US COVID-19 shutdown demonstrates importance of background NO_2 in inferring NO_x emissions from satellite NO_2 observations

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Key Points:

- COVID-19 US shutdown tested the ability of satellite NO_2 data to capture NO_x emission trends
- Satellite NO_2 show muted response to COVID-19 shutdown because of NO_2 background contribution to tropospheric column sensed from space
- Summertime NO_2 background has been rising in the US over the past decade and this is not captured by GEOS-Chem model

Abstract

Satellite NO_2 measurements are used extensively to infer nitrogen oxide (NO_x) emissions and their trends, but interpretation can be complicated by background contributions to the NO_2 column sensed from space. We use the step decrease of US anthropogenic emissions from the COVID-19 shutdown to compare the responses of NO_2 concentrations observed at surface network sites and from satellites (OMI, TROPOMI). After correcting for differences in meteorology, surface NO_2 measurements for 2020 show decreases of 20% in March-April and 10% in May-August compared to 2019. The satellites show much weaker responses in March-June and no decrease in July-August, consistent with a large background contribution to the NO_2 column. Inspection of the long-term OMI trend over remote US regions shows a rising summertime NO_2 background from 2010 to 2019 that is not captured by the GEOS-Chem model.

Plain Language Summary

Nitrogen oxides (NO_x) emitted from combustion are a major source of air pollution. Satellite observations of nitrogen dioxide (NO_2) have been used to infer NO_x emissions but this inference is complicated by NO_2 present in background air. Here we show that this NO_2 background results in a muted response of the satellite observations to the abrupt drop in NO_x emissions from the US COVID-19 shutdown. The NO_2 background over the US has increased in the past decade, masking the effect of emission decreases. Understanding this background NO_2 and its rise is important not only for inferring NO_x emissions but also for explaining the current rise in global tropospheric ozone.

1 Introduction

Nitrogen oxide radicals ($NO_x \equiv NO + NO_2$) are critical to air quality. They drive the production of ozone and nitrate particulate matter, and are responsible for acid and nitrogen deposition. Fossil fuel combustion is the main anthropogenic source of NO_x, while lightning and soils are important natural sources. Satellite observations of tropospheric NO_2 columns by solar backscatter, in particular from the Ozone Monitoring Instrument (OMI) launched in 2004, have enabled global monitoring of NO_x emissions and their trends (Martin et al., 2003; Stavrakou et al., 2008; Lamsal et al., 2011; Duncan et al., 2016; Krotkov et al., 2016; Miyazaki et al., 2017; Qu et al., 2020). The OMI observations over the contiguous United States (CONUS) show a 2005-2009 decrease consistent with the US EPA National Emission Inventory (NEI) and with the surface NO_2 monitoring network (Russell et al., 2012; Lamsal et al., 2015), but no further decrease after 2009 despite continued decrease of NO_x emissions according to the NEI (Jiang et al., 2018). It is not clear if this flattening of the NO_2 trend in the OMI data reflects major errors in the NEI (Jiang et al., 2018) or an increasing contribution from background NO_2 unrelated to surface anthropogenic NO_x emissions (Silvern et al., 2019). This background could originate from lightning, aircraft, soils, wildfires, and long-range transport of pollution (Zhang et al., 2012). Aircraft observations over the Southeast US in summer show that background NO_2 above 2 km altitude could contribute 70-80% of the summertime tropospheric NO_2 column sensed by satellite (Travis et al., 2016), reflecting the increased sensitivity to NO_2 with altitude as a result of gas, aerosol, and cloud scattering (Martin et al., 2002).

The shutdown of the US economy during the COVID-19 crisis provides an opportunity to investigate the response of the satellite NO_2 observations to the abrupt NO_x emission reductions.

40 out of 48 states in CONUS issued mandatory stay-at-home orders during March and April 2020 (Moreland et al., 2020), greatly decreasing emissions from transportation and to a lesser extent from industry. Liquid fuel consumption in the US dropped by 21% and coal consumption dropped by 25% in the second quarter of 2020 compared to 2019 (EIA, 2020). Anthropogenic NO_x emissions decreased by 10-40% in major US cities in March-April (Goldberg et al., 2020; Keller et al., 2020; Naeger & Murphy, 2020; Xiang et al., 2020).

