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Supplement of

Estimating background contributions and US anthropogenic enhancements to maximum ozone concentrations in the northern US

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S1. Additional features of ODV time series in the northeastern states

The manuscript briefly described some consistent general features of the ODV time series and the corresponding fits of Equation 1 to ODVs from selected groups of sites in the northeastern U.S. that guided the analysis. Here some additional features of interest are briefly discussed:

- New York currently has one non-attainment area. In addition to the New York-N. New Jersey-Long Island, NY-NJ-CT moderate non-attainment area with a population of more than 20 million, Chautauqua County (Jamestown), NY was once a marginal non-attainment area with a population of less than 100,000. In Figure S4 the two sites in this latter non-attainment area are highlighted in purple; the ODVs from these two sites do not differ markedly from the other upwind sites on the western border of the state. In this analysis ODVs from all of the upwind sites are considered together.
- Sites in the New York urban area and regions downwind with over-water transport paths from that urban area have recorded the largest observed ODVs. Consistent with this identification, Vermont, the only state with neither major urban areas nor an over-ocean transport path from the New York City area, records the smallest maximum ODVs (see Table 3 and Figure S10).
- Although some sites in the New York urban area record high ODVs, some other sites in central urban areas in the northeast U.S. record the lowest ODVs (e.g., New Haven, Connecticut; Providence, Rhode Island, particularly before 2000; and Boston Massachusetts; see Figures S6-S8, respectively). This behavior is consistent with fresh NOx emissions in urban areas reducing the ozone concentrations in air masses transported into those areas. This is evidently a very localized phenomenon, as the suburban sites adjacent to Boston (Figure S8) exhibit ODVs similar to coastal sites in the state.
- The farthest downwind coastal monitoring site in northeast Maine (Figure S11) records significantly lower ODVs than other coastal sites, suggesting that ozone concentrations may decrease during transport due to dilution and/or ozone loss to surface deposition.
- Connecticut had much higher maximum ODVs than any other state before 1985 (all points above 140 ppb in Figure 3); their cause is unknown. Since 1985 Connecticut ODVs have been similar to those of neighboring states until 2013.
- Through the measurement record, the differences between maximum and minimum ODVs have decreased, both within individual states and throughout the entire region.
- There is one monitoring site at a relatively elevated location in the northeastern U.S. - Mt. Washington in New Hampshire at 1.9 km above sea level (asl). Although the ODV record at this site (Figure S9) is generally not higher than others recorded in New Hampshire, the fit to Equation 1 shows a much smaller decrease than seen at any other site in the entire region. These ODVs followed a temporal evolution different from any of the other sites in the region (see curves in Figure 7 and parameters in Table 2). The A value (8 ± 8 ppb) is much smaller than that of any other selected group of sites, and the U.S. background ODV (y_0 = 66 ± 7 ppb) is significantly higher than the common y_0 value of 45.8 ± 1.7 ppb derived for the entire northeastern U.S. This difference is attributed to the vertical gradient of ozone over the northeastern U.S. Ozone concentrations in the free troposphere increase with altitude (e.g., see Figure 2 of Fehsenfeld et al., 2006), and it is these higher altitude air parcels that impact Mt. Washington. The y_0 value derived at Mt. Washington is in reasonable accord with the average ozone concentrations measured over the eastern U.S. by the MOZAIC program in the years near 2000 (Fehsenfeld et al., 2006). The U.S. anthropogenic enhancement of the ODVs (i.e., the A value) in the free troposphere observations at Mt. Washington is much smaller than the enhancements seen at the other sites, which are all located within the planetary boundary layer. Note that the temporal evolution described by the parameters in Table 2 and illustrated in Figures 7 and S9 implies that the Mt. Washington summit site will soon record the highest ODVs in New Hampshire and higher than other sites in the northeastern U.S. outside of and immediately downwind from the New York City urban area; in 2017 Mt. Washington did report the largest ODV in New Hampshire.
- The Cadillac Mountain coastal site at in Maine is at a somewhat elevated location (0.47 km asl). In contrast to Mt. Washington, the Cadillac Mountain ODVs (Figure S11) are generally similar to, although slightly higher than others recorded at the southwest Maine coastal sites. Evidently Cadillac Mountain receives primarily boundary layer air masses.

Reference

Fehsenfeld, F. C., et al.: International Consortium for Atmospheric Research on Transport and Transformation (ICARTT): North America to Europe—Overview of the 2004 summer field study, J. Geophys. Res. 2006, 111, D23S01, doi:10.1029/2006JD007829, 2006.

Table S1. Summary of data set of ODVs for eight northeastern U.S. states.

State	sites	years	ODV min (ppb)	ODV max (ppb)
Pacific Northwest States				
Washington	31	1979-2017	41	88
Oregon	16	1976-2017	50	91
Idaho	8	1995-2017	56	77
Rural Western States				
Montana	10	1979-2017	52	64
North Dakota	14	1982-2017	54	70
South Dakota	10	1990-2017	54	70
Yellowstone NP	1	1999-2017	60	67
Midwestern States				
Minnesota	16	1975-2017	55	92
Wisconsin	83	1975-2017	57	135
Michigan	47	1975-2017	57	117
Northeastern States				
Connecticut	20	1976-2017	67	169
Maine	32	1979-2017	50	117
Massachusetts	45	1976-2017	56	121
New Hampshire	31	1975-2017	54	118
New Jersey	26	1975-2017	62	132
New York	59	1973-2017	49	129
Rhode Island	4	1978-2017	66	130
Vermont	5	1978-2017	60	96

Table S2. Results of the intercepts of the linear regressions illustrated in Figures S12-S19 with the 1:1 lines.

State/sites	U.S. background ODV (ppb)		r^2	years fit
	OLR*	RMA*		
New York/rural upwind	43	25	0.75	2000-2017
New Jersey/all sites	52	31	0.83	2000-2017
Rhode Island/all sites	62	58	0.84	2000-2017
Massachusetts/Boston	47	45	0.75	2000-2017
Massachusetts/suburban	50	42	0.82	2000-2017
Massachusetts/coastal	53	48	0.89	2000-2017
New Hampshire/coastal	55	53	0.77	2000-2017
New Hampshire/northwest	49	46	0.60	2000-2017
Vermont /all sites	51	49	0.79	2000-2017
Maine/interior	39	28	0.44	2000-2017
Maine/NE coast	36	34	0.89	1991-2017
Maine/SW coast	56	53	0.77	2000-2017
Maine/Cadillac Mtn.	48	40	0.85	2000-2017

*OLR = standard linear regression; RMA = reduced major axis regression

Table S3. Results from linear fits to early years of three ODV data sets

Data set	Year 2000 value (ppb)		slope (ppb/yr)	
	original	corrected	original	corrected
NYC urban maximum	108.6 ± 3.8	110.1 ± 3.8	-2.16 ± 0.38	-2.91 ± 0.38
Vermont	78.2 ± 1.8	79.0 ± 1.8	-1.07 ± 0.18	-1.44 ± 0.18
Maine NE coast	70.6 ± 1.5	69.8 ± 1.5	-0.89 ± 0.13	-1.05 ± 0.13

Table S4. Results of iterative, non-linear regression analysis.

