

Societal shifts due to COVID-19 reveal large-scale complexities and feedbacks between atmospheric chemistry and climate change

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1 **The COVID-19 global pandemic and associated government lock-**
2 **downs dramatically altered human activity, providing a window into**
3 **how changes in individual behavior, enacted *en masse*, impact atmo-**
4 **spheric composition. The resulting reductions in anthropogenic ac-**
5 **tivity represent an unprecedented event that yields a glimpse into a**
6 **future where emissions to the atmosphere are reduced. While air pol-**
7 **lutants and greenhouse gases share many common anthropogenic**
8 **sources, there is a sharp difference in the response of their atmo-**
9 **spheric concentrations to COVID-19 emissions changes due in large**
10 **part to their different lifetimes. Here, we discuss two key takeaways**
11 **from modeling and observational studies. First, despite dramatic**
12 **declines in mobility and associated vehicular emissions, the atmo-**
13 **spheric growth rates of greenhouse gases were not slowed. Second,**
14 **it demonstrated empirically that the response of atmospheric compo-**
15 **sition to emissions changes is heavily modulated by factors includ-**
16 **ing carbon cycle feedbacks to CH₄ and CO₂, background pollutant**
17 **levels, the timing and location of emissions changes, and climate**
18 **feedbacks on air quality.**

COVID-19 | air quality | greenhouse gases | Earth system | mitigation

1 The effects of the COVID-19 pandemic and associated lock-
2 down measures have provided a way to observationally test
3 predictions of future atmospheric composition. This is illus-
4 trated conceptually in Figure 1. With many people working
5 from home and limiting travel, the pandemic caused a signifi-
6 cant decrease in anthropogenic emissions. These emissions
7 reductions can be thought of as a jump forward in time to
8 a future where additional systemic emissions controls have
9 been adopted. However, because these changes occurred in a
10 matter of months, the changes to the concentrations of key

air quality (AQ) and climate relevant gases in the atmosphere
were readily observable. Combining these observations with
current state-of-science models allows us an important win-
dow into the underlying processes governing the response of
the Earth system to reductions in anthropogenic emissions,
and thus a preview of the relative effectiveness of different
emissions control strategies.

Our goal is to synthesize some of the key results from
the past year into a coherent understanding of what we have
learned about the effectiveness of different strategies to reduce
greenhouse gas (GHG) emissions and improve AQ. We will do
so in four parts. First, we summarize the observed changes

Significance Statement

The COVID-19 pandemic and associated lockdowns caused significant changes to human activity that temporarily altered our imprint on the atmosphere, providing a brief glimpse of potential future changes in atmospheric composition. This event showed key differences in how air quality and atmospheric greenhouse gas concentrations respond to changes in anthropogenic emissions, with implications for future mitigation strategies.

JLL led the manuscript and the human activity analysis. JN, DS, and POW led the study team. K. Barsanti, K. Bowman, DS, AT, and EK led study subgroups. Remaining authors contributed data analysis. All authors helped revise the manuscript.

The authors declare no competing interests.

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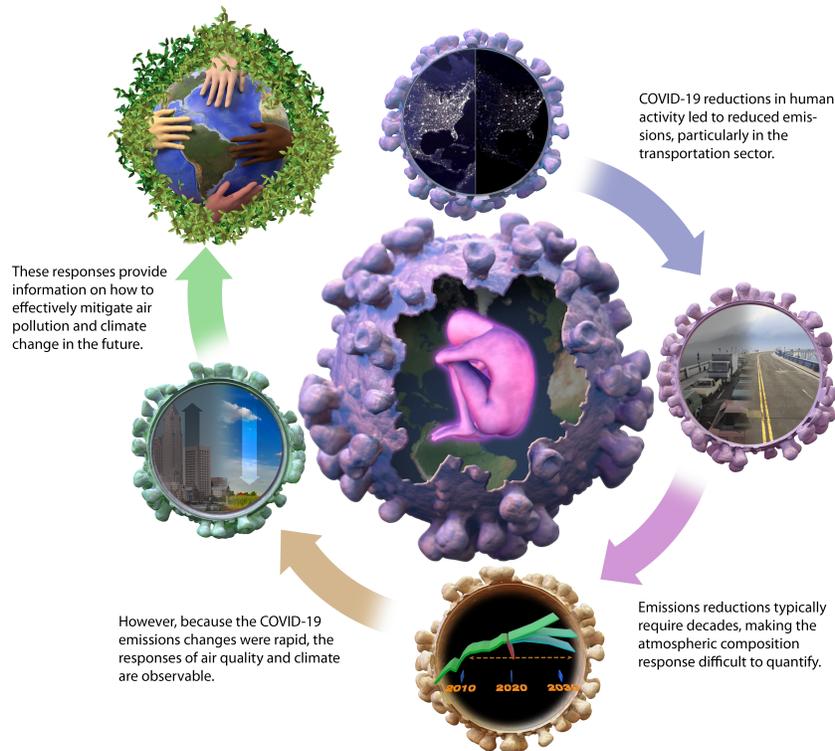


Fig. 1. Illustration of the conceptual foundation for this study. The COVID-19-induced reductions in human activity led to reduced anthropogenic emissions. The fact that these reductions occurred over months rather than decades allows us to observe how the atmosphere, land, and ocean are likely to respond in a future scenario with stricter emissions controls. This analysis helps to identify effective pathways to mitigate air pollution and climate change. Image credit: Chuck Carter / Keck Institute for Space Studies

23 in anthropogenic emissions during 2020. Second, we examine
 24 how the reduction in CO₂ emissions impacted the atmospheric
 25 CO₂ growth rate. Third, we show that the response of AQ
 26 to emissions reductions is very spatially heterogeneous, and
 27 summarize the causes of that heterogeneity. Fourth, we discuss
 28 the implications of these results for future AQ improvement
 29 strategies, our understanding of processes controlling GHG
 30 concentrations in the atmosphere, feedbacks between AQ,
 31 GHGs, and climate, and finally close by identifying strengths
 32 and gaps in our current observing networks. We draw three
 33 primary conclusions from this synthesis:

- 34 1. Despite drastic reductions in mobility and resulting vehi-
 35 cular emissions during 2020, the growth rates of GHGs
 36 in the atmosphere were not slowed.
- 37 2. The lack of clear declines in the atmospheric growth
 38 rates of CO₂ and CH₄, despite large reductions in human
 39 activity, reflect carbon cycle feedbacks in air-sea carbon
 40 exchange, large interannual variability in the land carbon
 41 sink, and the chemical lifetime of CH₄. These feedbacks
 42 foreshadow similar challenges to intentional mitigation.
- 43 3. The response of AQ to emissions changes is heavily mod-
 44 ulated by factors including background pollutant levels,
 45 the timing and location of emissions changes, and climate-
 46 related factors like heat waves and wildfires. Achieving
 47 robust improvements to AQ thus require sustained reduc-
 48 tions of both AQ and GHG emissions.

