Supplementary Materials for

High-resolution Mapping of Nitrogen Oxide Emissions in Large US Cities from TROPOMI Retrievals of Tropospheric Nitrogen Dioxide Columns

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Text S1.

We adapt the model function proposed by Liu et al. (2022) with minor adjustment to infer $b$ and $\tau$, following:

$$f(x) = \left(\frac{LD_{\text{calm}}(x) - b_{\text{calm}}}{v \times t}\right) \times L \times e^{-\frac{x}{v \times t}} + b,$$

where $LD_{\text{calm}}(x)$ is a function of distance from the city center in a particular direction $x$ and integrated over a given distance in a direction $y$ (perpendicular to that of $x$). The mean NO$_2$ VCDs maps (2D) under calm wind conditions (wind speed < 2 m s$^{-1}$) are reduced to 1D (so-called NO$_2$ line densities) along the respective direction $x$ by integration across the direction $y$.

$b_{\text{calm}}$ represents the NO$_2$ background under calm wind conditions for each city, which is derived by analyzing the distribution of NO$_2$ VCDs. We first calculate the mean NO$_2$ VCD under calm wind conditions for grid cells within the lowest 1st percentile of NO$_2$ VCDs for each city. This produces a good approximation of the mean NO$_2$ VCD for grid cells with low NOx emissions (i.e., the lowest 1st percentile of NO$_x$ emissions) as verified by our previous study (Liu et al., 2022). We then multiply this mean VCD value by the spatial width of the across-wind integration interval to derive $b_{\text{calm}}$.

$L$ is the average width of the grid cell in a given direction $x$. $v$ is the mean GEOS-IT wind speed averaged from surface to 1000 m altitude in a given direction $x$, and $*$ denotes convolution.

We perform a nonlinear least-squares fit of $f(x)$ to the observed line densities under windy conditions, with $b$ and $\tau$ as the fitting parameters. We use the package of scipy.optimize.curve_fit from the Python software library to perform the fitting. The fit intervals are set consistent with those in Liu et al. (2022). Fitting results of insufficient quality (i.e., the correlation coefficient $R$ between the fitted and observed NO$_2$ line densities < 0.9, normalized root-mean-square deviation (NRMSD) between the fitted and observed NO$_2$ line densities > 10%, one standard deviation error of $\tau$ > 10%, and error of $\tau$ > 1h) are discarded. We perform the fit for all wind direction sectors and then average the fitted $b$ and $\tau$ with good quality, using the fit residuals as inverse weights, to yield a best estimate of $<b>$ and $<\tau>$ for a given city. The derived $<b>$ and $<\tau>$ are used as inputs for the 2D MISATEAM to infer NO$_x$ emissions. The standard deviation of the fit results for different wind directions has been used to quantify uncertainties of the derived emissions. Additional technical details are available at Liu et al. (2022).

Figure S1 displays the observed line densities for calm (blue circles) and southeasterly winds (red circles) around New York and the fitted model function $f(x)$ (red lines). Generally, $f(x)$ describes the observed downwind patterns very well; the coefficients of determination ($R^2$) between observation and fit are 0.90–0.97 for different wind directions. Results for other wind direction sectors are discarded due to the fitting results being of insufficient quality.

Text S2.

We apply 2D MISATEAM to 70 major cities with populations > 200,000 over the US (Table S1). For the application using TROPOMI NO$_2$ VCDs, we exclude 18 cities with too weak emissions signals, i.e., $b_{\text{calm}}$/mean VCDs > 50%. We derive valid fitting results for 39 cities (Fig. S2). The other 13 cities without valid results either have small correlation coefficients ($R < 0.9$) or large RMSD (NRMSD > 10%) or large
fitting errors (standard deviation error of $\tau > 10\%$ or error of $\tau > 1h$); those cities tend to have larger temporal variations in winds, which do not satisfy MISATEAM’s requirement for steady winds prior to satellite overpass (see Fig. S3 of Liu et al. (2022)). For the validation using the NU-WRF simulation, cities on the boundary of the NU-WRF domain, e.g., Seattle and San Francisco, are excluded from the validation, because the data for their inflow/outflow plumes are partially missing from the model output and thus do not meet the requirements of MISATEAM. This filtering results in a total of 60 cities. Consistent with the application using TROPOMI data, we discard 10 cities with too weak emissions signals and 17 cities which have large fitting errors. We derive valid results for 33 cities for the validation.
Figure S1: NO$_2$ line densities around New York for different wind direction sectors. Circles: NO$_2$ line densities for calm (blue circles) and (A) southeasterly, (B) southwesterly, (C) northerly, and (D) northwesterly winds (red circles) as a function of the distance $x$ to New York center. Red line: the fit result $f(x)$. The numbers indicate the fitted NO$_2$ lifetime ($\tau$) and background ($b$). NO$_2$ line densities are derived from TROPOMI NO$_2$ VCDs averaged from May through September, 2018-2021. NO$_2$ line densities for the remaining wind direction sectors are discarded due to the fitting results having insufficient quality.
Figure S2: Geographic distribution of investigated cities over the US. Cities are labeled by their IDs (see Table S1). The background is the tropospheric NO$_2$ vertical column density map averaged from May to September 2019.

Figure S3. Average NO$_x$ emission rates around New York City from May through September, 2019. (a) TROPOMI-derived NO$_x$ emissions $E$, (b) upscaling (a) to the same spatial resolution as that of NEI, 12 km$x$12 km, (c) NEI NO$_x$ emissions $E_{NEI}$. 
**Table S1.** Summary of cities investigated in this study.

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