τ (years)	26.0 ± 6.0
y_0 (ppb)	41.8 ± 3.0
State	A (ppb)
Maine	51 ± 10
Massachusetts	56 ± 10
New Hampshire	46 ± 10
New Jersey	66 ± 10
New York	61 ± 10
Rhode Island	55 ± 10
Vermont	39 ± 10

Table S5. Estimates of U.S. background ODVs in five regions of the U.S. Units in ppb. The first column of results are based on observations, including those derived here plus the result of Parrish et al. (2017) for the Los Angeles urban area. The last three columns are model results as described in the text.

Region	Obs. based	Fiore et al. AM3	Fiore et al. GC	Emery et al. GC
Montana	55.4 ± 2.2	55 ± 2	45 ± 3	50 ± 5
North Dakota	59.3 ± 2.7	51 ± 2	44 ± 2	45 ± 5
South Dakota	61.5 ± 3.8	52 ± 2	46 ± 4	47 ± 5
Northeastern U.S.	45.8 ± 1.7	49 ± 2	37 ± 3	35 ± 5
South Coast Air Basin	62.0 ± 1.9	55 ± 2	48 ± 4	57 ± 5

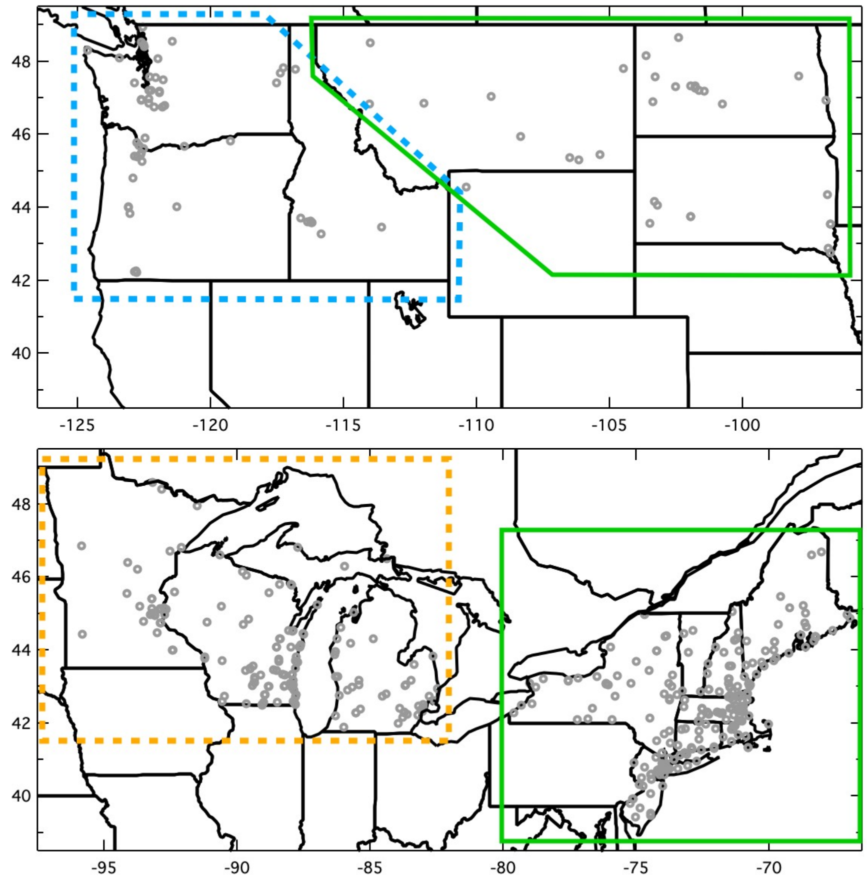

Figure S1: Maps of the northern U.S. with all ozone monitoring sites indicated by grey circles. The colored lines indicate four regions: the eight northeastern states and the three rural western states examined in detail (green solid), three Pacific Northwest states (blue dashed), and three midwestern states (orange dashed).

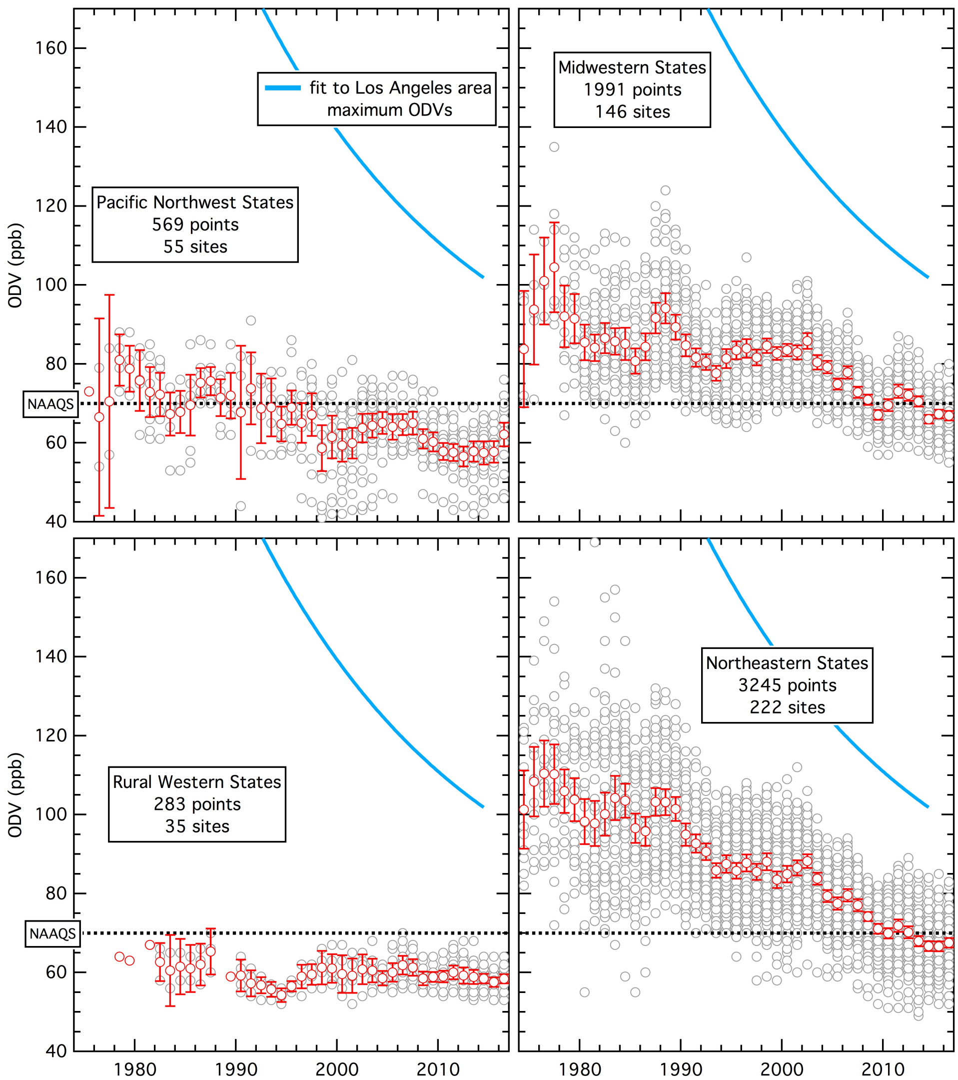

Figure S2: Time series of all ODVs (grey symbols) reported from all monitoring sites in the four northern U.S. regions shown in Figure S1. The numbers of monitoring sites and reported ODVs are annotated for each region. The red symbols give the averages and 2-σ confidence limits for all ODVs reported in each year. For comparison, the blue curve in each panel indicates a fit to the time history of the maximum ODVs recorded in the Los Angeles urban area (Parrish et al., 2017). The dotted line indicates the 2015 NAAQS of 70 ppb.

