



Supporting Information for:

Disentangling the impact of the COVID-19 lockdowns on urban NO₂ from natural variability

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This PDF file includes:

Further description of TROPOMI NO₂ processing technique and associated uncertainties

Further description of methodologies used to calculate NO₂ drops during COVID-19

Figures S1 to S5

Table S1

1. TROPOMI NO₂

1.1 Air Mass Factors and Uncertainty Estimates

The slant tropospheric column is converted to a vertical column using a quantity known as the air mass factor (Palmer et al., 2001). The air mass factor is the most uncertain quantity in the retrieval algorithm (Lorente et al., 2017), and is a function of the surface reflectance, the NO₂ vertical profile, and scattering in the atmosphere among other factors (Lamsal et al., 2014).

Using accurate and high-resolution data (spatially and temporally) as inputs in calculating the air mass factor can significantly reduce the overall errors of the air mass factor (Choi et al., 2019; Goldberg et al., 2017; Laughner et al., 2016, 2019; Lin et al., 2015; Liu et al., 2019; Russell et al., 2011; Zhao et al., 2020) and thus the tropospheric vertical column content.

Operationally, the TM5-MP model ($1 \times 1^\circ$ resolution) (Williams et al., 2017) is used to provide the NO₂ vertical shape profile and the climatological Lambertian Equivalent Reflectivity ($0.5 \times 0.5^\circ$ resolution) (Kleipool et al., 2008) is used to provide the surface reflectivities. The operational air mass factor calculation does not explicitly account for aerosol absorption effects, which are accounted for in the effective cloud radiance fraction. While the operational product does have larger uncertainties in the tropospheric column contents than a product with higher spatial resolution inputs, we limit our analysis to relative trends, which dramatically reduces this uncertainty. The uncertainty in any daily measurement in the operational slant column data has been assigned to be approximately 5.7×10^{14} molecules-cm⁻² (van Geffen et al., 2020). This equates to roughly a 5-10% uncertainty over polluted areas. However, because we are averaging over many days (~20-40), we assume that random errors will cancel due to the large number of observations used. This leaves only the systematic errors. Here, we assign the AMFs and tropospheric vertical column contents a systematic uncertainty of 20% in the trends (McLinden et al., 2014). This systematic uncertainty may be largest over areas with changing snow cover, such as Minneapolis, Chicago, Toronto, and Montreal. We calculate total uncertainty as the quadrature of the uncertainty associated with this potential systematic bias and the standard deviation of the three Methods. These are listed in Table S1.

1.2 Re-gridding of TROPOMI NO₂

For our analysis we re-grid the operational TROPOMI tropospheric vertical column NO₂, with native pixels of approximately 3.5 × 7 km², to a newly defined 0.01° × 0.01° grid (approximately 1 × 1 km²) centered over the continental United States (CONUS; corner points: SW: 24.5° N, 124.75° W; NE: 49.5° N, 66.75° W). Before re-gridding, the data are filtered so as to use only the highest quality measurements (quality assurance flag (QA_flag) > 0.75).

2. Description of Methodologies 2 & 3

2.1 Method 2: Normalization of Daily TROPOMI NO₂ using ERA5

We use TROPOMI NO₂ data from 2018 – 2019 as analog data to normalize 2020 data. Essentially, our method is searching through the 2018 – 2019 archive to find a meteorological analog to the current conditions and then adjusting the current day's conditions based off that analog.

For each day of the record, we modify the original observed TROPOMI NO₂ based on its value compared to a "baseline" which we set as a weekday in April with 3 m/s southwest winds. For each day, n , and each city, i , the normalized NO₂, $\widehat{NO}_{2n,i}$, is calculated as follows:

$$\widehat{NO}_{2n,i} = \frac{NO_{2n,i}}{f_{totaln,i}}$$

The subscript i represents a city-specific average within a 0.4° × 0.4° box (i.e., ~20 km radius) surrounding the city center.

