Electronic Supplementary Material (ESI) for Environmental Science: Processes & Impacts.

Supporting Information for: Atmospheric Cycling of Indium in the Northeastern United States

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8 pages, 5 figures, 2 tables

Methods: Correlation of indium with other metals in PM_3

Correlations were determined between indium concentrations and other metals, using particle-normalized concentrations. After separating by location and wind direction (described in main text), regression lines and their standard deviations were generated by a Monte Carlo technique, whereby random subsets of the data were selected and linear regressions were calculated for each subset. The mean and standard deviation of these subsets were plotted as the mean regression line for the complete data set and its $\pm 1\sigma$ uncertainty.

The Kolmogorov-Smirnov test was used to obtain a statistical measure of how much the two wind directions differ with respect to their In:M ratios. The Kolmogorov-Smirnov test is a nonparametric test to determine the probability that two data sets of unknown distribution are drawn from the same distribution[1]. Both data sets are plotted as a cumulative distribution function, $S_{N_1}(x)$ and $S_{N_2}(x)$, and the maximum value of the absolute difference between the two distribution functions, D, is determined: $D = \max|S_{N_1}(x) - S_{N_2}(x)|$.

The significance of D (i.e. the probability that the two data sets are the same) can be calculated as

$$Q_{KS}(\lambda) = 2\sum_{j=1}^{\infty}(-1)^{j-1}e^{-2j^2}\lambda^2 \tag{S1}$$

with the limits of $Q_{KS}(0) = 1$ and $Q_{KS}(\inf) = 0$. The significance level of a calculated D can be approximated by:

$$Probability(D > observed) = Q_{KS}([\sqrt{N_e} + 0.12 + 0.11/\sqrt{N_e}]D) \tag{S2}$$

and

$$N_e = \frac{N_1 N_2}{N_1 + N_2} \tag{S3}$$

where N_1 and N_2 are the number of data points in the first and second distribution, respectively. This approximation tends to be good for $N_e \geq 4$, and is asymptotically accurate as N_e gets large[1]. Here $N_e \geq 9$.

Figures and Data Tables

The following figures include a map of potential sources of indium to the atmosphere in the northeastern United States in 1995 (the year the samples were taken), correlations between indium concentrations in Massachusetts and New York, indium's enrichment factor in the atmospheric samples, cumulative distribution functions for In:metal mass ratios in air from the north and air from the west, and indium concentrations in the coarse air particulate fraction. Table S1 includes the raw data for indium concentrations in PM_3 in five locations in the Northeastern United States.

Table S1: Indium atmospheric concentration data for PM_3 in five locations in the northeastern United States.

	Boston		Quabbin		Reading		Brockport		Rochester	
Date	pg/m^3	μg/g	pg/m^3	μg/g	pg/m^3	μg/g	pg/m^3	μg/g	pg/m^3	μg/g
1/3/95	2.15	0.15	1.06	-	1.54	0.16	-	-	0.94	-
2/2/95	2.31	-	1.44	0.36	2.17	0.23	1.32	0.94	3.86	0.40
2/26/95	4.39	0.37	-	-	1.34	0.19	3.83	0.50	5.06	0.38
3/4/95	4.49	0.20	5.29	0.50	2.49	0.10	2.52	0.10	2.40	0.07
3/10/95	-	-	-	-	-	-	6.54	0.87	2.99	0.23
3/16/95	4.85	0.19	2.74	0.08	7.76	0.25	-	-	-	-
4/3/95	2.78	0.15	2.22	0.16	2.06	0.13	2.26	0.11	3.51	0.19
4/15/95	-	-	-	-	1.31	0.11	-	-	-	-
4/27/95	2.42	0.13	2.71	0.15	1.24	0.06	2.55	0.25	2.59	0.17
5/21/95	-	-	-	-	1.84	0.10	-	-	-	-
5/27/95	-	-	-	-	-	-	2.21	0.26	1.20	0.20
6/2/95	1.69	0.10	-	-	-	-	1.13	0.03	3.12	0.13
6/8/95	-	-	-	-	-	-	-	-	0.98	0.07
7/2/95	0.48	0.04	-	-	-	-	0.13	0.02	0.32	0.03
7/26/95	5.04	0.13	-	-	-	-	1.31	0.08	2.45	0.11
8/13/95	0.61	0.10	-	-	-	-	3.19	0.32	5.45	0.61
9/6/95	2.24	0.22	0.79	-	1.02	0.08	1.40	0.06	1.88	0.16
9/30/95	0.71	0.14	0.41	0.06	0.51	0.03	0.94	0.31	0.69	0.07
10/30/95	0.84	0.15	0.70	0.16	0.95	-	0.75	0.58	1.42	0.31
11/23/95	0.80	0.09	1.28	0.09	0.83	0.08	1.08	0.19	2.84	0.30
11/29/95	1.63	0.22	1.30	0.11	0.96	0.10	6.22	1.07	5.21	0.72
12/5/95	2.32	0.15	1.70	0.28	-	-	1.41	0.19	2.85	0.39
12/29/12	1.68	0.11	1.23	0.07	0.90	0.12	1.15	0.07	1.83	0.07