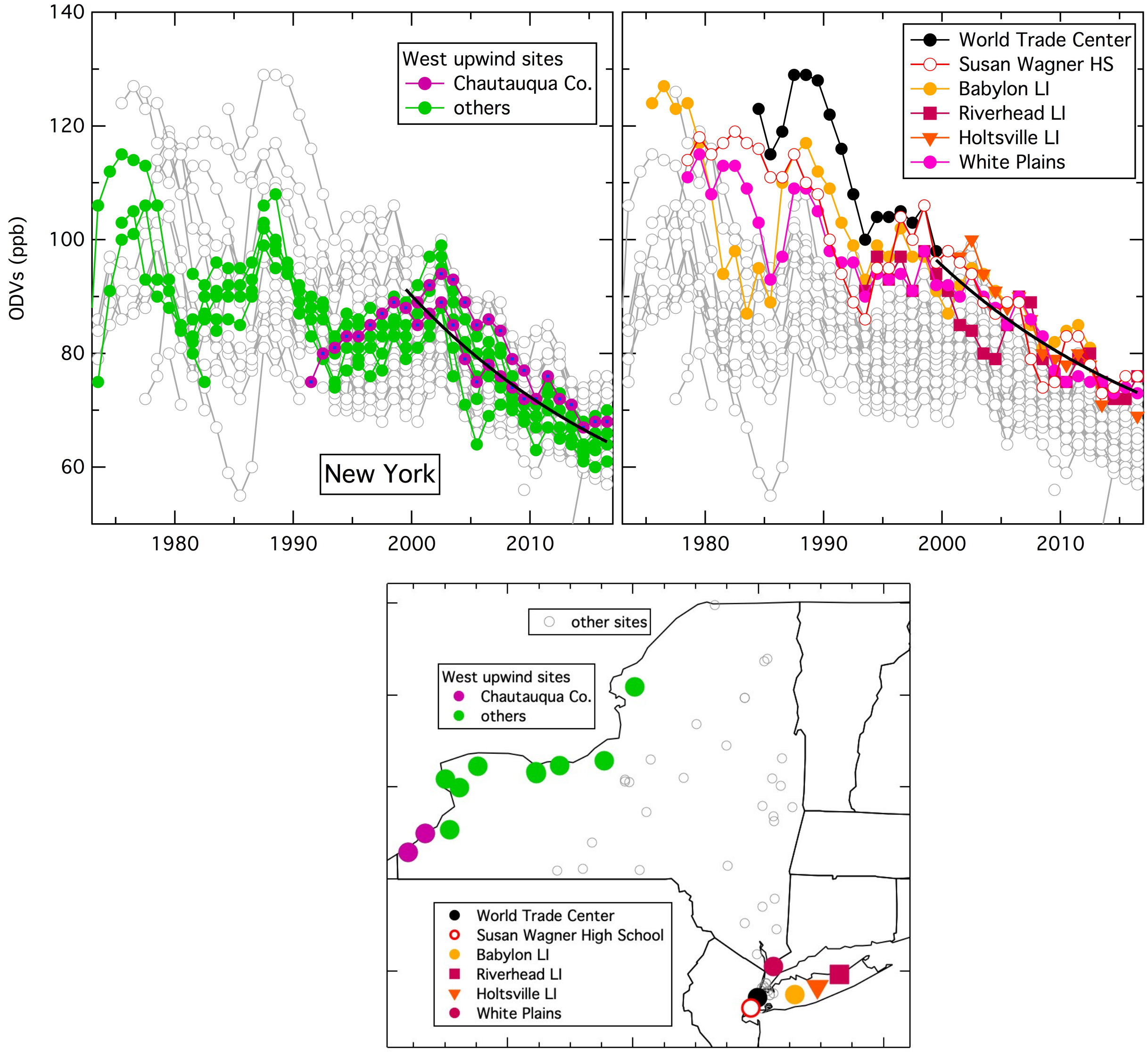

Figure S3. Temporal evolution of the ODVs in New York and map of all monitoring sites reporting ODVs. The upper panels highlight the sites on the western border (left) and the urban New York city area sites with the largest ODVs (right); these sites are also highlighted on the map with corresponding symbols and color-coding. The sites in the Chautauqua Co., NY marginal nonattainment area are highlighted in purple. Black curves show fits of Equation 1 for 2000-2017 to the color-coded points in the upper panels.

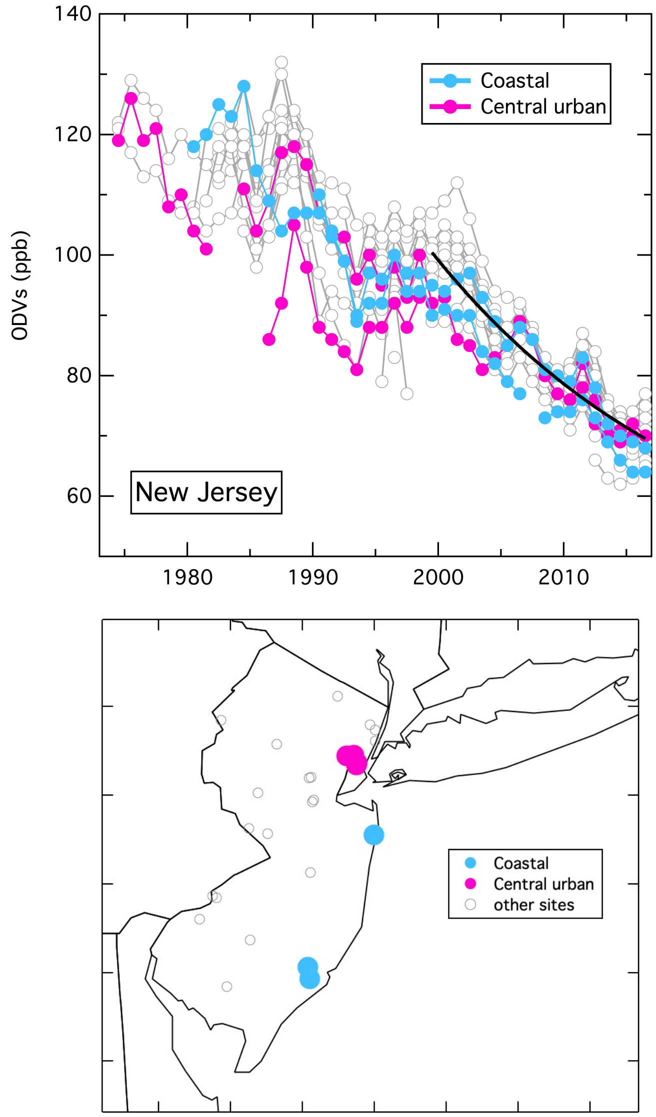

Figure S4. Temporal evolution of the ODVs in New Jersey and map of all monitoring sites reporting ODVs. The two panels highlight the coastal and central urban sites with corresponding symbols and color-coding. A fit of Equation 1 to ODVs from all sites is shown for 2000-2017.

