Supplement of

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 τ, following:

$$f(x) = \frac{[LD_{calm}(x) - b_{calm}] \times L}{v \times \tau} * e^{-\frac{x}{v \times \tau}} + b, \tag{1}$$

where $LD_{calm}(x)$ 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_{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_{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 τ 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 τ > 10%, and error of τ > 1h) are discarded. We perform the fit for all wind direction sectors and then average the fitted b and τ 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_{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.
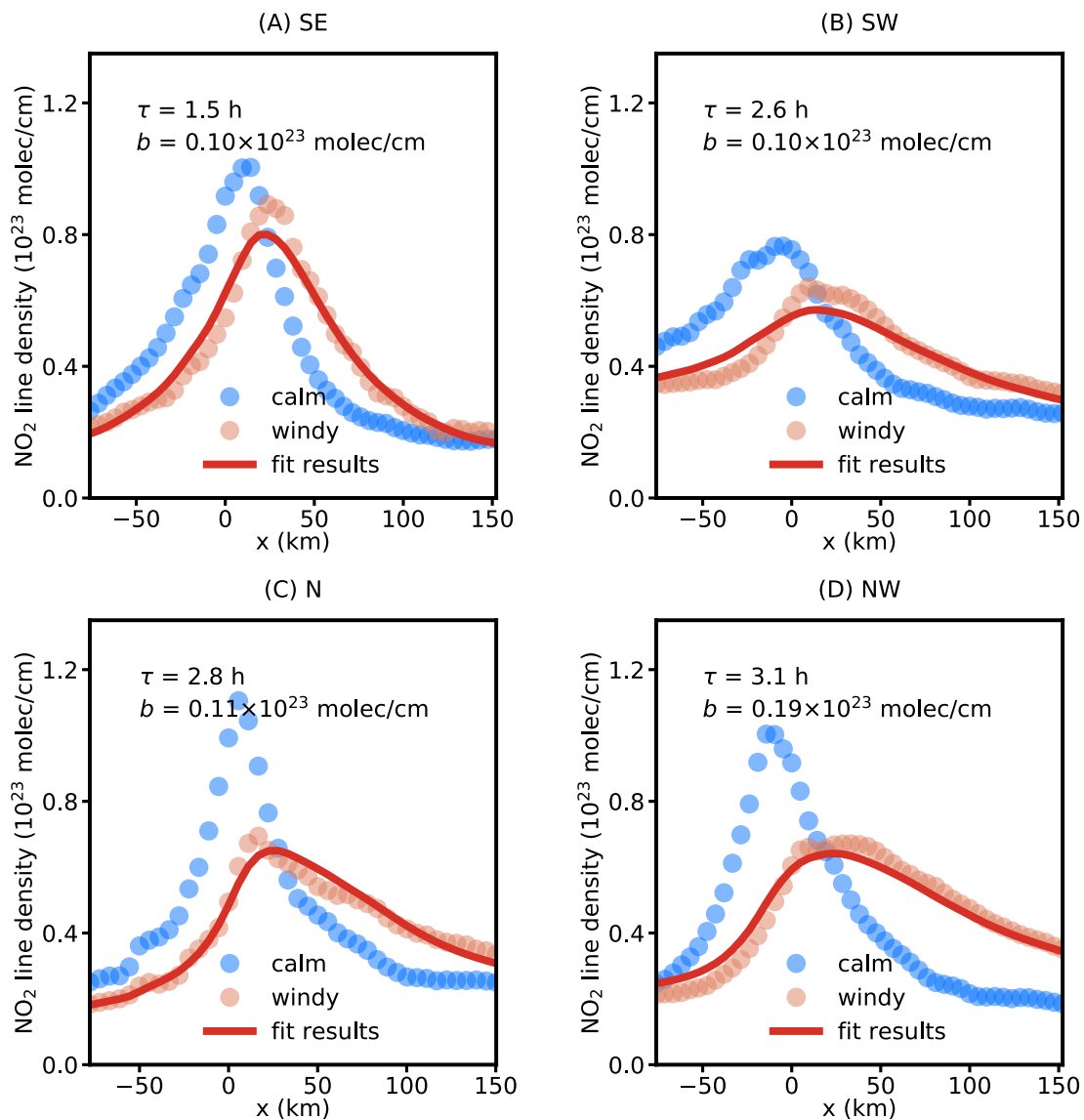
Figure S1: NO₂ line densities around New York for different wind direction sectors. Circles: NO₂ line densities for calm (blue circles) and (A) southeasterly, (B) southwesterly, (C) northerly, and (D) northwesterly winds (red circles) as a function of the distance x to New York center. Red line: the fit result $f(x)$. The numbers indicate the fitted NO$_x$ lifetime (τ) and background (b). NO₂ line densities are derived from TROPOMI NO₂ VCDs averaged from May through September, 2018-2021. NO₂ line densities for the remaining wind direction sectors are discarded due to the fitting results having insufficient quality.
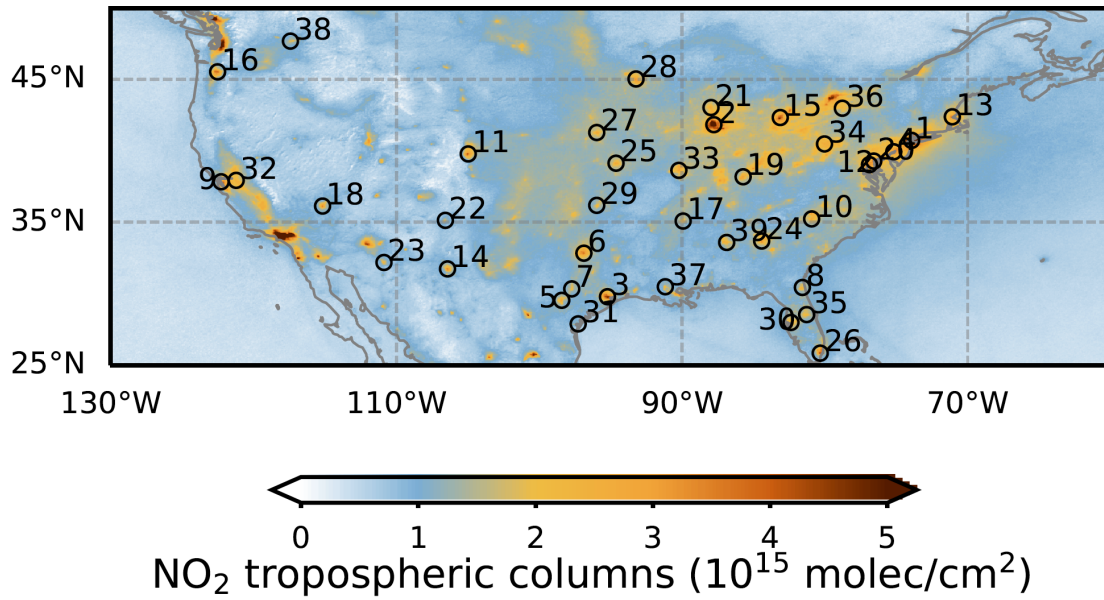
Figure S2: Geographic distribution of investigated cities over the US. Cities are labeled by their IDs (see Table S1). The background is the tropospheric NO₂ vertical column density map averaged from May to September 2019.