Here we examine the response of NO_2 satellite observations to the COVID-19 shutdown in CONUS and compare it to trends in the NO_2 surface monitoring network. This allows us to determine if satellites can accurately track changes in surface NO_x emissions or if background NO_2 plays a confounding role, addressing the conundrum posed by the flattening of the OMI NO_2 trend over the past decade. We go on to further examine the 2005-2019 OMI trends in the context of the surface NO_2 observations and the GEOS-Chem model.

2 Data and model

We use hourly surface measurements of NO_2 concentrations from the EPA Air Quality System (AQS) accessed through the application programming interface (API, aqs.epa.gov/aqsweb/documents/data_api.html). The measurements are made by a chemiluminescence analyzer with a molybdenum converter, which has been reported to have interferences from NO_x oxidation products (Dunlea et al., 2007; Reed et al., 2016), but we assume here that these do not affect the relative long-term trends. For the 2019-2020 analysis, we only include AQS sites that have reported measurements for each month in both 2019 and 2020, for a total of 328 sites. We average the data on a 0.5° × 0.625° grid to define collocation with satellite data and for meteorological trend correction. For the 2005-2019 trend analysis, we only include grid cells with continuous AQS records over the period, for a total of 168 0.5° × 0.625° grid cells.

Satellite observations of tropospheric NO_2 vertical column densities are from two instruments: TROPOMI (2018-present) (Veefkind et al., 2012) and OMI (2005-present) (Levelt et al., 2006, 2018). Pixel resolutions are 3.5 ×5.5 km^2 for TROPOMI (3.5× 7 km^2 before August 2019) and 13×24 km^2 for OMI. The TROPOMI retrieval is the version 1 offline product (www.tropomi.eu/data-products/nitrogen-dioxide). We use two different OMI retrievals: the version 4 NASA NO_2 product (disc.gsfc.nasa.gov/datasets/OMNO2_003/summary) (Lamsal et al., 2020) and the QA4ECV OMI NO_2 retrieval from KNMI (temis.nl/qa4ecv/NO2.html) (Boersma et al., 2018). For both OMI and TROPOMI retrievals, we filter the data using the quality flags and only include observations with cloud fraction < 0.2, surface albedo < 0.3, solar zenith angle < 75°, and view zenith angle < 65°. We also exclude OMI data affected by the so-called row anomaly. We average the satellite data on the same 0.5° × 0.625° grid as the AQS surface NO_2 data.

We use the GEOS-Chem chemical transport model version 12.9.2 (doi.org/10.5281/zenodo.3959279) to simulate the contribution of meteorology to changes in surface and column NO_2 concentrations at AQS sites between 2019 and 2020, assuming the same emissions in both years in the model. GEOS-Chem is driven by MERRA-2 assimilated meteorological data from the NASA Global Modeling and Assimilation Office (GMAO) and has been used in many studies of NO_x sources and chemistry over the US (Zhang et al., 2012; Fisher et al., 2016; Lee et al., 2016; Travis et al., 2016; Jaeglé et al., 2018).. We conduct nested simulations over the US (126°-66°W, 13°-57°N) at a horizontal resolution of 0.5° × 0.625° with

dynamic boundary conditions generated from a global simulation with 4° × 5° resolution. Emissions are from the NEI 2011 scaled to 2019 using national emission totals (EPA, 2020). We also use 2019 open fire emissions from GFED4 (van der Werf et al., 2017) for both 2019 and 2020 simulations. The spatial and monthly climatology of lightning NO_x emissions is constrained by LIS/OTD satellite observations averaged over 1995-2013. Lightning emissions in the model can vary from year to year as determined by MERRA-2 convective mass flues (Murray, 2016). Soil NO_x emissions are calculated following Hudman et al. (2012).

Long-term GEOS-Chem simulations from 2005 to 2017 are from Silvern et al. (2019), using version 11-02c of the model at 0.5° × 0.625° grid resolution driven by MERRA-2 meteorology and including yearly NEI emission trends. Open fire emissions in that simulation are from the daily Quick Fire Emissions Database (QFED) (Darmenov & da Silva, 2013). Soil NO_x emissions are decreased by 50% in the midwestern US in summer based on a comparison with OMI NO_2 (Vinken et al., 2014). A full description of this long-term GEOS-Chem simulation is provided in Silvern et al. (2019).