Summary of emissions in 2020

49
 50 As AQ-relevant gases and CO₂ are co-emitted by combus-
 51 tion processes, decreases in human activity are expected to
 52 drive decreases in both of these species. Figure 2 summarizes
 53 changes to key sectors of human activity during the COVID-
 54 19 pandemic. Figure 2a shows the Oxford Stringency Index
 55 (1), which quantifies the severity of government-imposed res-
 56 trictions on travel, businesses, schools, and other aspects of
 57 society. Panels b, c, and d show changes in air travel & mar-
 58 itime shipping, traffic, and United States (US) electricity use,
 59 respectively. There is a clear decrease in air travel and traffic
 60 for most of the world in March 2020, when the first major
 61 wave of COVID-19 led governments to institute quarantine
 62 measures (see also high values of the Stringency Index). Mar-
 63 itime shipping (to west coast US ports) and power generation
 64 (in the US) were less affected. Power generation in particular
 65 remained within approximately 5% of 2019 levels.

66 Reductions in NO_x emissions were apparent in both in situ
 67 (5) and satellite (6) observations of NO₂ concentrations due
 68 to the short atmospheric lifetime of NO_x (< 1 day). Esti-
 69 mates of NO_x emissions reductions from assimilating satellite
 70 data in global models (7), combining global chemical models
 71 with machine learning trained on surface measurements (8), or
 72 activity data (including electricity use, traffic/mobility data,
 73 flight data, etc.) (9–11) find regional reductions of 10% to
 74 40% during the strictest lockdown periods. Generally, meth-
 75 ods assimilating satellite data report smaller reductions (10%
 76 to 20%) than studies based on activity data (25% to 40%).

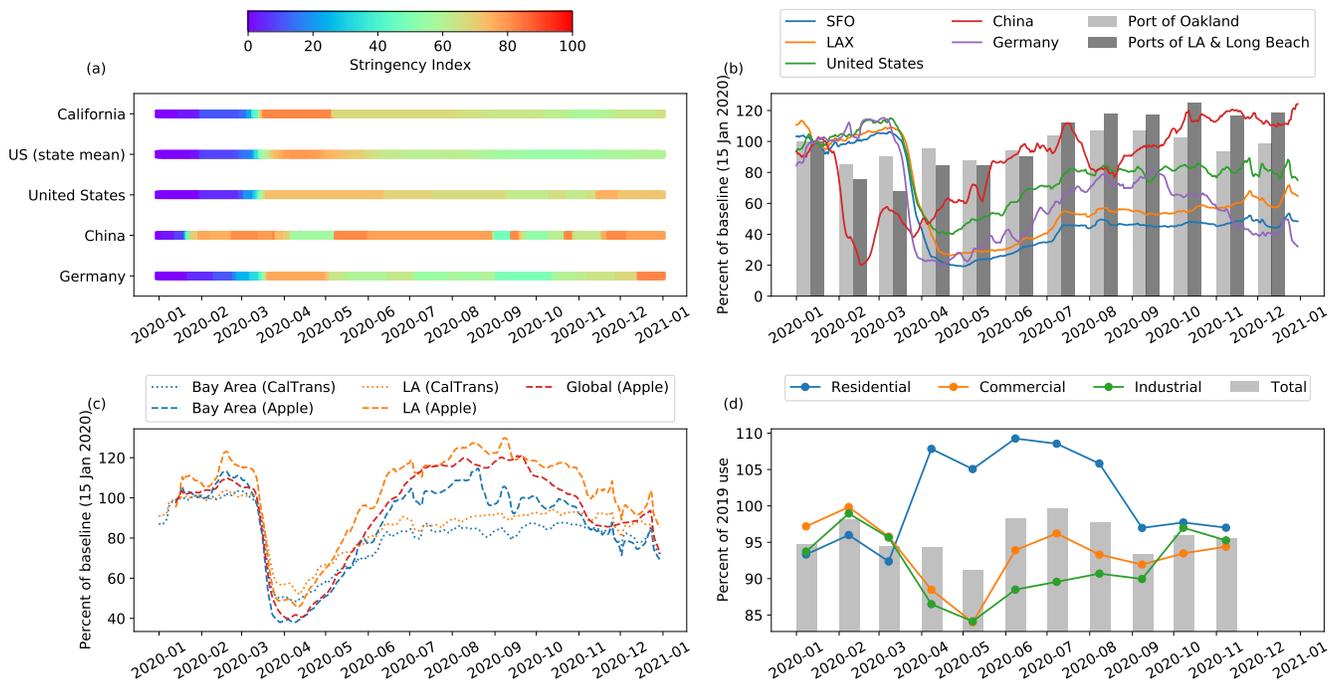


Fig. 2. Metrics for change in human activity at different scales show that the strongest impact of COVID-19 lockdowns were in the transportation sections, and that these impacts varied substantially from country to country. Panel (a) shows the Oxford stringency index (1) for the regions used in this figure. “US (state mean)” is the average of individual states’ indices, “United States” is the index attributed to the US as a whole (not individual states, see SI for discussion). Panel (b) shows the percent change in flights (2–4) for two California airports and three countries (lines) and container moves for three California ports (bars) Panel (c) shows traffic metrics for two California urban areas, and 26 countries (“global”). CalTrans indicates Caltrans PEMS data; Apple indicates Apple driving mobility data. Panel (d) shows electricity consumption in the US by sector, relative to the same month in 2019. The three sectors shown constitute > 96% of US power consumption. In (b) and (c), daily metrics are relative to 15 Jan 2020 and presented as 7 day rolling averages and monthly metrics are relative to Jan 2020. Electricity consumption not available after Nov 2020 at time of writing.

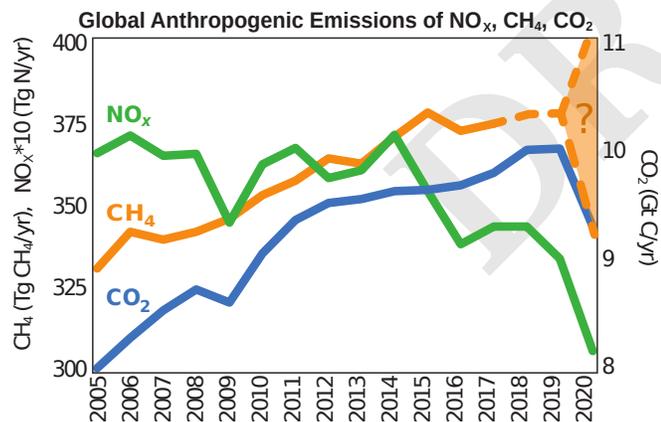


Fig. 3. 2020 saw reductions in CO₂, CH₄, and NO_x emissions. CH₄ and NO_x are plotted along the left axis, CO₂ on the right. The dashed line for CH₄ after 2017 indicates it is estimated from the average rate of increase. 2020 emissions are represented as a range: the IEA estimated a 10% decrease in CH₄ emissions in 2020(12), but this is uncertain, as the CH₄ growth rate increased in 2020. Full details are in the SI.

decrease in CO₂ over the remainder of 2020 (14). The largest decreases occurred in the first half of 2020, as shown in Fig. 4a and were primarily associated with reductions in ground transportation (15). The response of atmospheric CO₂ mixing ratios can be observed near the emissions sources; during the strictest lockdowns, Turner et al. were able to use CO₂ observations from a local ground-based network to estimate a 48% reduction in traffic CO₂ emissions in the San Francisco Bay Area (16). Liu et al. found a 63% (41 ppm) decrease of the typical on-road CO₂ enhancement in Beijing, China (17). Distinguishing these signals in CO₂ at regional scales is more challenging. Buchwitz et al. infer peak decreases in anthropogenic CO₂ emissions from China of 10% from space-based total column CO₂ measurements (18). However, they note that the uncertainty is approximately 100%, and that the expected CO₂ concentration signal is 0.1 to 0.2 ppm, out of a background of over 400 ppm.