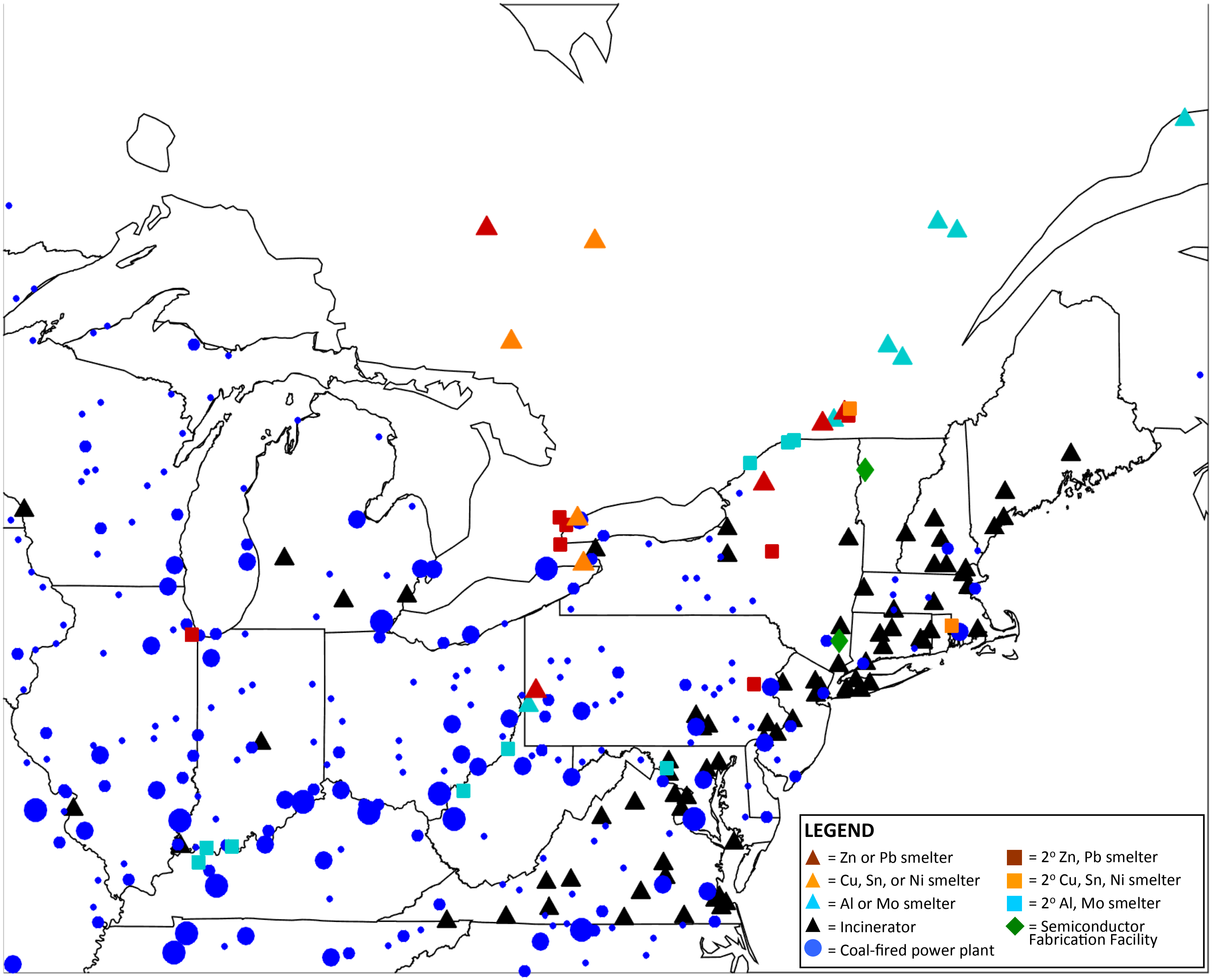

Figure S1: Map of potential sources of indium in the northeastern United States in 1995, enlarged. The size of the circles representing coal-fired power plants roughly denote the scale of their energy production. Note that Canada does not rely heavily on coal-fired power plants; those in operation in 1995 are mapped.