3 Responses of NO_2 observations to the COVID-19 shutdown

Figure 1 shows the relative changes in 24-hour average NO_2 surface air concentrations measured at the AQS sites in March-April 2020 compared to March-April 2019. Most of the monitoring sites are in urban areas, though a number are in oil/gas production regions (Edwards et al., 2014). We expect the AQS NO_2 trends to closely track NO_x emission trends, after corrections for meteorology given below. The decreases in Figure 1 average 21% across CONUS with no apparent regional patterns. Thick-rimmed circles identify the 0.5° × 0.625° grid cells with the 5% highest mean NO_2 concentrations (exceeding 11 ppb) in March-April 2019. The decreases at these sites average 26%, similar to the CONUS average.

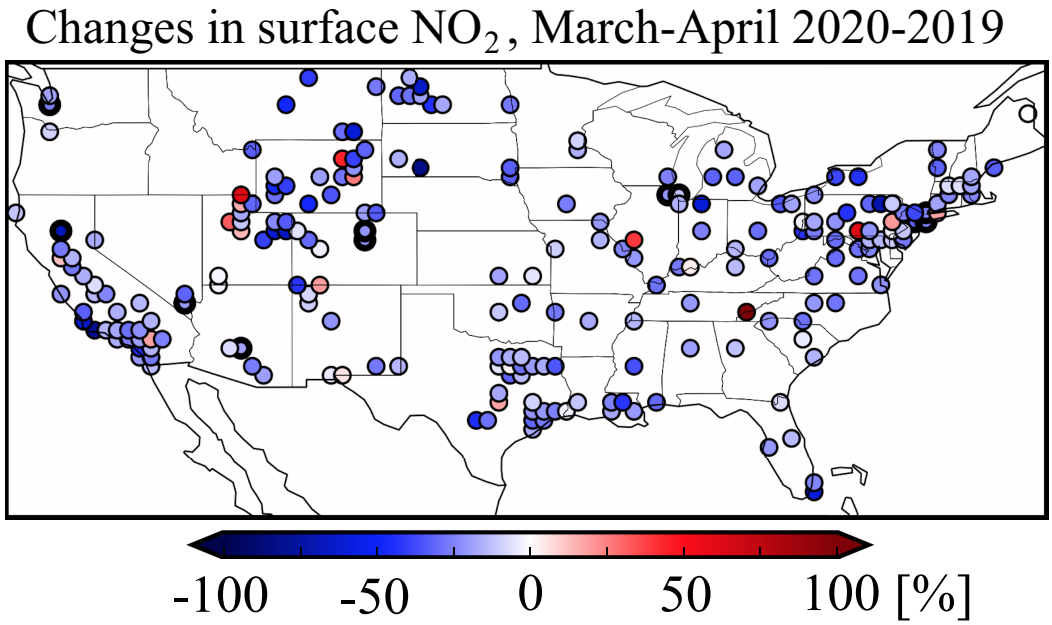

Figure 1. Changes in 24-hour mean surface NO_2 concentrations in March-April 2020 relative to March-April 2019. Observations are from the US EPA Air Quality System (AQS) network binned

into 0.5° × 0.625° grid cells. Thick rims identify grid cells with the 5% highest concentrations in March-April 2019.

Figure S1 shows the changes in OMI NO_2 vertical column densities between March-April 2019 and 2020. The data are noisy, presumably reflecting the degradation of the instrument in recent years (KNMI, 2020). Similar observations but with higher pixel resolution (3.5×5.5 km^2 at nadir) are available from the TROPOMI satellite instrument launched in October 2017 (Veefkind et al., 2012; Griffin et al., 2019). Figure 2 shows the TROPOMI NO_2 vertical column densities observed over CONUS in March-April 2019 and 2020 on the 0.5° × 0.625° grid. Differences between 2019 and 2020 are much less uniform than for the AQS sites, and large rural areas show increases. The average decrease from March-April 2019 to March-April 2020 is 4% across CONUS, 11% at the ensemble of AQS monitoring sites in Figure 1, and 21% at the AQS sites with 5% highest NO_2. The weaker decrease of TROPOMI NO_2 relative to the AQS data is consistent with a dampening effect from background NO_2. The dampening effect is least where surface NO_2 is highest.