Anthropogenic CH₄ emissions are dominated by sources such as landfills, oil and gas production, and agricultural activities. The International Energy Agency (IEA) estimates that CH₄ emissions dropped by 10% in 2020 (Fig. 3), largely due to the decrease in demand for oil and gas. However, it is unclear whether reduced demand during 2020 was the primary driver of emissions. It is likely that decreased maintenance of landfills and oil and gas infrastructure during the COVID-19 pandemic led to new leaks in some areas, which can result in those locations becoming CH₄ “superemitters” (19). In general, the type, maintenance level, and throughput of CH₄ infrastructure can have a large impact of the amount of fugitive

Estimates of the reduction in global NO_x emissions in the first half of 2020 range from 5% (8) to 13% (7).

The change in global CO₂ emissions was comparable to that of NO_x emissions, as seen in Fig. 3. Liu et al. report a peak global reduction of approximately 15% (4 Tg C or 15 Mt CO₂) in April, and an annual total of 5.4% (13). In March 2020, Le Quéré et al. projected a slightly larger 7%

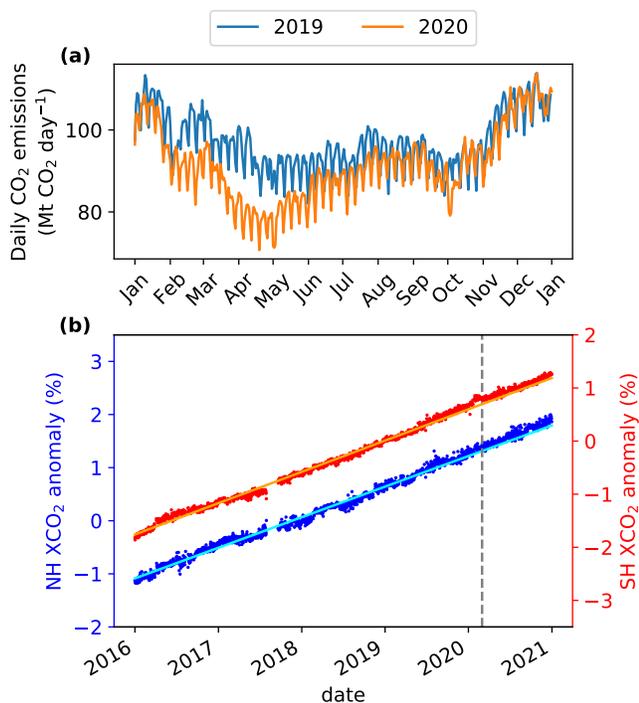


Fig. 4. Despite substantial reductions in anthropogenic CO₂ emissions in early 2020, the annual atmospheric CO₂ growth rate did not decline. Panel (a) shows daily global CO₂ emissions for 2019 and 2020, calculated following Liu et al. (13). Panel (b) shows trends in atmospheric column average CO₂ from the Orbiting Carbon Observatory 2 (OCO-2). The small blue and red symbols indicate daily, deseasonalized values as percent anomalies relative to the global 2018 mean. The solid cyan and orange lines are linear fits to 2016 through 2019 data. In panel (b) the vertical gray dashed line marks 1 March 2020 as the approximate beginning of lockdowns in response to COVID-19. A version of (b) showing the absolute trends and the data including the seasonal cycle is available as Fig. S8 in the SI.

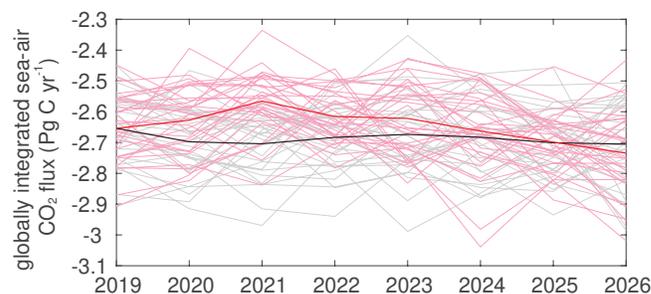


Fig. 5. Sea-air carbon exchange responded quickly to the reduction in anthropogenic CO₂ emissions during 2020. Shown here are annual mean, globally integrated sea-to-air carbon dioxide fluxes predicted from the CanESM5-COVID ensemble (24, 25). Black/gray lines derive from simulations forced with SSP2-RCP4.5 CO₂ emissions, while red/pink lines derive from simulations forced with a 25% peak CO₂ emissions reduction in 2020. See (24, 25) for more details. Thick lines are ensemble averages, and thin lines are individual ensemble members, each with different phasing of internal variability.

growth rate of 2.45 ppm/year since 2016 (Fig. S8b and 22), the 5.4% total reduction in CO₂ emissions calculated by Liu et al. (13) equals a 0.13 ppm/yr decrease in the CO₂ growth rate for 2020—well within the natural variability observed by OCO-2 (Fig. S8) and surface networks (22).

Wildfires are one element of the variability in CO₂ growth rate. The 2019/2020 Australian wildfires emitted 173 Tg C (634 Mt CO₂) between Nov 2019 and Jan 2020, over 6 times more than Australia's average Nov.-Jan. CO₂ emissions for 2001 through 2018 (23). This drove an early increase in CO₂ in 2020, evident in the deseasonalized southern hemisphere OCO-2 XCO₂ (Fig. 4b, red series) and growth rate derived from the OCO-2 data (Fig. S8b). This wildfire anomaly offset a third of the 518 Tg C (1901 Mt CO₂) reduction in anthropogenic CO₂ (13) and so does not fully explain the offset between emissions and atmospheric mixing ratios for CO₂.

The atmospheric CO₂ growth rate led to a reduction in the rate of oceanic CO₂ uptake. Figure 5 shows the magnitude of ocean carbon fluxes over 8 years as computed from a model ensemble under normal and COVID-like emissions. There is significant variation in the sea-air and CO₂ flux among the model ensemble members. This spread represents the potential interannual variability in CO₂ flux; given that variability, the true change in CO₂ flux in 2020 is uncertain, in part due to corresponding variability in the land carbon sink (Fig. S9). However, the ensemble mean indicates that while on short time scales the land carbon flux is insensitive to the change in emissions (Fig. S9), the ensemble mean ocean uptake was reduced by 70 Tg C/yr in 2020. This would offset 14% of the approximately 520 Tg C/yr (1901 Mt CO₂/yr) reduction in anthropogenic CO₂ emissions in 2020 (13), further dampening the signal from emissions reductions in atmospheric CO₂.