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×12 km, (c) NEI NO_x emissions E_{NEI}.

Table S1. Summary of cities investigated in this study.

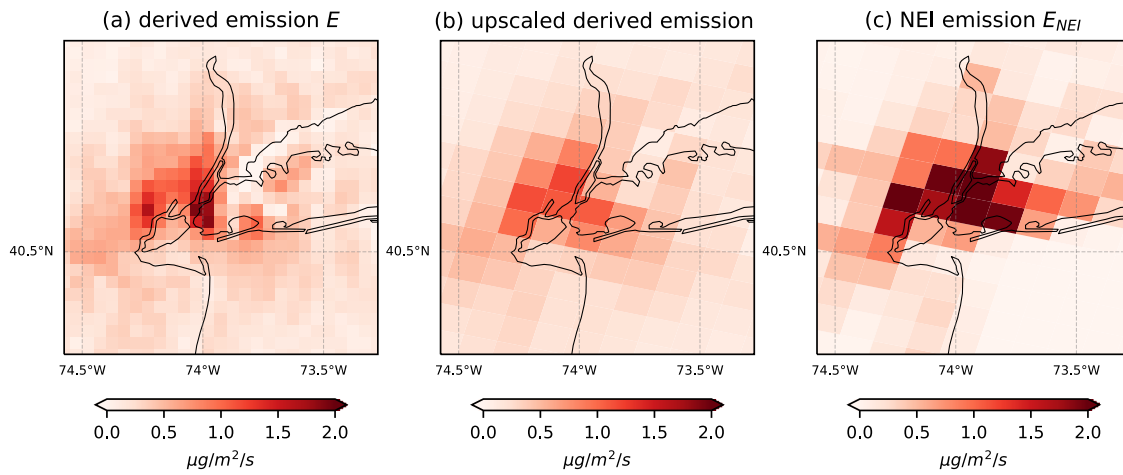| population rank	ID	name	lat	lon	tau (h)	b (10^{23} molec/cm)	significant source	large error	NU-WRF validation	interannual tau*
1	1	NewYork	40.7	-73.9	2.6	0.8	Y		Y	
2		LosAngeles	34.0	-118.3	na	na	Y	Y		Y
3	2	Chicago	41.8	-87.7	2.4	0.8	Y		Y	Y
4	3	Houston	29.8	-95.4	2.0	0.5	Y			
5		Phoenix	33.4	-112.1	na	na	Y	Y		
6	4	Philadelphia	40.0	-75.2	3.6	1.0	Y			
7	5	SanAntonio	29.5	-98.5	2.2	0.6	Y		Y	
8		SanDiego	32.8	-117.1	na	na	Y	Y		Y
9	6	Dallas	32.8	-96.8	2.2	0.7	Y		Y	
10		SanJose	37.3	-121.9	na	na	Y	Y		
11	7	Austin	30.3	-97.8	3.7	0.6	Y			Y
12	8	Jacksonville	30.3	-81.7	2.1	0.7	Y		Y	
13		Columbus	40.0	-83.0	na	na			Y	
14	9	SanFrancisco	37.8	-122.4	1.8	0.4	Y			Y
15	10	Charlotte	35.2	-80.8	1.6	0.7	Y		Y	
16		Indianapolis	39.8	-86.1	na	na			Y	
17		Seattle	47.6	-122.3	na	na	Y	Y		
18	11	Denver	39.7	-105.0	1.6	0.1	Y			
19	12	Washington	38.9	-77.0	2.7	0.8	Y		Y	
20	13	Boston	42.3	-71.1	2.2	0.7	Y		Y	Y
21	14	ElPaso	31.8	-106.4	1.8	0.3	Y		Y	
22	15	Detroit	42.4	-83.1	2.0	0.9	Y		Y	
23		Nashville	36.2	-86.8	na	na				
24		Portland	45.5	-122.7	na	na	Y	Y		
25	16	Memphis	35.1	-90.0	1.9	0.7	Y			
26		OklahomaCity	35.5	-97.5	na	na			Y	Y
27	17	LasVegas	36.2	-115.2	1.8	0.4	Y			
28	18	Louisville	38.3	-85.8	1.5	0.9	Y			
29	19	Baltimore	39.3	-76.6	2.1	0.8	Y		Y	
30	20	Milwaukee	43.1	-88.0	2.0	0.9	Y		Y	
31	21	Albuquerque	35.1	-106.6	1.8	0.3	Y		Y	
32	22	Tucson	32.3	-111.0	2.4	0.3	Y			
33		Fresno	36.8	-119.8	na	na	Y	Y		
34		Sacramento	38.6	-121.5	na	na	Y	Y		Y
35	23	Atlanta	33.8	-84.4	2.6	0.6	Y		Y	
36	24	KansasCity	39.1	-94.6	1.9	0.8	Y		Y	Y