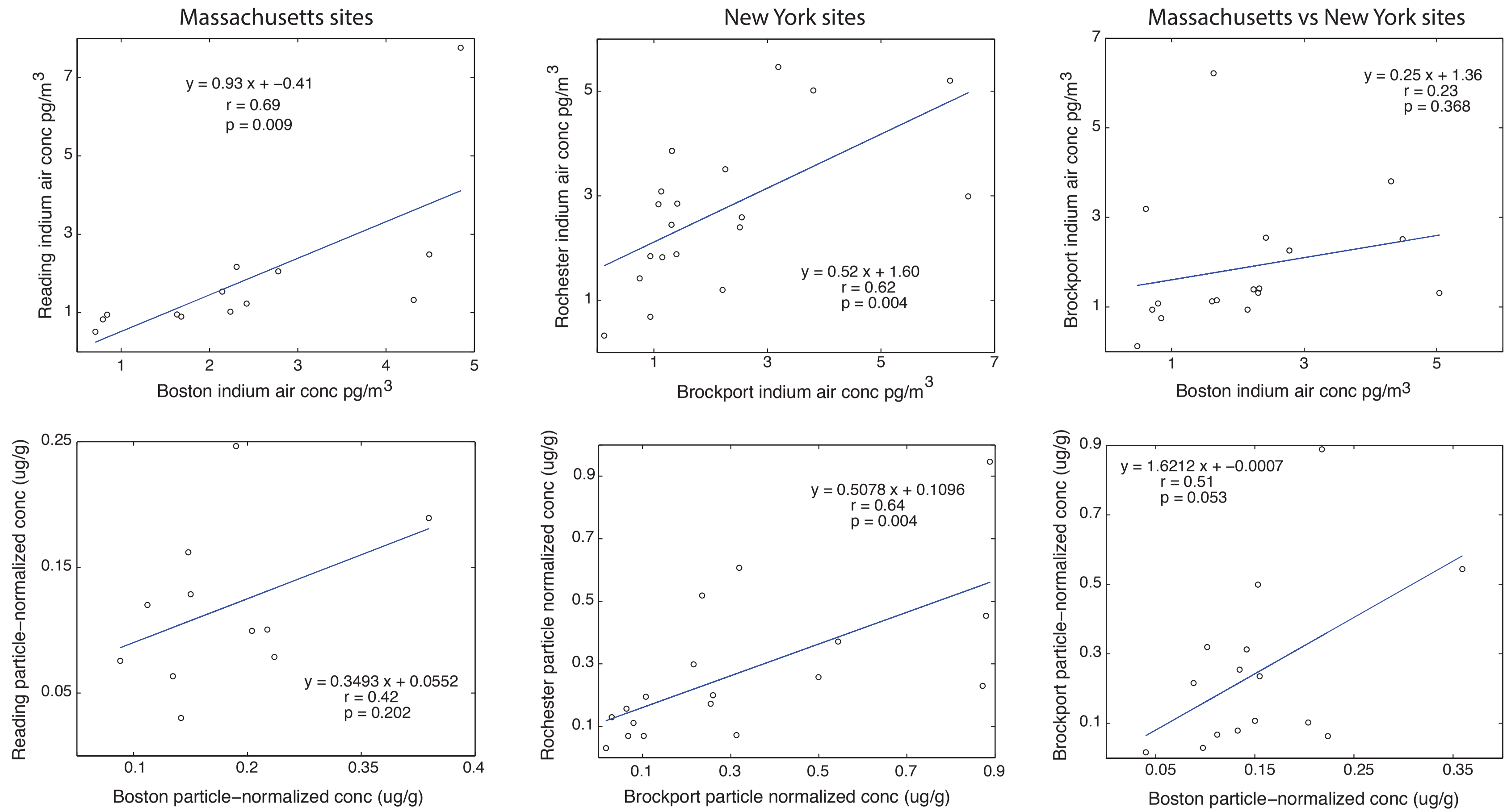

Figure S2: Indium concentrations in the Massachusetts sites correlate relatively well with one another, as do the New York sites. The pattern of indium concentrations in the Massachusetts sites are distinct from the New York sites.