The growth rate of CH₄ was also not slowed by the pandemic. Figure 6a shows trends in column average CH₄ (XCH₄) from two ground based spectrometers in the Total Carbon Column Observing Network (TCCON, 26, 27) located in Park Falls, Wisconsin, US (28) and Lauder, New Zealand (29, 30). The XCH₄ values after 1 March 2020 lie approximately 0.3% above the 2016 to 2019 trend in both hemispheres. Similarly, NOAA reported the single largest increase in CH₄ in its record (31).

Because the lifetime of CH₄ depends on the abundance of

emissions (20, 21). On a positive note, some of the decrease in emissions estimated by the IEA was associated with the installation of new oil and gas infrastructure and the adoption of new CH₄ regulations in a number of countries (12). Such decreases would likely be sustained beyond the pandemic period.

CO₂ and CH₄ atmospheric growth rates

The effect of CO₂ emissions reductions, especially from ground transport, were clearly apparent in urban-scale observations of atmospheric CO₂ mixing ratios (16, 17). This does not, however, transfer to global-scale observations. Figure 4b shows deseasonalized trends in column-average CO₂ mixing ratios (referred to as XCO₂) observed by the Orbiting Carbon Observatory 2 (OCO-2) instrument. Despite the reduction in CO₂ emissions in 2020 (Fig. 4a), there is no clear deflection of the observed XCO₂ below what would be projected based on previous years' growth rates. We compared the variability in actual atmospheric CO₂ growth rates derived from the OCO-2 data with that computed from fossil fuel emissions (Fig. S8b) and found that the change in atmospheric CO₂ growth caused by the COVID-19 pandemic is smaller than the natural year-to-year variability. This is expected, because the percent change in the CO₂ growth rate, in the absence of feedbacks, will match the percent change in emissions. For a typical

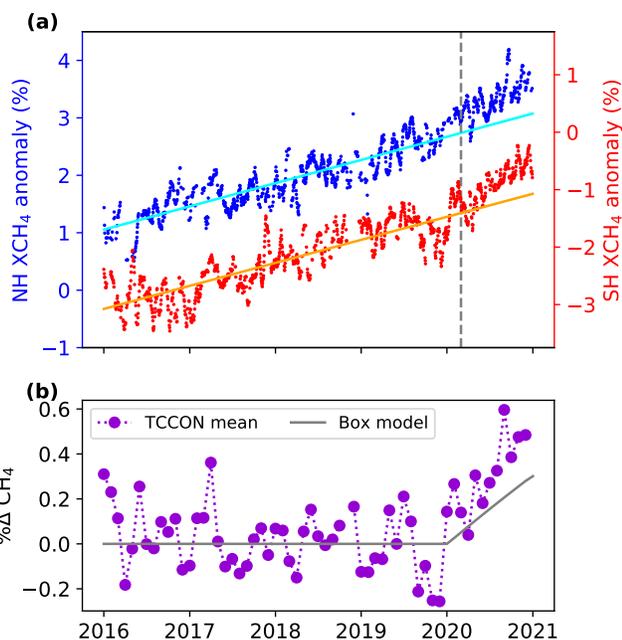


Fig. 6. Atmospheric mixing ratios of CH₄ increased more rapidly in 2020 than they had in the past decade. The increase is consistent with no change in CH₄ emissions and a 3% decrease in OH (predicted from decreased NO_x emissions) during 2020. Panel (a) is similar to Fig. 4b, except it shows trends in column-average CH₄ (XCH₄) from two TCCON sites: Park Falls, WI, USA in the northern hemisphere and Lauder, New Zealand in the southern hemisphere instead of OCO-2 XCO₂. Panel (b) compares the TCCON XCH₄ trend to that predicted by a box model. The purple series are the monthly mean percent differences between the TCCON XCH₄ and linear fits from (a). The grey line represents the percent difference in CH₄ predicted by a box model (33) with a 3% decrease in OH during 2020 compared to no change in 2020 OH.

the hydroxyl radical (OH), the concentration of CH₄ varies with atmospheric pollution levels. In fact, we find compelling evidence that the jump in CH₄ mixing ratios during 2020 is partly due to reductions in NO_x emissions. In a model incorporating the decreased NO_x emissions associated with COVID-19 (32), the resulting decrease in global ozone (7) leads to a 2% to 4% decrease in global OH concentrations. As oxidation by OH is the primary loss process for atmospheric CH₄, this acts to increase CH₄ mixing ratios in the atmosphere. Figure. 6b compares the trend in XCH₄ measured by TCCON to that predicted by a box model (33). The purple series is the monthly percent difference of TCCON XCH₄ from the linear trends shown in Fig. 6a, and the grey line is the percent difference between a box model run with and without a 3% decrease in OH during 2020. The box model closely matches the extra growth in atmospheric CH₄ during 2020, indicating that the change in OH was an important driver of the observed CH₄ growth. However, this is inconsistent with the 10% decrease estimated by the IEA (12), as our box model assumes constant CH₄ emissions after 2012.

If decreases in anthropogenic NO_x emissions during 2020 were responsible for the increase in CH₄ lifetime that led to its higher than expected growth rate, what does this imply for the effect of future efforts to reduce NO_x emissions to improve AQ? To understand this, we need to examine how the 2020 NO_x decreases affected AQ around the world. In the next section, we will describe the ozone and particulate matter (PM) response to these NO_x reductions. Afterward, we will

explore the implications of this AQ-GHG in the discussion.

Heterogeneity in air quality response

Most parts of the world saw significant decreases in NO_x emissions during the pandemic, but the magnitude and timing of these emissions changes varied with location. Figures 7a-c compare timeseries of NO₂ column densities measured by TROPOMI for three cities. Following the beginning of lockdown measures (indicated by the dotted lines), the 2020 NO₂ column densities are clearly less than in 2019. However, in Los Angeles, the drop in NO₂ occurred very rapidly when lockdowns were enacted in early March, but by May there was little difference between 2019 and 2020. In Lima, on the other hand, the difference between 2019 and 2020 grew from March through May. In Shanghai, we see a very large drop in NO₂ associated with the early lockdown in January and a smaller drop during the second lockdown in late February.

These changes in NO_x emissions drove changes in secondary pollutants, such as ozone and PM. However, the ozone and PM responses depended on the local chemical regime and meteorology, as well as the magnitude and timing of the NO_x emissions reductions. In this section, we describe the factors controlling the ozone response first, followed by PM.

Ozone. Ozone is a secondary pollutant produced in the atmosphere from the reaction of NO_x and OH with volatile organic compounds (VOCs). The response of ozone concentrations to changes in NO_x emissions is characterized by the ozone production efficiency (OPE), which is the ratio of the change in ozone for a given change in NO_x.

Figures 7d-f show the ozone production efficiency (OPE) calculated in a global model that assimilates multiple satellite measurements. The OPE values shown represent the change in ozone mass burden per unit change in mass of reactive nitrogen emissions, using the COVID-19 reduction in emissions as the ΔNO_x. More detail is given in the SI.