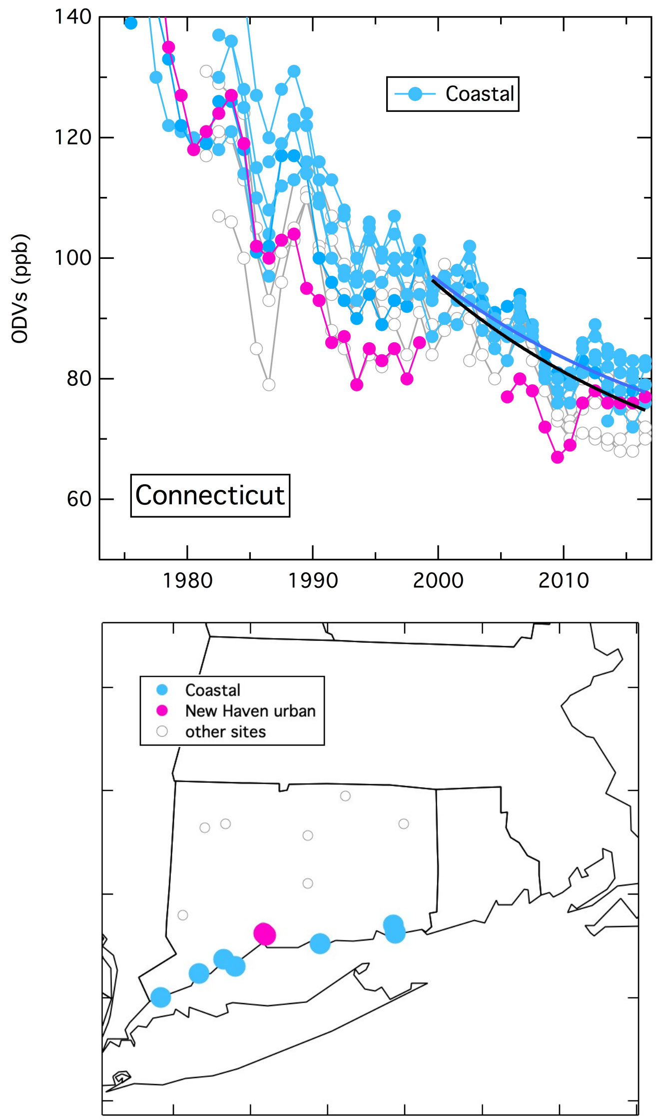

Figure S5. Temporal evolution of the ODVs in Connecticut and map of all monitoring sites reporting ODVs. The two panels highlight the coastal and New Haven urban sites with corresponding symbols and color-coding. Fit of Equation 1 to all ODVs (black curve) and to the coastal sits (colored cureve) are shown for 2000-2017.

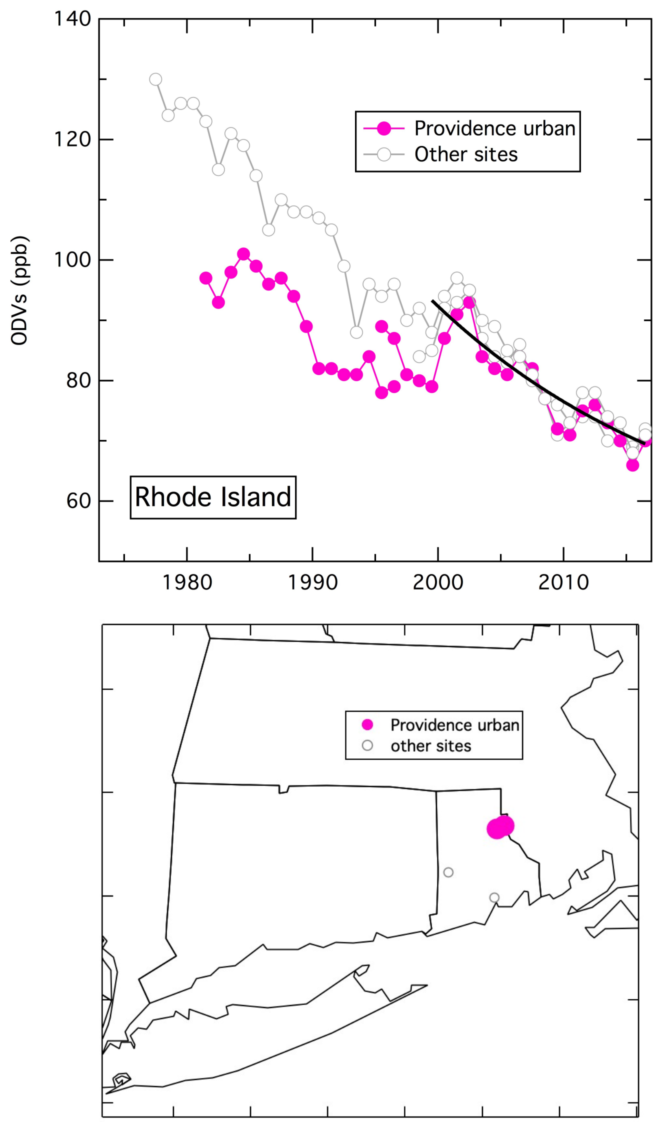

Figure S6. Temporal evolution of the ODVs in Rhode Island and map of all monitoring sites reporting ODVs. The two panels highlight the rural and urban Providence sites with corresponding symbols and color-coding. A fit of Equation 1 to all ODVs is shown for 2000-2017.

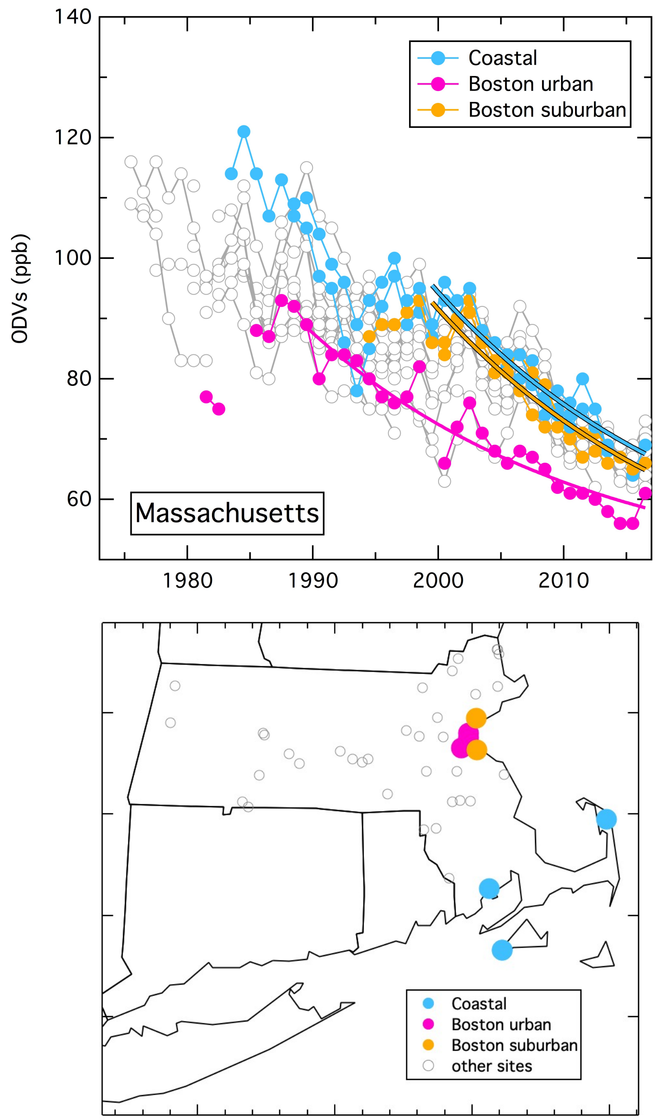

Figure S7. Temporal evolution of the ODVs in Massachusetts and map of all monitoring sites reporting ODVs. The two panels highlight the coastal and Boston urban and suburban sites with corresponding symbols and color-coding. Fits of Equation 1 to the selected ODVs are shown for 2000-2017 for the coastal and suburban sites, and for 1990-2017 for the Boston urban sites.

