

Supplemental Material, Table 1. Bias and absolute precision values for the post-1999 and pre-1999 PM<sub>2.5</sub> models from cross-validation.

Grouping		Post-1999 model			Pre-1999 model <sup>a</sup>		
		N <sup>b</sup>	Bias <sup>c</sup> (µg/m <sup>3</sup> )	RMSPE <sup>d</sup> (µg/m <sup>3</sup> )	N <sup>b</sup>	Bias <sup>c</sup> (µg/m <sup>3</sup> )	RMSPE <sup>d</sup> (µg/m <sup>3</sup> )
By state (1988-2002)	CT	449	-0.3	2.1	79	0.1	2.3
	DE	268	-0.1	1.8	59	-0.5	2.1
	ME	664	-0.5	2.3	132	-0.8	2.0
	MD	539	-0.2	1.6	46	-1.6	2.6
	MA	773	-0.6	2.1	164	-0.2	2.1
	MI	1,054	0.0	2.3	162	0.2	1.9
	NH	355	-0.3	3.1	79	-1.5	4.6
	NJ	765	0.5	2.4	151	-0.2	3.2
	NY	1,758	-0.1	2.3	126	0.5	3.2
	OH	1,713	-0.3	2.2	298	-1.2	2.7
	PA	1,575	0.0	2.2	245	0.1	2.7
	RI	282	0.1	2.0	52	0.5	1.7
	VT	249	-0.3	2.6	49	0.5	2.0
By season (across study region)	Winter	2,411	-0.3	2.6	310	-0.7	3.1
	Spring	2,523	-0.2	2.0	360	-0.2	2.3
	Summer	2,699	-0.1	2.4	451	-0.2	2.9
	Fall	2,811	-0.1	2.0	521	-0.4	2.4
By quartiles of urban land use	1	2,622	-0.1	2.1	409	-0.4	2.7
	2	2,618	-0.2	2.1	411	-0.5	2.6
	3	2,608	-0.2	2.3	414	-0.5	2.6
	4	2,596	-0.1	2.4	408	-0.1	2.8
By quartiles of block group population density	1	2,593	-0.1	2.0	410	0.0	2.3
	2	2,619	-0.4	2.6	408	-0.9	3.1
	3	2,621	0.0	2.2	414	-0.3	2.6
	4	2,611	-0.1	2.1	410	-0.1	2.7
By monitoring network	AQS <sup>e</sup>	9,856	-0.2	2.2	1,575	-0.4	2.6
	IMPROVE <sup>f</sup>	494	0.5	1.9	49	0.6	2.3
	CASTNet <sup>f</sup>	94	0.0	3.2	18	1.2	4.8
By monitoring objective <sup>g</sup>	Unknown (Other)	588	0.4	2.2	67	0.8	3.2
	Unknown (AQS)	1,169	-0.2	1.9	135	0.3	2.2
	Population Exposure	6,264	-0.1	2.1	973	-0.3	2.6

	Highest Concentration	1,177	-0.1	3.3	248	-1.6	3.5
	General/Background	429	0.0	2.0	81	-0.2	2.0
	Other	354	0.4	1.9	76	0.6	1.9
	Regional Transport	196	-0.3	1.9	26	-0.6	1.5
	Upwind Background	134	0.2	1.6	13	0.3	1.5
	Extreme Downwind	48	0.0	1.4	11	1.0	1.3
	Maximum Ozone Concentration	44	-0.3	2.2	12	-0.4	1.8
	Source Oriented	41	-0.6	2.0	NA <sup>h</sup>	NA <sup>h</sup>	NA <sup>h</sup>
Across all observations		10,444	-0.2	2.2	1,642	-0.3	2.7

<sup>a</sup>Using 1999 data for cross-validation, excluding four observations from one site in New York as outliers.

<sup>b</sup>"N" is number of pairs of measurements and cross-validation predictions.

<sup>c</sup>Mean prediction error, where prediction errors are cross-validation predictions minus measurements.

<sup>d</sup>RMSPE is root mean squared prediction error, describing the absolute precision of the model predictions.

<sup>e</sup>From the U.S. EPA's Air Quality System (AQS) monitoring network.

<sup>f</sup>IMPROVE is Interagency Monitoring of Protected Visual Environments, CASTNet is Clean Air Status and Trends Network.

<sup>g</sup>Defined for sites in the U.S. EPA Air Quality System (AQS) network only.

<sup>h</sup>"NA" indicates that no data in this category were available.