Two patterns in the OPEs demonstrate the significant spatial and temporal variability in the relationship between NO_x emissions and ozone concentrations. First, in Fig. 7f, the OPE in the Northern hemisphere increases between February and June. This is mostly due to increasing sunlight driving key photolysis reactions more rapidly. Thus, the timing of NO_x emissions changes plays a significant role in the magnitude of the ozone response in the mid- and high-latitudes, with a smaller ozone response to a given NO_x change during spring than during summer. Second, in Fig. 7d, tropical and subtropical cities have the largest, most positive OPEs. Furthermore, there is little change in OPE with season for these cities (Fig. 7e) due to the relatively small changes in insolation at low latitudes. Figure 7d indicates that most of the northern mid-latitude cities have small, positive OPEs. Two cities, however, have slightly negative OPEs (Beijing -0.10, Karachi -0.06); a negative OPE indicates that ozone increased when NO_x emissions decreased. Other studies have, in fact, identified large ozone increases in China (34) associated with the decreased NO_x emissions during the pandemic. Additional increases in ozone were observed in Europe (35), with smaller but still positive changes in ozone in the United Kingdom (36).

We use a steady-state model (Fig. S10) to interpret the patterns in Fig. 7. From the steady-state model, we know OPE is small at both low and high NO_x concentrations, but large at

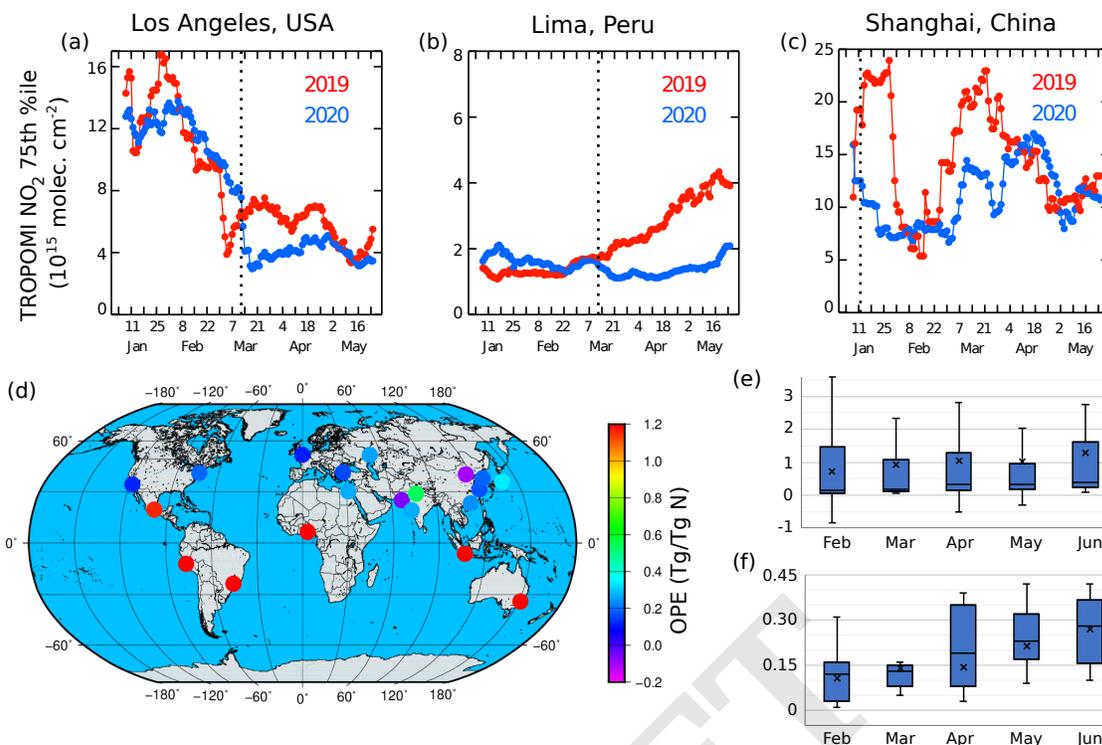


Fig. 7. COVID-19 lockdowns dramatically reduced urban NO₂ levels, which in turn drove changes in O₃ production. Panels (a–c) show 15 day rolling averages of 75th percentile TROPOMI NO₂ column densities in three cities for 2019 and 2020. The vertical dotted line indicates the beginning of lockdown measures in 2020. Panel (d) shows OPE modeled in 17 megacities, averaged from February to June 2020. Panel (e) shows modeled monthly global averaged tropospheric O₃ production efficiency (OPE). The whiskers are the minimum and maximum, the horizontal lines the quartiles and median, and the X is the mean. Panel (f) is similar to (e), but averaged over 30° N to 90° N.

intermediate NO_x concentrations. Overall OPE also increases with VOC reactivity (VOC_R, the total rate of reaction of all VOCs with OH in a given parcel of air) for NO_x concentrations greater than ~ 0.1 ppb. Thus, in Fig. 7, areas with negative OPE are in the high-NO_x part of the OPE curve; sustained efforts to reduce NO_x emissions will bring them closer to the maximum-OPE tipping point, after which NO_x reductions should lead to ozone reductions. Cities in the tropics and subtropics have large, positive OPE values. This is partly due to plentiful sunlight to drive photochemistry, but these regions also have large VOC_R values due to the abundance of biogenic VOCs (37). The steep dependence of OPE on NO_x follows because NO_x is the limiting reactant in ozone production in these high-VOC_R conditions. Thus, these cities should see large ozone reductions from NO_x reductions. However, of the equatorial cities shown in Figure 6, only those located in South Asia had large enough reductions in NO_x emissions during the COVID-19 pandemic to produce substantial reductions in surface ozone (3–5 ppb) (7).

We also see this heterogeneity in ozone response to NO_x emissions reductions at the intraurban scale. Measurements of daily maximum NO₂ and ozone at monitoring sites throughout the Los Angeles Basin show consistent reductions in NO₂ throughout the basin in March and April of 2020, but smaller reductions in ozone in the central northern part of the basin than elsewhere (Figs. S1, S2). This is consistent with the near-0 OPE for Los Angeles in Fig. 7d, i.e. for a city on the verge of reducing NO_x emissions to the point where NO_x is the limiting factor in ozone production. While the overall basin chemistry is at this tipping point, local differences in emissions

as well as transport of pollutants within the basin can lead to these small scale differences in ozone response (38).

However, the behavior of ozone in the Los Angeles Basin also illustrates that NO_x controls may become less effective in a warmer climate. Figure 8 shows time series of daily maximum NO₂ and ozone (top and middle panels). NO₂ and ozone concentrations are clearly lower in March and April 2020 compared to the 2015 to 2019 average, in part due to the reduction in NO_x emissions at the beginning of the lockdown. However, these two months were significantly cooler than the 2015 to 2019 average as well. When temperatures rose above average during an unusual heat wave in late April and May of 2020, ozone daily maxima rose above the range seen in 2015 to 2019, despite the fact that NO₂ remained similar to 2015 to 2019 concentrations. An increase in ozone during April and May was also seen in a previous study (39). The response of ozone per degree increase in temperature is shown in Fig. S3. Typical values for the O₃ season (May–Sep) in 2020 throughout the basin were 1.8 to 5.8 ppb K⁻¹. This is higher than a previous prediction of about 1 ppb K⁻¹ in the basin (40), suggesting the ozone climate penalty may be stronger than expected; however, analysis is ongoing.