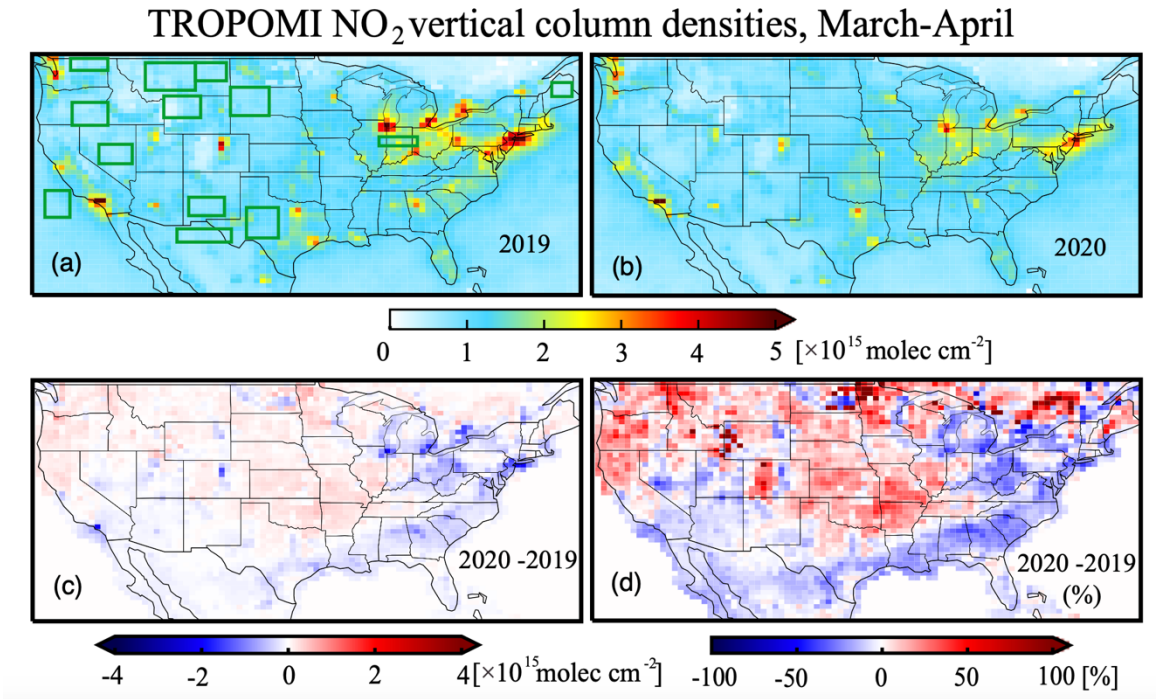

Figure 2. Mean tropospheric vertical column densities of NO_2 measured by TROPOMI in March-April (a) 2019 and (b) 2020. Panels (c) and (d) show the absolute and relative differences between 2020 and 2019. The green rectangles in panel (a) represent the 13 remote regions used in the long-term trend analysis of Figure 4 (b).

Variations in meteorology can be a confounding factor when interpreting year-to-year changes in concentrations (Goldberg et al., 2020). We diagnosed this meteorological influence in both AQS and satellite NO_2 by conducting GEOS-Chem simulations for 2019 and 2020 with no changes in anthropogenic and open fire emissions. Results are shown in Figure S2. This meteorological

influence is relatively small in March-April but more important in other months. We subtract it from the observed changes in what follows.

Figure 3 shows the mean meteorology-corrected changes of NO_2 concentrations at AQS sites for January-August 2020 relative to 2019 and the changes of TROPOMI tropospheric NO_2 columns for the same sites. Here we have again segregated the 5% of sites with the highest AQS NO_2 concentrations, where the relative background influence would be expected to be least. For those sites, January-February 2020 (before the COVID-19 shutdown) had 5% lower NO_2 than the same period in 2019. The COVID-19 shutdown decreased surface NO_2 concentrations at these top 5% of AQS sites by 22% in March-April, 13% in May-June, and 14% in July-August. TROPOMI tracks these changes except in July-August where it shows no significant change in 2020 relative to 2019. For the other AQS sites, trends in surface NO_2 concentrations are similar to the top 5% but TROPOMI shows no trend in May-June and an increase in July-August. We performed the same analysis with OMI data (Figure S3) and obtained similar results.