The four adjustment factors are: sun angle, wind speed, wind-direction, and day-of-week. While other conditions affect NO₂ amounts they are either interrelated to the aforementioned factors or can be considered secondary. Each of the four individual factors are multiplied together to get a "total adjustment factor". The "total adjustment factor", f_{total} is calculated for each day, n , and each city, i , as follows:

$$f_{totaln,i} = [f_{sun-angle}]_n [f_{day-of-week}]_n [f_{wind-speed}]_{n,i} [f_{wind-dir}]_{n,i}$$

For the sun angle factor, we calculate this using a cosine fit. For each julian date, n , the sun angle factor ($f_{sun-angle}$) can be calculated as follows:

$$f_{sun-angle}_n = \frac{0.75 + 0.25 * \cos \left[2\pi \frac{n + 11}{365} \right]}{0.75 + 0.25 * \cos \left[2\pi \frac{n_d + 11}{365} \right]}$$

At the winter solstice, December 21st ($n = -11$ or $n = 354$) the numerator value is 1 and at the summer solstice, June 21st ($n = 171$) the numerator value is 0.5. The variable n_d represents the normalization day, in this case April 15th ($n_d = 105$). The aforementioned equation is only valid for locations north of the Tropic of Cancer (23.4°N).

For the wind speed factor, we fit a third-order polynomial using analog winds speeds from the 2018 – 2019 TROPOMI time frame. Wind speeds of 5 m/s would yield a correction factor of 1. Values larger than 1 represent winds slower than 5 m/s and values smaller than 1 represent winds faster than 5 m/s. This fit allows us to calculate a correction factor given any city-specific wind speed.

For the wind direction factor, we calculate a correction factor normalized to southwest winds. Wind directions are grouped into the following categories: 0 – 90 ° are southwest, 90 – 180 ° are northwest, 180 – 270 ° are northeast, and 270 – 360 ° are southeast. Once the wind speed is grouped into a specific category, the factor is defined based on its relation to the climatological wind direction; northwest for New York City and Washington D.C., and northeast for Los Angeles. Daily winds which are typical of the climatological wind direction yield a correction factor of 1.

Lastly, for the day-of-week factor, we assume 15% lower values on Saturdays and 30% lower values on Sundays. We assume all weekdays have similar emissions rates to each other. Weekdays have a factor of 1, Saturdays a factor of 0.85 and Sundays a factor of 0.70. These assumptions are broadly consistent with literature demonstrating day-of-week NO_x emissions patterns.

As an example, a stagnant day in January may be lowered by a factor of ~2 to "normalize" to a 5 m/s April weekday, whereas a very windy weekend day in April might be increased by a factor of 1.5 to account for the faster than normal winds and the weekend effect.

Method 3: Normalization of Daily TROPOMI NO₂ using a CTM

We infer expected NO₂ columns (V_{ex}) during the lock-down period (t_{covid}) using the output from the GEM-MACH model (Moran et al., 2009; Pendlebury et al., 2018). The operational version of the model, used in this study, has a 10×10 km² grid cell size with 80 vertical levels (from the surface to about 0.1 hPa), provides hourly output, and includes emissions, chemistry, dispersion, and removal processes of 41 gaseous and eight particle species. The emissions used in the model are processed using the Sparse Matrix Operator Kernel Emissions (SMOKE) (Coats, n.d.) and account for seasonal changes; changes in emissions due to the COVID-10 lock-downs are not considered in the model framework.

In a first step the GEM-MACH NO₂ vertical levels in the boundary layer (up to approximately 2 km) are summed to a column amount using the model's pressure and temperature profile (Côté et al., 1998). Since the GEM-MACH model currently does not contain any NO_x sources in the free troposphere (such as aircraft or lightning emissions), the NO₂ model concentrations decrease to 0 above the planetary boundary layer (PBL). A free tropospheric column (from 2 km to 12 km) is added to the GEM-MACH PBL vertical column densities (VCDs) using a monthly GEOS-Chem run (0.5x0.67° resolution, version v8-03-01; <http://www.geos-chem.org>) (Bey et al., 2001; McLinden et al., 2014). The model VCDs are then mapped in space and time to the TROPOMI observations, and treated like the observations, where data with $qa < 0.75$ are filtered and averaged over the city center using a 28-day running mean.

The expected VCDs (V_{ex}) are the 28-day running means of the modelled VCDs (V_M) during the lockdown period (t_{covid}). V_{ex} is scaled to remove any bias between the model and satellite (V_T) for the pre-lockdown period (t_{pre} , between February 1st and March 1st 2020):

$$V_{ex}(t_{covid}) = V_M(t_{covid}) \cdot \text{mean} \left(\frac{V_T(t_{pre})}{V_M(t_{pre})} \right).$$

Depending on the city, some dates within the t_{pre} time period may not be considered for the scaling, if there is a strong divergence between the model and the observations.