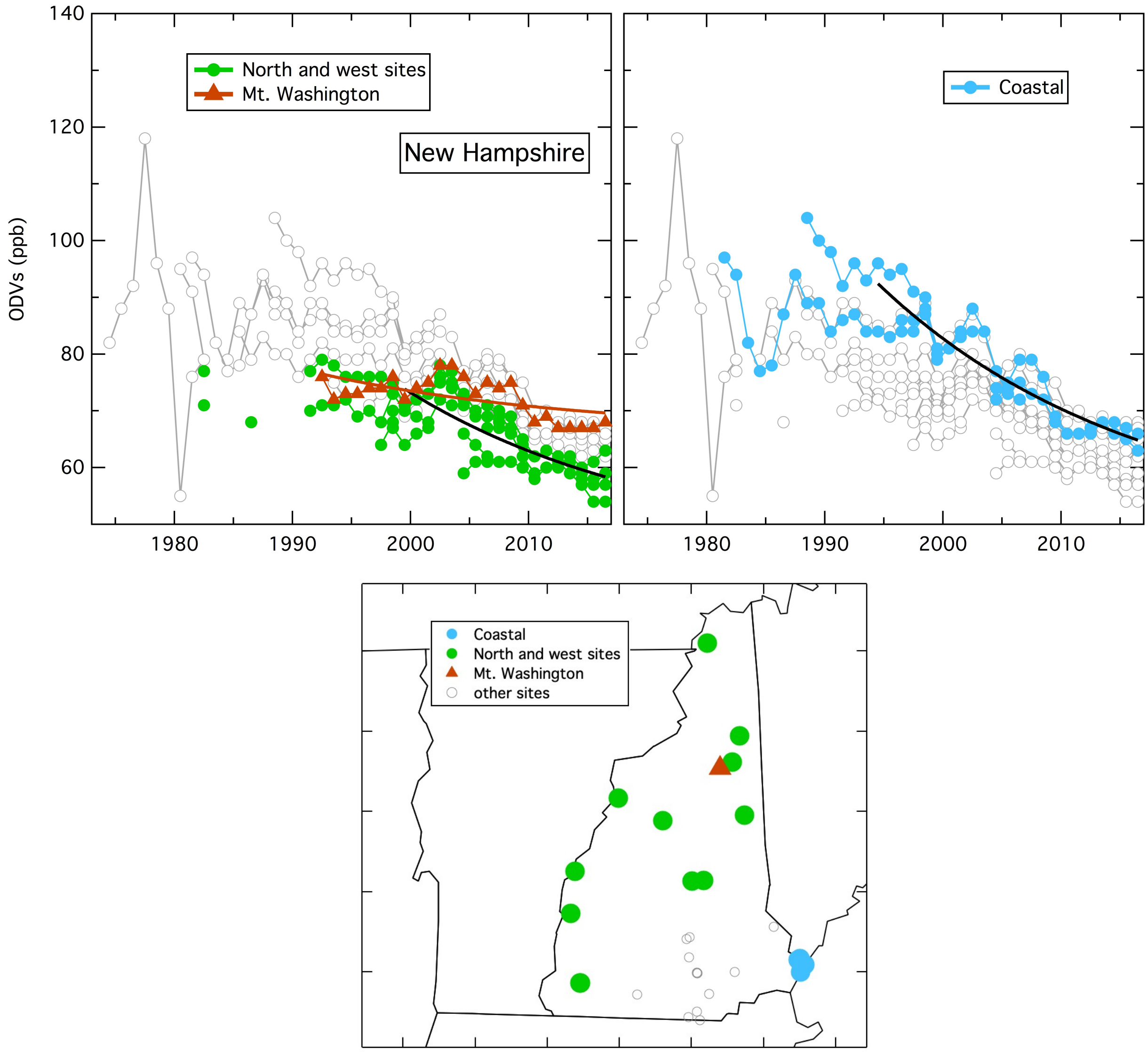

Figure S8. Temporal evolution of the ODVs in New Hampshite and map of all monitoring sites reporting ODVs. The two upper panels highlight the coastal (including Kittery, ME) and the rural north and west sites, including Mt. Washington. The lower panel shows all sites with corresponding symbols and color-coding. Fits of Equation 1 to the selected ODVs are shown for 1995-2017 for the coastal sites, 2000-2017 for the north and west sites, and the full data range (1993-2017) for Mt. Washington.

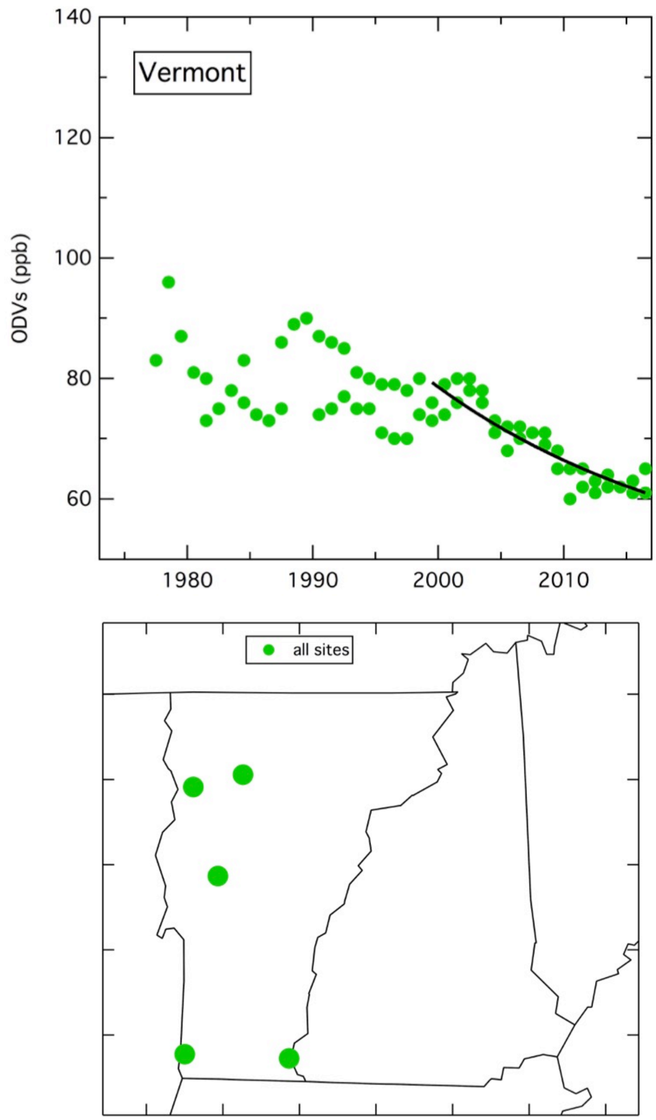

Figure S9. Temporal evolution of the ODVs in Vermont and map of all monitoring sites reporting ODVs. Fit of Equation 1 to all sites is shown for 2000-2017.