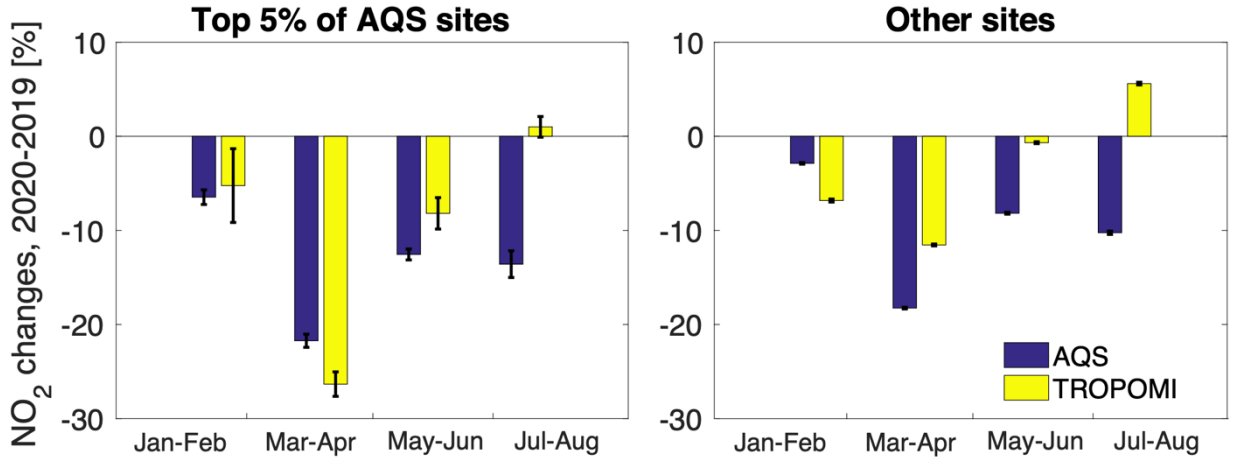

Figure 3. Mean bimonthly changes in NO_2 concentrations at AQS sites between 2019 and 2020. Changes in AQS surface air NO_2 concentrations are compared to changes in TROPOMI tropospheric NO_2 columns sampled at the same sites. AQS sites with the 5% highest 2019 NO_2 concentrations on a 0.5^ox0.625^o grid (Figure 1) are segregated. The effects of meteorological changes have been subtracted with a GEOS-Chem simulation. The error bars represent the normalized standard errors of the changes.

The results in Figure 3 show that TROPOMI is increasingly unable to capture the decrease of NO_x emissions documented by the AQS sites in the seasonal progression from spring to summer. This is consistent with an obfuscating contribution of background NO_2 from lightning, soils, and wildfires, which would be lowest in winter and highest in summer (Zhang et al., 2012).

4 Implications for the long-term trend of NO_2 observed from satellite

The muted response of the satellite NO_2 observations to the COVID-19 shutdown over CONUS implies a large background contribution to the tropospheric NO_2 columns sensed from space. It supports the previous argument from Silvern et al. (2019) that the non-decreasing NO_2 background would have dampened the long-term 2005-2017 trend of OMI NO_2 averaged over CONUS relative to that expected from declining NO_x emissions. We find little dampening when sampling OMI NO_2 observations at grid cells with continuous AQS records for 2005-2019 (Figure 4a), which likely reflects the urban location of these long-term sites (Figure S4). The OMI NO_2 post-2010 observations show a weaker trend in summer than winter that is not seen in the AQS data but is captured by GEOS-Chem simulations. The weaker trend in summer is attributed in GEOS-Chem to an increasing relative contribution of background NO_2 as NO_x emissions decrease.