Particulate matter. Achieving long-term reductions in PM (especially PM 2.5, particles with a diameter < 2.5 μm) concentrations is a matter of great importance due to the large health impacts of PM compared to ozone (41). Our interest here is to use observations from the pandemic period to better understand some of the factors controlling atmospheric PM concentrations, rather than focusing on the question of whether

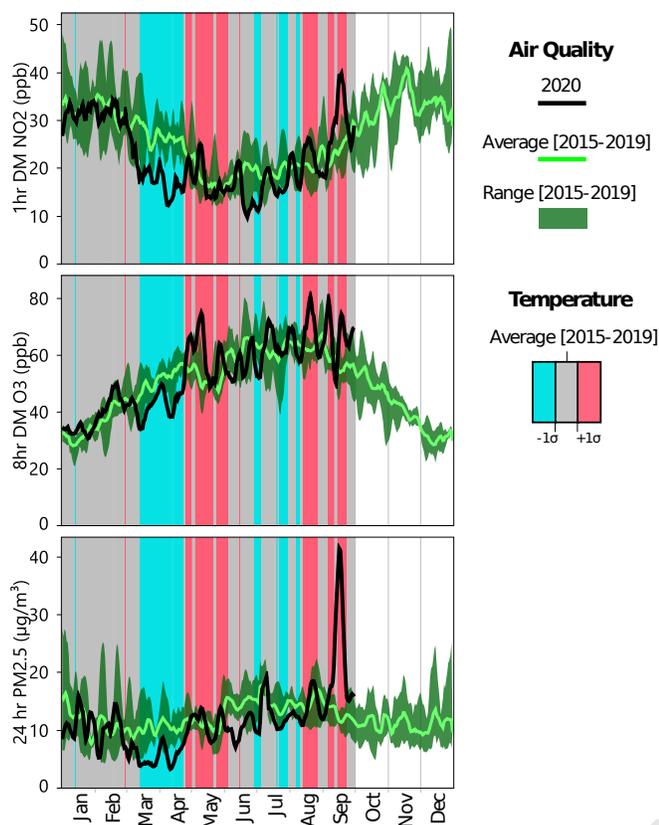


Fig. 8. In Los Angeles, temperature and wildfires drove ozone and PM pollution, respectively, more than changes in traffic. The three panels show 7-day rolling average of 24hr PM_{2.5}, 1hr daily maximum (DM) NO₂, and 8hr DM O₃, respectively, by day of year in 2020 and in the past five years (2015-2019) in the LA Basin. Bars in the background show the 7-day rolling average of basin-average 1 hr DM temperature in 2020 relative to the 2015 to 2019 average ($\pm 1\sigma$) by day of year. 2020 data are preliminary, unvalidated, and subject to change.

PM production throughout North America, Europe, and East Asia.

The relative availability of NO_x and ammonia elsewhere in the US plays an important role in whether NO_x emissions reductions lead to reduced nitrate PM. Simulations of nitrate chemistry over the continental US show that Los Angeles is somewhat unique as an urban area that experienced a significant shift to NO_x-limited nitrate chemistry. Other urban areas in the northeast, southeast, and northwest largely remained ammonia-limited (Figs. S5–S7). This could explain, at least in part, the scattered response of PM to NO_x emissions reductions across US cities seen in other studies (44). It also implies that continuing the long-running trajectory of NO_x emissions reductions in Los Angeles in order to reach the tipping point where ozone becomes NO_x limited will also benefit AQ via reduced production of nitrate PM.

However, Los Angeles also represents a cautionary tale about attributing AQ changes to the COVID-19 pandemic without accounting for other confounding factors. Weather and wildfires also played a large role in determining the PM concentrations in Los Angeles during 2020. When the lockdowns were first instituted in March, news outlets and social media attributed the clean air in the Los Angeles Basin to the lack of traffic. However, as seen in Fig. 8, the lower PM concentrations in March and April 2020 than 2015 to 2019 (Fig. 8, bottom) coincide with anomalously cool weather, which was accompanied by higher than average precipitation (Fig. S1 in (38)). Precipitation removes PM from the atmosphere through wet deposition (45, 46), and was at least partially responsible for the clean air during this period. The extreme spike in PM concentrations seen in September 2020, on the other hand, coincides with a time period when major wildfires were burning in close proximity to Los Angeles. Like the April-May heatwave, this event also points to the fact that climate change can erase progress in AQ improvement through emissions reductions.

Discussion

The changes in atmospheric composition throughout 2020 unequivocally demonstrate that AQ and GHGs cannot be treated as separate problems, despite the disparate time scales of AQ and GHG responses to changes in human activity. AQ is most dependent on local changes in emissions because of the shorter atmospheric lifetime and rapid chemistry of AQ-relevant pollutants. In contrast, the global total GHG emissions matter more than local emissions, as it is the overall GHG atmospheric growth rate that drives climate change. As discussed above, improvements in AQ made by reducing pollutant emissions locally can be offset by changes in meteorology or non-anthropogenic (e.g. biogenic or wildfire) emissions driven by climate change. Likewise, changes in AQ can affect climate change, as decreases in AQ-relevant emissions could lead to increased lifetimes for shorter-lived GHGs (such as CH₄), increasing their global warming potential.

Reductions in NO_x emissions during the pandemic did show the potential benefits cities can gain by promoting systemic change to accomplish these same reductions. For most countries, the pandemic-induced emissions reductions can be seen as going back in time to a period when NO_x emissions were lower. In the US, Europe, and China, where NO_x emissions have been trending downward, these reductions were more

PM exposure increases the chance of death from COVID-19.

The factors controlling PM concentration are more complicated than those for ozone. PM arises from primary emissions and natural sources, as well as secondary chemistry in the atmosphere. One such secondary pathway is the formation of nitrate PM from the reaction of higher oxides of nitrogen (such as HNO₃) with ammonia (42). Nitrate PM formation via this pathway may be limited by either available NO_x or ammonia.

Model simulations (Fig. S4) demonstrate the effect that NO_x emissions reductions had on nitrate PM formation in Los Angeles. Under COVID-19 emissions, the nitrate PM concentrations decreased by approximately 60% in April 2020. At the same time, the model reported a shift towards NO_x-limited (rather than ammonia-limited) chemistry. This implies that the NO_x emissions decreases in April, when the shift in the chemical regime shows the largest change, were more efficient at reducing nitrate than the reductions in other months. Compared to the measured total PM reductions shown in the bottom panel of Fig. 8, our results suggest that NO_x emissions reductions account for about 10% of the total PM reduction in the Los Angeles Basin during the COVID-19 lockdowns. This agrees with other recent work (43) which indicate that traffic NO_x emissions contribute less than 10% of secondary

COVID-19 Equivalent NO_x Emissions Year by Country

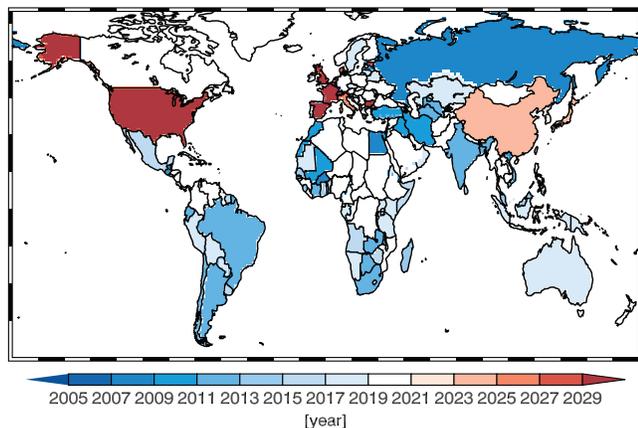


Fig. 9. The emissions reductions during the pandemic are, in a sense, like moving forward or back in time. Countries are colored by the year to which their 2020 NO_x emissions are equivalent, projected forward in time where emissions have been decreasing and backward elsewhere. Details of emissions estimates given in the SI.