The estimated NO_2 drop is the average of the difference between the expected VCDs, $V_{ex}(t_{covid})$, and the observed TROPOMI VCDs, $V_T(t_{covid})$, between March 28th and April 16th, 2020 using the daily 28-day running means as shown in Figure 4.

3. Supplemental Figures

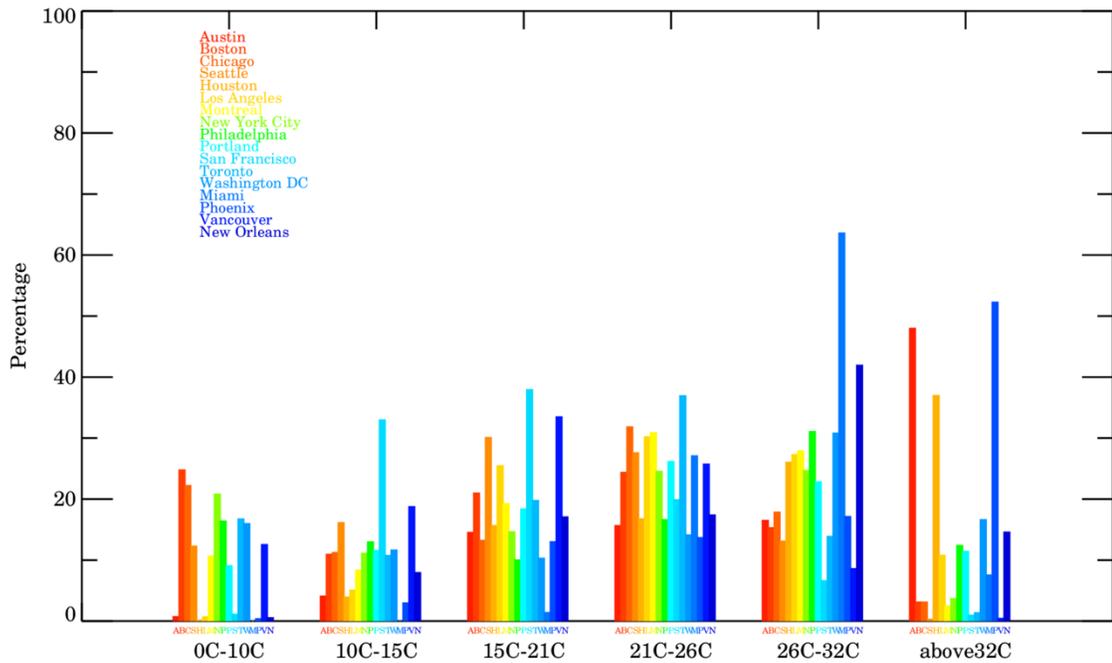


Figure S1. Frequency of daily maximum 2-m temperature within each bin, according to the ERA5 re-analysis. Each bar is a different city as noted by list in top left.