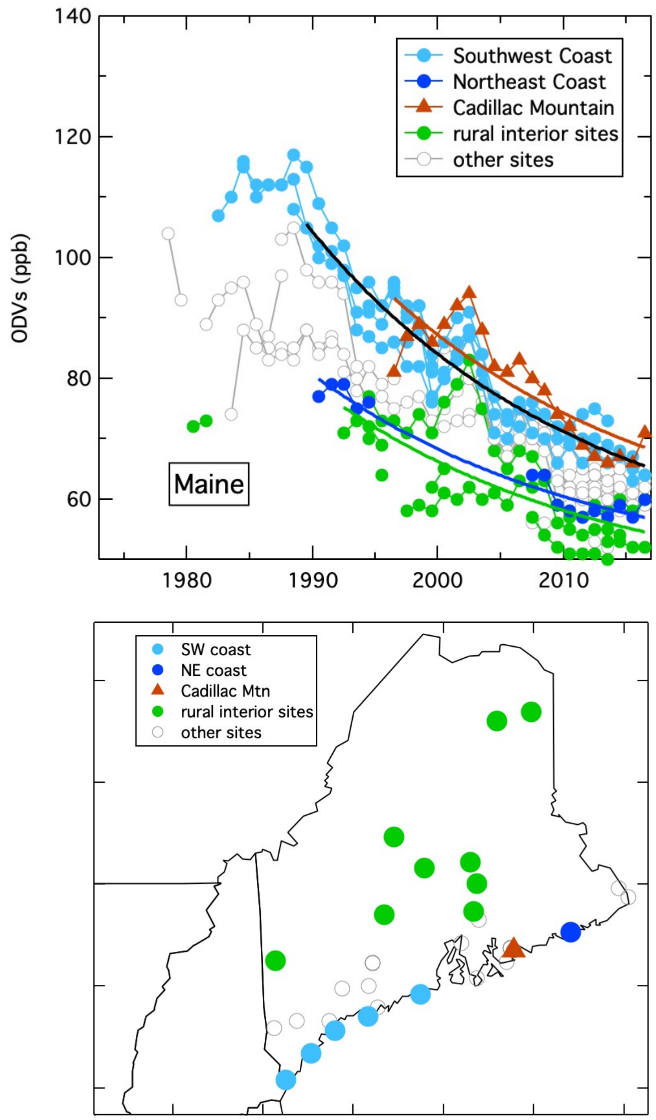

Figure S10. Temporal evolution of the ODVs in Maine and map of all monitoring sites reporting ODVs. Fits of Equation 1 to four sets of sites are shown.

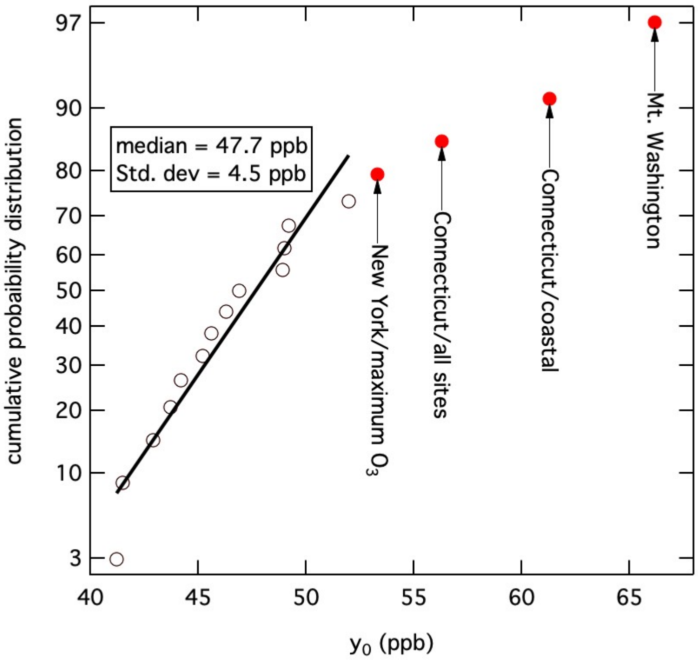

Figure S11. Cumulative probability distribution plot of the y_0 determinations listed in Table 2 of the manuscript. The ordinate scale of the figure is designed so that a normal distribution will lie on a straight line. The line shows a linear regression fit to the open points, which defines a normal distribution with the median and standard deviation annotated in the figure. The four points at the higher y_0 values correspond to the colored curves in Figure 7 of the paper.

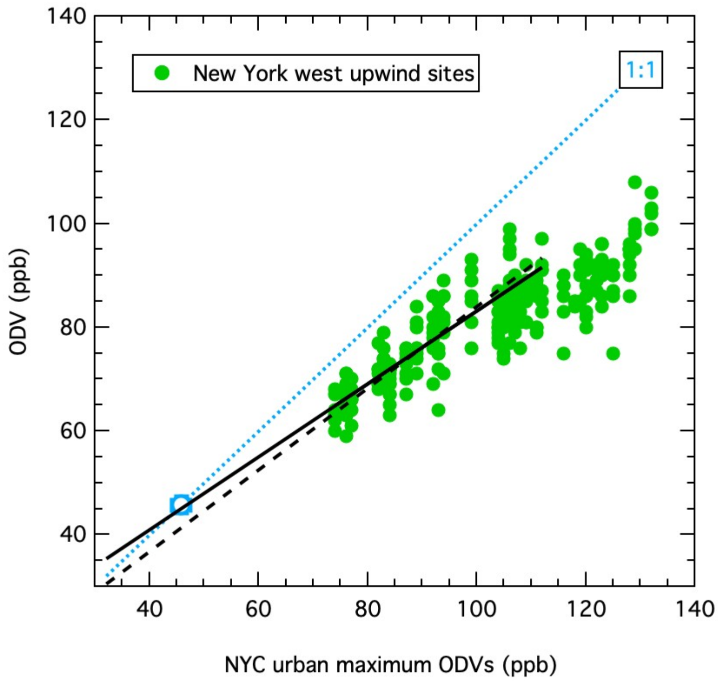

Figure S12. Correlation between the ODVs at the New York west upwind sites and the maximum ODVs recorded in the New York city urban area. Black lines show ordinary linear regression (solid) and reduced major axis regression with equal weighting (dashed) fits of the two correlated data sets for the ODVs recorded in 2000-2017. The blue symbol shows the mean U.S. background ODV derived from the exponential fits to the ODV time series and the blue dotted line indicates the 1:1 relationship.