akin to a jump forward in time to a lower emissions future. Figure 9 shows the equivalent year for each country's NO_x emissions during the pandemic, assuming recent trends in NO_x emissions hold constant. Most striking is how much more quickly China could reach pandemic-like emissions levels than the US or Europe. Though all three regions' emissions reductions had similar peak magnitudes (18% to 20%), Europe and especially the US are further along their respective NO_x reduction pathways than China. This, combined with China's higher pre-pandemic emissions levels, means that China can make progress quickly if they are able to maintain the aggressive pace of emissions reductions they have set over the past decade (32).

Many cities in the US and Europe are close to reaching a point at which NO_x emissions will be a very effective control on ozone concentrations. In Fig. 7d, cities with an OPE near 0 are likely at the tipping point between VOC-limited and NO_x-limited chemistry. Further NO_x reductions should move them firmly into NO_x-limited chemistry, where NO_x is the primary control on ozone formation. While sustaining these emissions reductions may be challenging due to the decreasing contribution of on-road gasoline emissions (47) and the impact of emissions reductions being offset in part by increases in chemical lifetime (48), the rewards in doing so are likely substantial. In addition, since NO_x and CO₂ are co-emitted by combustion processes, regulations such as those that encourage a transition to electric vehicles will also benefit climate. In fact, recent work has shown that the cost savings associated with reduced health impacts from air pollution will outweigh the cost of transition to a clean carbon economy and that the increased radiative forcing from longer-lived CH₄ and ozone is balanced by the decrease in forcing from smaller CO₂ mixing ratios (49). On the other hand, measures such as NO_x removal from coal-fired power plants will benefit AQ but not climate; as discussed below, this will eventually limit their effectiveness for improving AQ.

The same strategies to improve AQ will not be equally effective in all locations. On one hand, the tropical and subtropical cities with large, positive OPE values in Fig. 7d can

immediately realize substantial ozone reductions through reductions in NO_x emissions. On the other hand, cities such as Beijing and Karachi with negative OPEs, or locations such as the United Kingdom where in situ studies found a negative correlation between NO_x emissions and ozone concentrations (36) would do better to reduce volatile organic compound (VOC) reactivity simultaneously with NO_x emissions. Such an approach would allow them to avoid the chemical regimes with the largest OPEs (50) (Fig. S10a). Similarly, while chemical formation of ammonium nitrate PM in Los Angeles became NO_x-limited during the pandemic, most other cities in the US remain ammonia-limited and would see stronger reductions in PM by controlling primary emissions, organic precursors, or other key species.

Unfortunately, 2020 has also shown that improvements in AQ are likely to be offset by climate feedbacks. Such effects were most apparent in Los Angeles, where warmer than average May temperatures led to ozone concentrations above the 2015 to 2019 average, greater than average precipitation in March and April likely contributed to the reduction in PM, and severe wildfires from late August through September caused PM concentrations four times that of the 2015 to 2019 average. Changing climate will affect each of these variables, leading to warmer temperatures, more wildfires (51), and potentially more intense but less frequent precipitation (52), giving PM more time to accumulate between wet deposition events.

Changes in AQ-relevant emissions, particularly NO_x emissions, have potential to feed back into climate change as well. As we showed in Fig. 6, there is compelling evidence that reductions in OH stemming from reduced anthropogenic NO_x emissions drove a ~ 0.3% jump in CH₄ during 2020. While tropical cities have the greatest potential for decreasing ozone by reducing NO_x emissions (Fig. 7d), they also have an outsized impact on atmospheric CH₄ lifetime, as the largest share of CH₄ oxidation occurs in the tropics (33). Since only tropical cities in South Asia had substantial changes in NO_x emissions during 2020 (7), 2020 represents a minimum benchmark for the effect of NO_x reductions on the CH₄ growth rate. It is therefore essential to invest strategies to reduce fugitive CH₄ emissions (such as updated CH₄ storage and transportation infrastructure to prevent and limit leaks, landfill CH₄ capture, and confined animal feed operation CH₄ mitigation) ahead of decreases in tropical NO_x emissions.

In terms of climate, despite a reduction in global emissions equivalent to going back in time nine years (to 2011-equivalent CO₂ emissions), any change to the global CO₂ growth rate was smaller than typical interannual variability. As mentioned earlier and discussed in more detail below, this is partly due to the offsetting reduction in ocean carbon uptake (Fig. 5), but also arises because the sharp decreases in CO₂ emissions during the first half of 2020 were not sustained. By the second half of 2020, emissions due to power generation, industry, and residential consumption had nearly returned to 2019 levels (13). If we assume that these emissions levels represent a balance between reduced activity to limit the spread of COVID-19 and sufficient activity to maintain a minimum economic productivity, this suggests that reducing activity in these sectors is not practical. Reducing these sectors' emissions permanently will require their transition to low carbon emitting technologies.

One interesting aspect of the GHG emissions reductions during the pandemic was that they provided a chance to study

509 the feedback in ocean carbon uptake. The model simulations
510 using COVID-like CO₂ emissions shown in Fig. 5 indicate
511 that the sea-air carbon flux adjusts rapidly in response to
512 changes in anthropogenic emissions. That model ensemble
513 mean indicates a response time of about one year. Though
514 this basic response - a decline of the ocean carbon sink in
515 response to mitigation - is accounted for the RCP scenarios
516 (53), much uncertainty remains as to the accuracy of these
517 ocean sink predictions. This uncertainty is due both to the
518 forced response of the ocean and to interannual variability
519 Lovenduski et al. found that, for a change in ocean carbon
520 uptake to be observable with our current network of ocean
521 buoy measurements, it would need to be four times larger
522 than the COVID-19 emissions reductions (25). This will be a
523 challenge as we work to quantify the effect of future permanent
524 CO₂ emissions reductions on atmospheric CO₂ mixing ratios.

525 The pandemic does offer insight into how the atmospheric
526 GHG growth rates could be curtailed: systemic changes are
527 required to enable sustained reductions in emissions. The
528 efficacy of sustained reductions (without systemic changes to
529 the energy sector) can be seen in the contrast between CO₂
530 emissions from ground transport and international shipping
531 and aviation (“international bunkers”) reported by Liu et al.
532 (13) The peak reduction in international bunkers’ emissions was
533 only approximately 1/3rd that of the reduction in emissions
534 from ground transport, by mass. However, while ground
535 transport recovered fairly quickly, the international bunkers’
536 emissions remained at about half of 2019 levels throughout
537 the second half of 2020. As a result, the cumulative reduction
538 in 2020 emission due to international bunkers was 75% that of
539 the reduction due to traffic, despite the comparatively small
540 magnitude of the daily emissions from international bunkers.