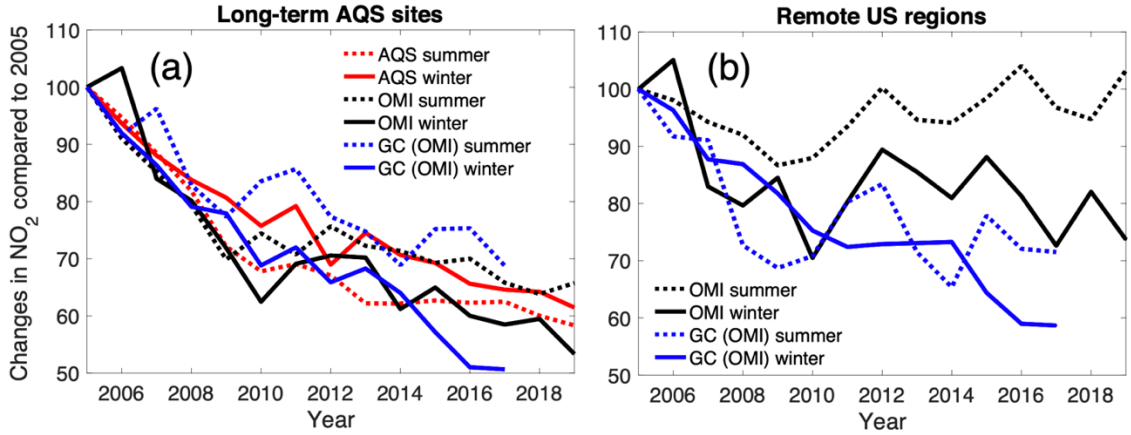

Figure 4. Long-term trends in NO_2 over CONUS, 2005-2019. (a) Trends averaged over the (mainly urban) AQS sites with continuous records of surface NO_2 concentrations for 2005-2019 (Figure S4). The trends are relative to 2005 and shown separately for summer (JJA) and winter (DJF). Trends in OMI tropospheric NO_2 columns (from NASA retrieval) averaged over the same sites are also shown, along with a GEOS-Chem (GC) simulation of the OMI NO_2 data previously reported by Silvern et al. (2019). (b) Trends in OMI and GEOS-Chem tropospheric NO_2 columns for summer and winter over 13 remote US regions (Figure 2) as defined by Russell et al. (2012).

To better isolate the contribution of background NO_2, we examined the long-term OMI trends averaged over 13 remote regions previously defined by Russell et al. (2012) and mainly in the western US (Figure 2). Results shown in Figure 4b indicate a decrease over 2005-2009 but not afterward. For the 2010-2019 period, OMI over these remote regions shows no trend in winter and a 19% increase in summer, implying a decadal rise in summertime background NO_2 that GEOS-Chem does not capture. Better understanding of this background and its trend is obviously needed.

That background cannot be easily subtracted from the satellite observations because it is not uniform (Marais et al., 2018, 2020). Cloud-sliced satellite observations could isolate free tropospheric NO_2 but need better accuracy than available at present (S. Choi et al., 2014; Marais et al., 2018). This may be achievable with upcoming geostationary observations (Zoogman et al., 2017; W. J. Choi et al., 2018). Aside from complicating the interpretation of NO_x emission trends from satellite data, a rising NO_2 background could help explain the current rise in background tropospheric ozone (Gaudel et al., 2018).

5 Conclusions

We have used the unintended experiment of the COVID-19 economic shutdown in the US starting in March 2020 to demonstrate the impact of background NO_2 on interpreting NO_x emission trends from satellite observations of NO_2 vertical column densities. After subtracting the impact of meteorology in both surface and satellite NO_2 observations, we find that the satellite observations can capture the magnitude of NO_x emission reductions from March to June 2020 only for the sites with the highest levels of surface NO_2. At other sites, the response of the satellite observations to the changes in emissions is strongly muted. The satellite data show no reduction in July-August 2020 when background NO_2 is expected to be seasonally highest. Further inspection of long-term trends in the satellite NO_2 data over remote US regions shows a 2010-2019 increase in summer, implying a rise in background NO_2 that is not captured by the GEOS-Chem model. Better quantitative understanding of the factors contributing to background NO_2 and its trend is urgently needed for the interpretation of satellite data, in particular from the upcoming geostationary constellation for air quality.

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