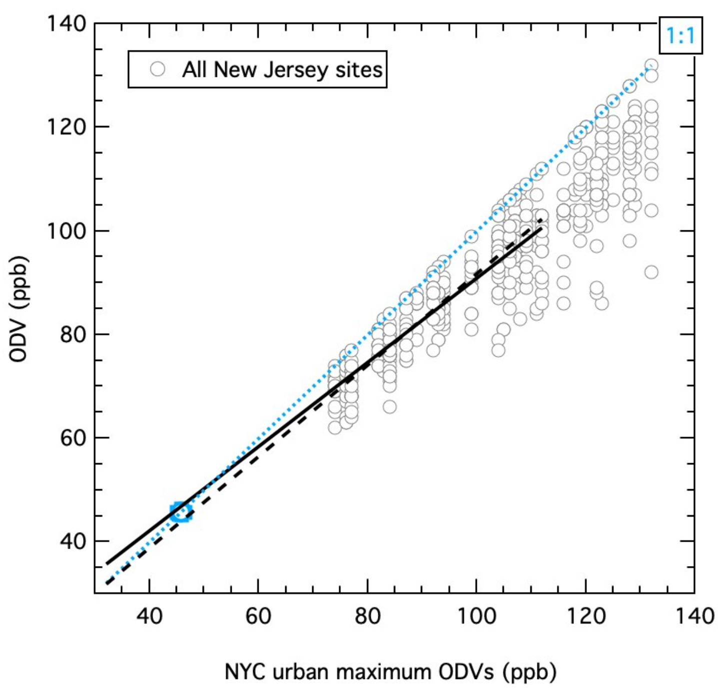

Figure S13. Correlation between the ODVs recorded at all New Jersey sites and the maximum ODVs recorded in the New York city urban area in 2000-2017. Format is the same as in Figure S12.

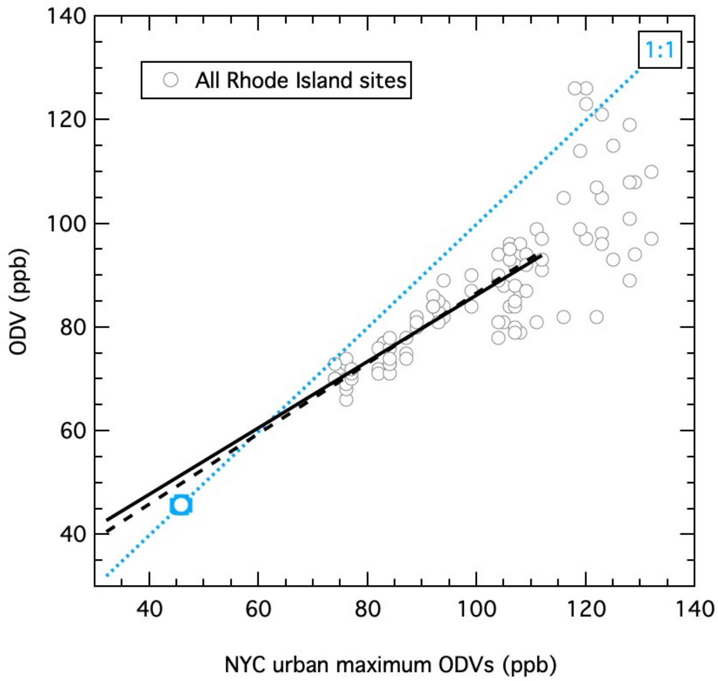

Figure S14. Correlation between the ODVs at all Rhode Island sites and the maximum ODVs recorded in the New York city urban area in 2000-2017. Format is the same as in Figure S12.

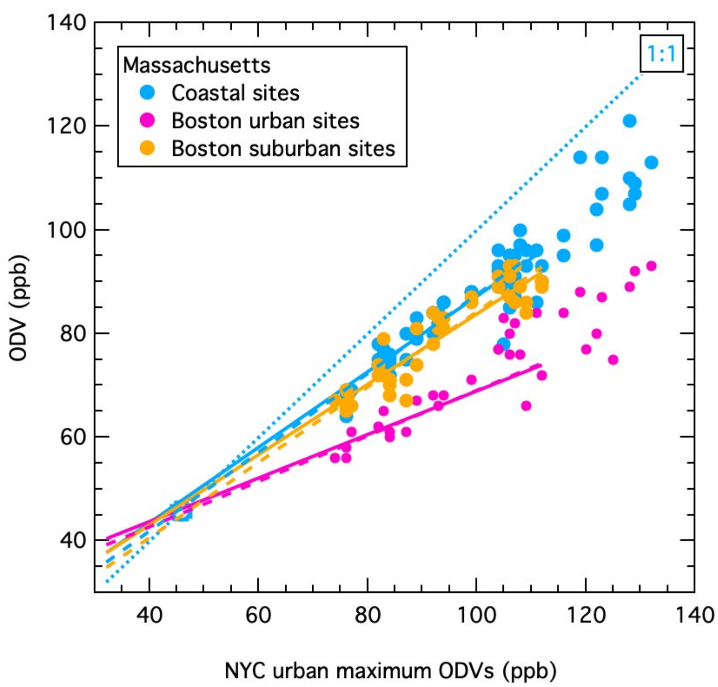

Figure S15. Correlation between the ODVs at three sets of Massachusetts sites and the maximum ODVs recorded in the New York city urban area in 2000-2017. Format is the same as in Figure S12.

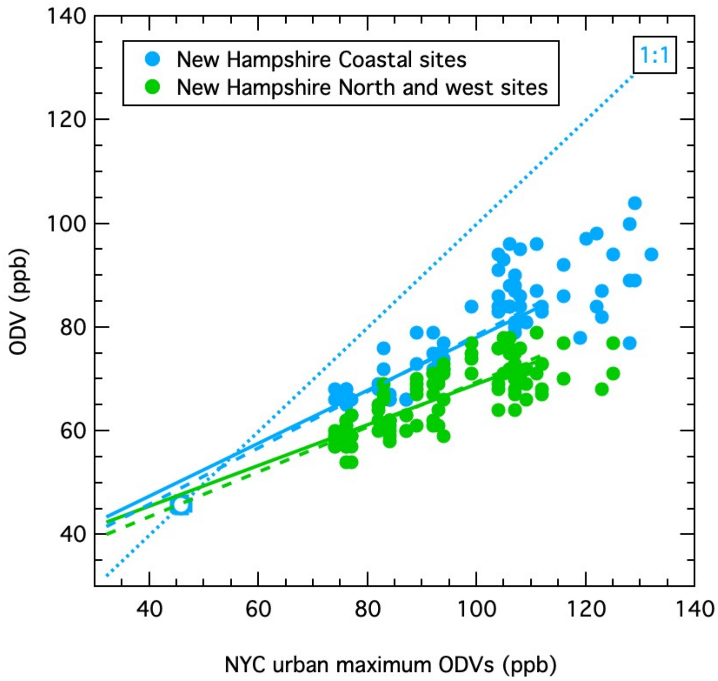

Figure S16. Correlation between the ODVs at two sets of New Hampshire sites and the maximum ODVs recorded in the New York city urban area in 2000-2017. Format is the same as in Figure S12.

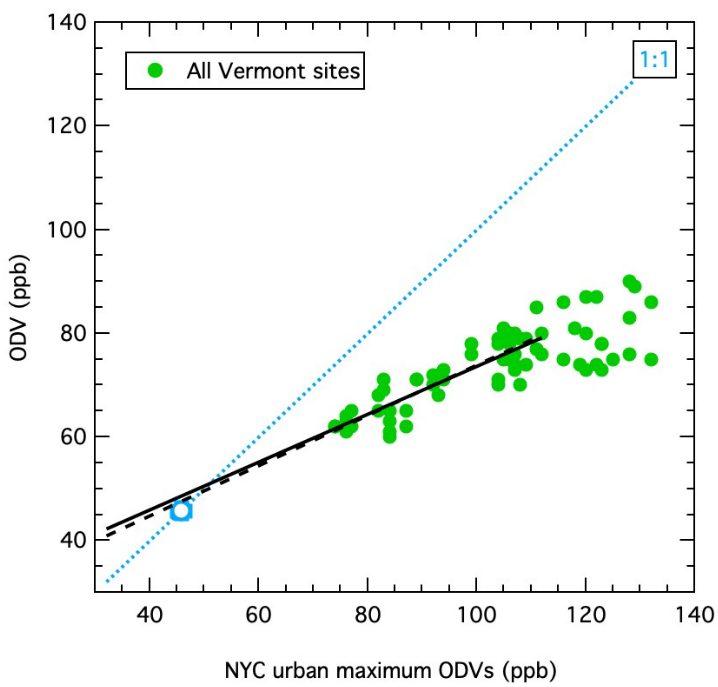

Figure S17. Correlation between the ODVs at all Vermont sites and the maximum ODVs recorded in the New York city urban area in 2000-2017. Format is the same as in Figure S12.