541 Sustained reduction in other sectors will require investment
542 in renewable energy and new technologies to support current
543 levels of productivity with lower carbon emissions, that is, to
544 reduce the carbon intensity of our economy. Such investment
545 is essential, as several studies (54, 55) have documented the
546 harm to employment, family connections, and other critical
547 human connections from the reduction in personal mobility
548 due to the pandemic. Liu et al. (13) note that Spain’s 2020
549 emissions due to power generation were almost 25% lower
550 than in 2019 due to investment in renewable energy. A post-
551 COVID economic recovery represents an opportunity to invest
552 in carbon-reducing technologies (56), as long as the need to
553 balance short-term job creation with long-term retraining is
554 accounted for (57). If this investment was able to continue
555 the trend of a 5.4% decrease in global CO₂ emissions per year,
556 we would reach “preindustrial” (circa 1850) emissions levels
557 in approximately 18 years.

558 Strengths and weaknesses of current observing systems

560 Understanding how the COVID-19 pandemic has altered AQ
561 and the carbon cycle has relied heavily on the multifaceted
562 observing system built over the past two decades, including satel-
563 lites, dense ground-based observing networks, Earth system
564 and chemical transport models, and techniques to assimilate
565 observations into these models. Novel data on human activity
566 (particularly internet-of-things mobility data, crowdsourced
567 air traffic data, and even news reports) have also played a
568 vital role in both understanding how human behavior changed

during the pandemic and quantifying the effect of that change
on anthropogenic emissions.

569
570
571 Nevertheless, there remain important gaps in our observ-
572 ing network. First, space-based detection of VOCs remains a
573 challenging problem, yet quantitative measurements of key bio-
574 genic (e.g. isoprene, terpenes) and anthropogenic (e.g. ethene,
575 propene) contributors to VOC OH reactivity are needed to
576 identify the dominant chemistry governing AQ around the
577 globe. Second, as we saw in the LA Basin case study, disen-
578 tangling primary PM emission, secondary PM formation, and
579 meteorological drivers of PM concentration is crucial to under-
580 stand which processes control PM exposure. Given the serious
581 health impacts of PM exposure, work towards an integrated
582 surface and space-based system that can differentiate these
583 processes is needed to elucidate the optimum approaches to
584 reducing PM exposure.

585 In regards to climate-relevant observations, spatiotempo-
586 rally broader and denser space-based GHG observations would
587 provide a highly valuable empirical constraint on changes to
588 anthropogenic and biogenic carbon fluxes. A satellite instru-
589 ment that provided comparable observations to the BEACO₂N
590 network in the San Francisco Bay area (~ 2 km resolution,
591 strong sensitivity to the near-surface atmosphere, urban-scale
592 coverage) could apply similar inversion techniques as Turner
593 et al. (16) to infer key sectors’ emissions in cities around the
594 world. It is also clear that our current network of near-real
595 time ocean carbon uptake measurements are not sufficient
596 to disentangle internal variability in the air-sea carbon flux
597 from changes driven by reductions in anthropogenic emissions
598 (25). Expanding this network or developing new methods to
599 constrain the air-sea carbon flux from space will be necessary
600 to quantify the impact of anthropogenic emissions reductions
601 on atmospheric CO₂ mixing ratios.

602 Conclusions

603 The COVID-19 pandemic and associated changes in human
604 behavior represent an unprecedented rapid change in anthro-
605 pogenic emissions to the atmosphere. Due to the large differ-
606 ences in relevant atmospheric lifetimes for constituents central
607 to AQ and climate, clear changes in local AQ but not global
608 GHG trajectories were observed. Changes in AQ were very
609 spatially heterogeneous, demonstrating that the same strate-
610 gies to improve AQ do not apply equally well to all regions.
611 Additionally, changes in AQ in the Los Angeles Basin corre-
612 lated with temperature, precipitation, and severe wildfires,
613 indicating that shifts in these quantities associated with cli-
614 mate change will at least partially offset gains in AQ made
615 from past and future reductions in anthropogenic emissions.

616 Despite large disruptions in transportation emissions sec-
617 tors, the global-scale change in the CO₂ growth rate was less
618 than interannual variability. This is due to a combination of re-
619 duced ocean uptake of CO₂, a recovery of CO₂ emissions in the
620 second half of 2020, and large interannual variability in land
621 carbon fluxes. That recovery indicates that expecting changes
622 to individual behavior to be sufficient to halt the increase of
623 GHGs in the atmosphere is unrealistic. Instead, incentives to
624 deploy new methods to systematically and sustainably reduce
625 carbon intensity are needed. Given the bidirectional feedback
626 between climate and AQ, it is clear that climate and AQ can
627 no longer be considered separate problems; prompt action to
628 reduce anthropogenic carbon emissions is essential not only to

629 avert direct climate impacts, but to avoid giving up decades
630 of hard-won progress in improving urban AQ.

631 Materials and Methods

632 Full methods are available in the SI. Analysis of LA Basin AQ used
633 data from CA Air Resources Board monitors, filtered for complete
634 data records in the 2015 to 2020 period. 1 h daily maximum (DM)
635 NO₂ and temperature, 8 h DM O₃, and 24 h average PM were
636 calculated from this data. OPE was derived from model simulations
637 using multiconstituent assimilation of multiple satellite measure-
638 ments in the MIROC-CHASER model (32). OPE calculated by
639 comparing modeled O₃ production and NO_x emission difference
640 between baseline (2010 to 2019) and reduced 2020 emissions. Separate
641 PM_{2.5} simulations used GEOS-Chem v9-02 with NO_x emissions
642 consistent with the OPE simulations: baseline NO_x emissions used
643 HTAP v2 scaled to 2017 using satellite-derived emissions reduction
644 ratios and COVID NO_x emissions were scaled down by the same
645 factor as in the OPE simulations. The TROPOMI timeseries analy-
646 sis first regridded native TROPOMI pixels to a 0.01° × 0.01° grid
647 and filtered to primarily remove cloud and snow/ice contaminated
648 scenes. The timeseries show the 75th percentile of 15-day rolling
649 average NO₂ columns in a 1° × 1° box around each city.

650 Global CO₂ emissions estimates were derived from an array of
651 near-real time data on power generation, industry, transport, and
652 fuel consumption. XCO₂ growth rates were derived from OCO-2 v10
653 ocean glint data and XCH₄ growth rates from TCCON GGG2014
654 data. The data shown are 15-day running averages deseasonalized by
655 fitting a four-harmonic curve. Expected CH₄ trends we computed
656 from a two-box model (representing the two hemispheres) using
657 prescribed OH concentrations and constant CH₄ emissions after
658 2012. TCCON data can be obtained from the TCCON Data Archive
659 hosted by CaltechDATA (<https://tccondata.org/>). The authors thank
660 the TCCON science team for their effort in providing this data.

661 Publicly available datasets are listed along with data generated
662 from this study and stored in public facing repositories in the SI,
663 table S1. Emissions data for Figs. 3 and 9 are given in Table S2.
664 Data for the OPE values in Fig. 7 is given in Table S4. Emissions
665 and OPE data also included as Excel SI files.

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