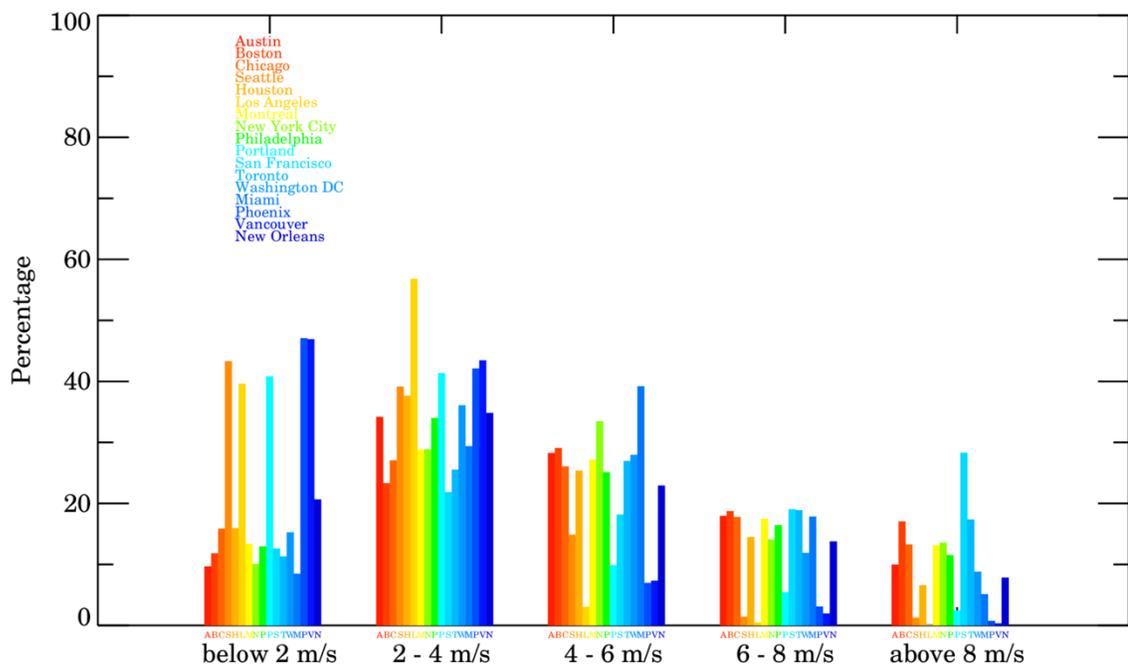


Figure S2. Frequency of 100-m afternoon (16Z-21Z) wind speed within each bin, according to the ERA5 re-analysis. Each bar is a different city as noted by list in top left.

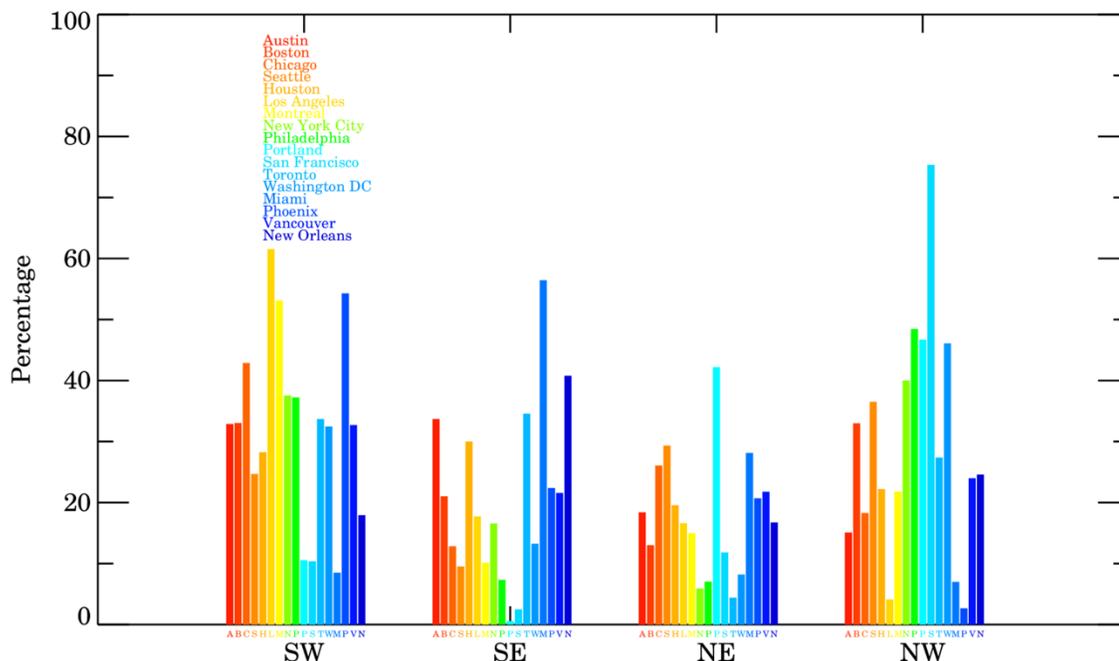


Figure S3. Frequency of 100-m afternoon (16Z-21Z) wind direction within each bin, according to the ERA5 re-analysis. Each bar is a different city as noted by list in top left.

