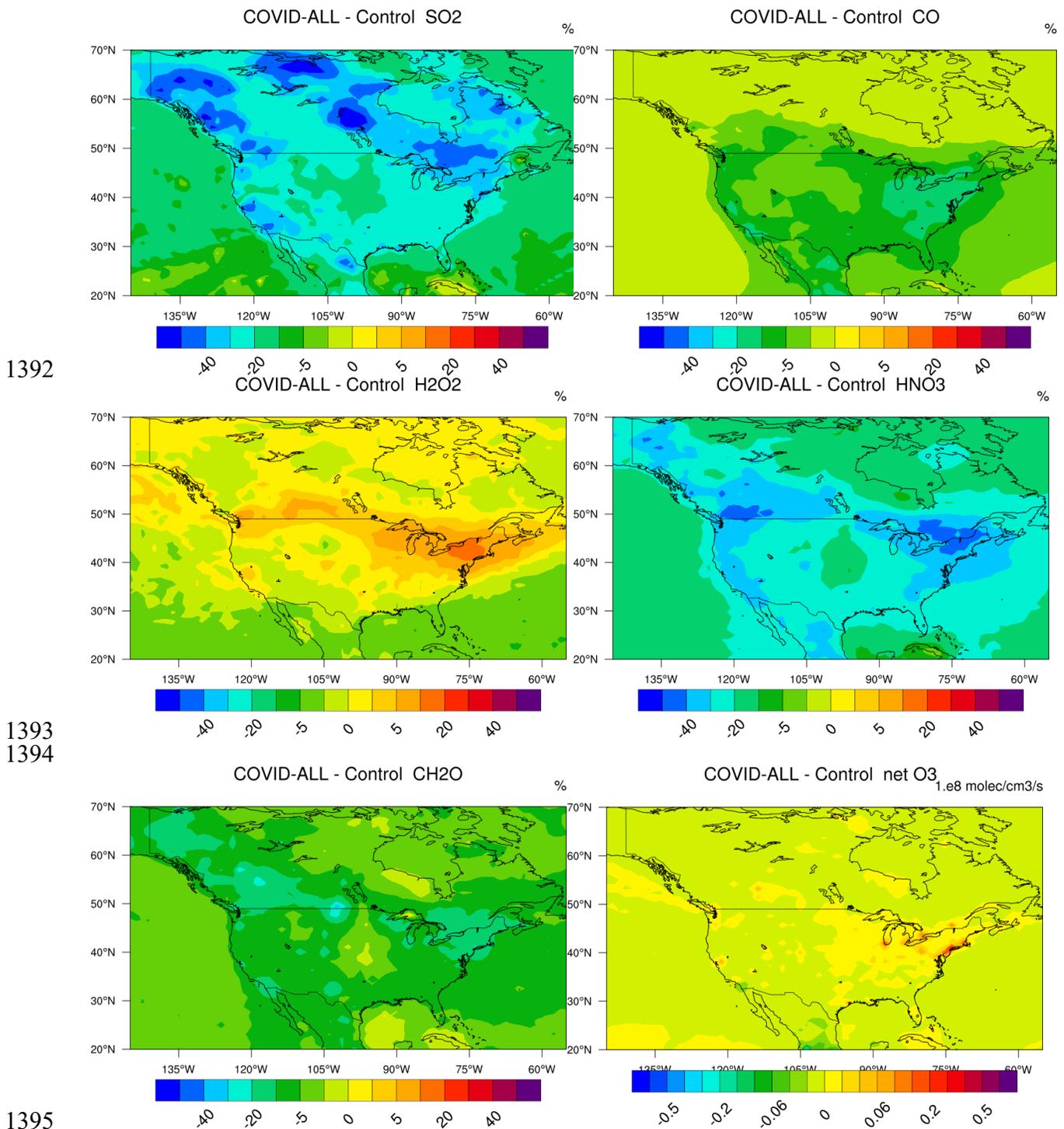
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1386 **Figure S4.** Percentage change in several chemical variables in China in response to reduced emissions of
 1387 primary pollutants in February 2020 during the COVID-19 pandemic. From the top panels to the bottom
 1388 panels: H₂O₂, HNO₃, HO₂, CH₃O₂, HCHO. Left column: reduction in all emissions; center panel: reduction
 1389 in NO_x emissions only; right panel: reduction in VOC and CO emissions only.

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1396 **Figure S5.** Change (from the top left panel to the bottom left panel) in the surface concentrations of SO₂,
 1397 CO, H₂O₂, HNO₃, HCHO [percent] and in the net ozone production rate [cm⁻³ s⁻¹] in North America in
 1398 response to reduced emissions of primary pollutants during the period 15 March - 14 April 2020 during the
 1399 COVID-19 pandemic.

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