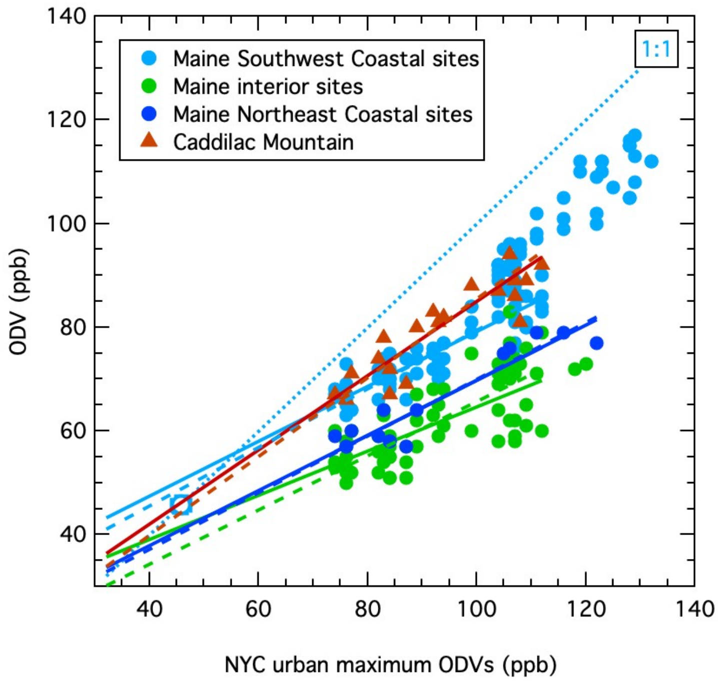

Figure S18. Correlation between the ODVs at four sets of Maine sites and the maximum ODVs recorded in the New York city urban area in 2000-2017. Format is the same as in Figure S12, except for the Maine northeast coastal sites all ODVs recorded (1991-2017) are included in the fit.

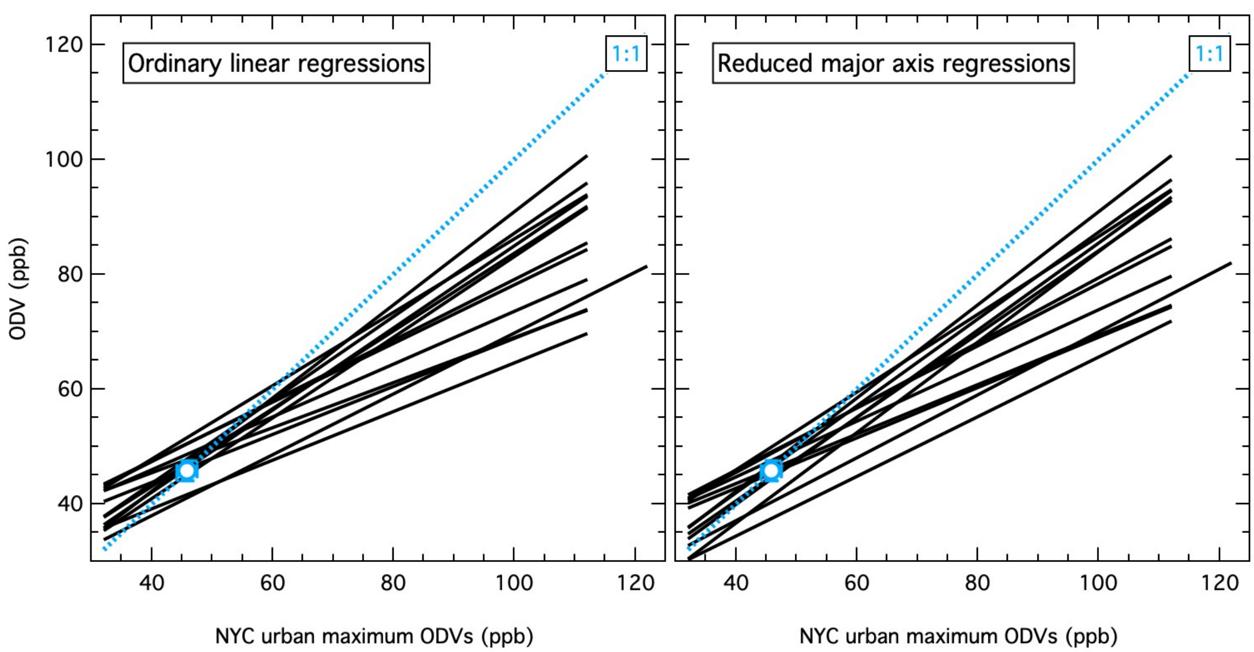

Figure S19. Correlations between the ODVs at 13 sets of northeastern U.S. sites and the maximum ODVs recorded in the New York city urban area in 2000-2017. All correlation lines from Figures S12-S18 are included. The blue symbols show the mean U.S. background ODV derived from the exponential fits to the ODV time series and the blue dotted lines indicate the 1:1 relationship. Table S2 summarizes the results of all fits.

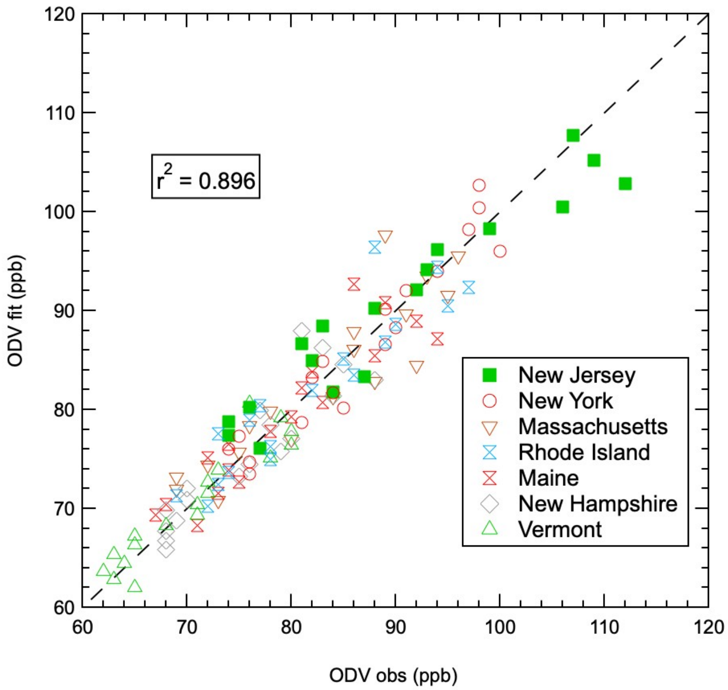

Figure S20. Comparison of state maxima ODV from 9 parameter iterative regression fit with those from the original values derived from observations.