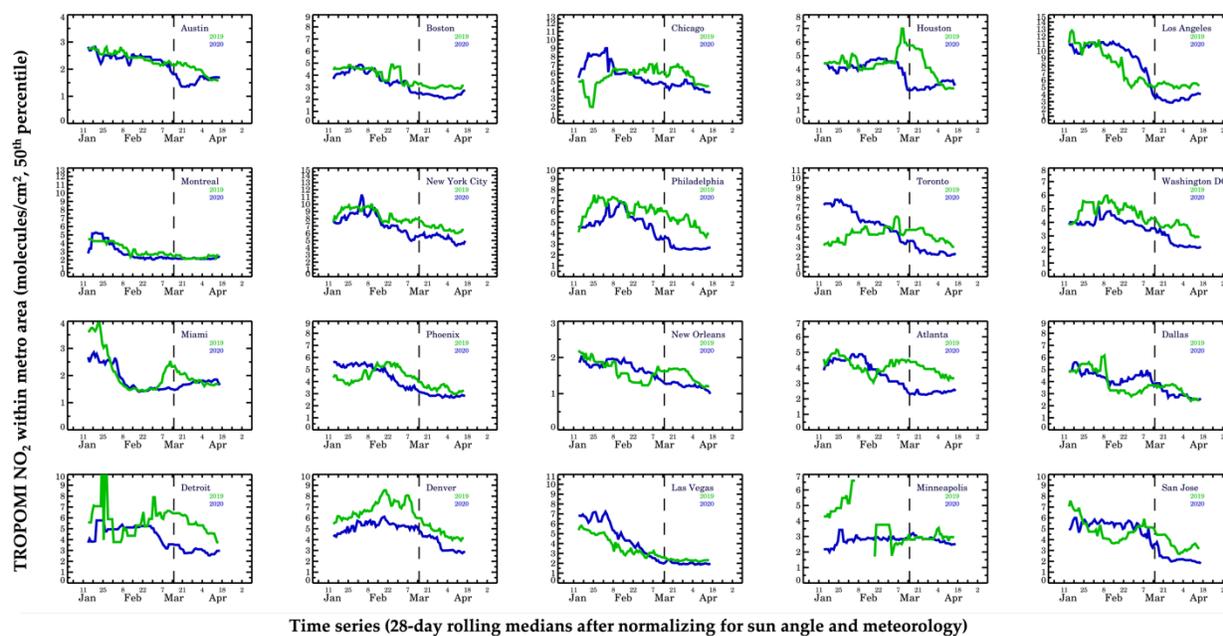


Figure S4. Trends in TROPOMI NO₂ since January 1 in 2019 and 2020. The lines represent the 28-day rolling median value (50th percentile) in a $0.4^\circ \times 0.4^\circ$ box centered on the city center for the largest cities (New York City, Los Angeles, Chicago, Toronto, Houston) and $0.2^\circ \times 0.2^\circ$ box in all other cities.

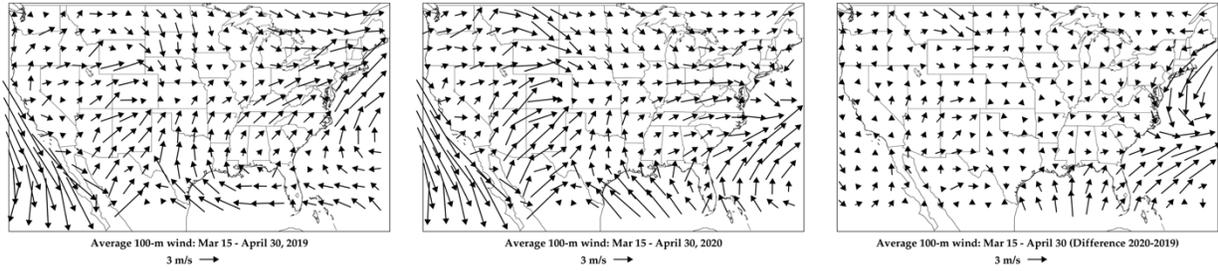


Figure S5. Average 100-m afternoon (16Z-21Z) wind speed and direction for March 15 – April 30 in (left) 2019, (center) 2020, (right) difference between the two years, according to the ERA5 re-analysis.

4. Supplemental Table

Table S1. Uncertainties associated with our methodology. Uncertainties are calculated as the quadrature of any potential systematic bias (20%) and the standard deviation of Methods 1 – 3.

City Name	Uncertainty
San Jose	20.1%
Toronto	20.3%
Los Angeles	23.6%
Philadelphia	21.3%
Atlanta	21.5%
Detroit	21.4%
Denver	21.2%
Montreal	20.2%
Boston	23.5%
Washington DC	24.2%
New York City	20.0%
New Orleans	20.5%
Las Vegas	26.1%
Phoenix	23.4%
Chicago	22.4%
Houston	23.6%
Austin	20.3%
Dallas	21.1%
Miami	22.0%
Minneapolis	20.3%

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