#	#	City	Lat	Lon						
37	25	Miami	25.8	-80.2	2.1	0.5	Y			
38		Raleigh	35.8	-78.6	na	na			Y	
39	26	Omaha	41.3	-96.0	1.9	1.0	Y		Y	
40	27	Minneapolis	45.0	-93.3	1.8	0.7	Y		Y	
41	28	Tulsa	36.1	-95.9	2.0	0.6	Y			Y
42	29	Tampa	28.0	-82.5	2.1	0.6	Y			Y
43	30	NewOrleans	29.9	-90.1	2.5	0.6	Y			
44		Wichita	37.7	-97.3	na	na				
45		Cleveland	41.5	-81.7	na	na			Y	
46		Bakersfield	35.3	-119.0	na	na	Y	Y		
47	31	CorpusChristi	27.7	-97.4	4.3	0.3	Y			
48		Lexington	38.0	-84.5	na	na				
49		Stockton	38.0	-121.3	na	na	Y	Y		Y
50	32	St.Louis	38.6	-90.2	1.8	0.6	Y		Y	
51	33	Cincinnati	39.1	-84.5	2.4	0.8	Y		Y	Y
52	34	Pittsburgh	40.4	-80.0	2.5	0.7	Y		Y	
53		Greensboro	36.1	-79.8	na	na				
54		Lincoln	40.8	-96.7	na	na				
55	35	Orlando	28.5	-81.4	2.3	0.7	Y		Y	
56		Toledo	41.7	-83.6	na	na			Y	
57		FortWayne	41.1	-85.1	na	na			Y	
58		Laredo	27.6	-99.5	na	na				
59	36	Buffalo	42.9	-78.9	6.8	0.7	Y		Y	
60		Lubbock	33.6	-101.9	na	na				
61		Reno	39.5	-119.8	na	na	Y	Y		
62		Norfolk	36.9	-76.2	na	na			Y	
63		Boise	43.6	-116.2	na	na	Y	Y		
64		Richmond	37.5	-77.5	na	na			Y	
65	37	BatonRouge	30.4	-91.1	2.8	0.4	Y		Y	
66	38	Spokane	47.7	-117.4	2.0	0.5	Y			
67		Modesto	37.6	-121.0	na	na	Y	Y		Y
68	39	Birmingham	33.5	-86.8	2.2	0.7	Y			
69		Fayetteville	35.1	-79.0	na	na			Y	
70		Montgomery	32.3	-86.3	na	na				

[*]Cities have valid single-year NO_x lifetimes for all four individual years from 2018 to 2021. Single-year NO_x lifetimes are inferred from TROPOMI NO_2 VCDs, averaged from May to September for an individual year.