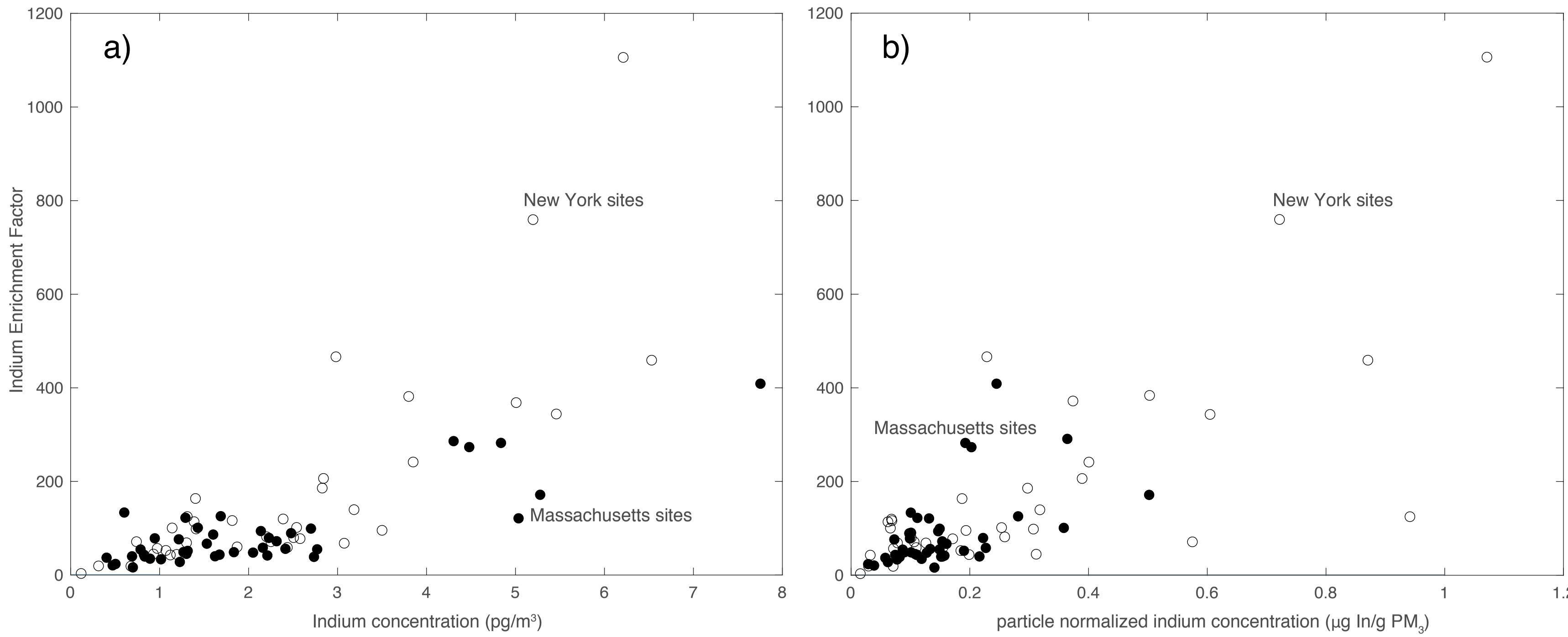

Figure S3: Enrichment factors for indium ranging from 15-1100 suggest that the source of indium to this region is likely not dust. Air concentration of indium versus Enrichment Factor (a) is relatively linear, with higher air concentrations generally reflecting larger enrichment factors. Particle-normalized indium concentrations correlate less well with the Enrichment Factor (b) but higher particle normalized concentrations still tend to reflect higher Enrichment Factors. Crustal values used[2,3]: Indium = 0.052 mg/kg; Aluminum = 8.4 weight %. Open circles = New York sites; closed circles = Massachusetts sites.

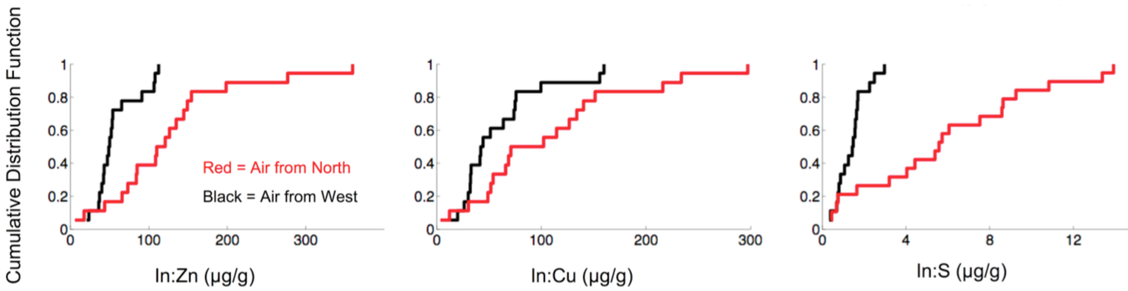

Figure S4: Air from the north has a distinctly different chemical makeup than air from the west. This can be seen in a correlation plot (Fig. 6), in cumulative distribution functions (this figure), and by computing a Kolmogorov-Smirnov statistic (Supplementary Table S2).

Table S2: Probability that air from the north is chemically different than air from the west (Kolmogorov-Smirnov test results for In:M mass ratios).

	P
In:Zn	0.997
In:Cu	0.87
In:Pb	0.85
In:S	0.9999
In:Fe	0.98
In:Ag	0.98

References

(1) Press, W. H.; Teukolsky, S. A.; Vetterling, W. T.; Flannery, B. R. *Numerical Recipes in C*; Cambridge University Press: Cambridge, UK, 1992; Vol. Second Edition; p 994.

(2) Wedepohl, K. H. The Composition of the Continental-Crust. *Geochimica et Cosmochimica Acta* **1995**, *59*, 1217–1232.

(3) Rudnick, R. L.; Gao, S. In *Composition of the Continental Crust*, 1st ed.; Holland, H. D., Turekian, K. K., Eds.; Treatise on Geochemistry; Elsevier/Pergamon: Amsterdam ; Boston, 2004; Vol. 3; Chapter 3.01, pp 1–64.

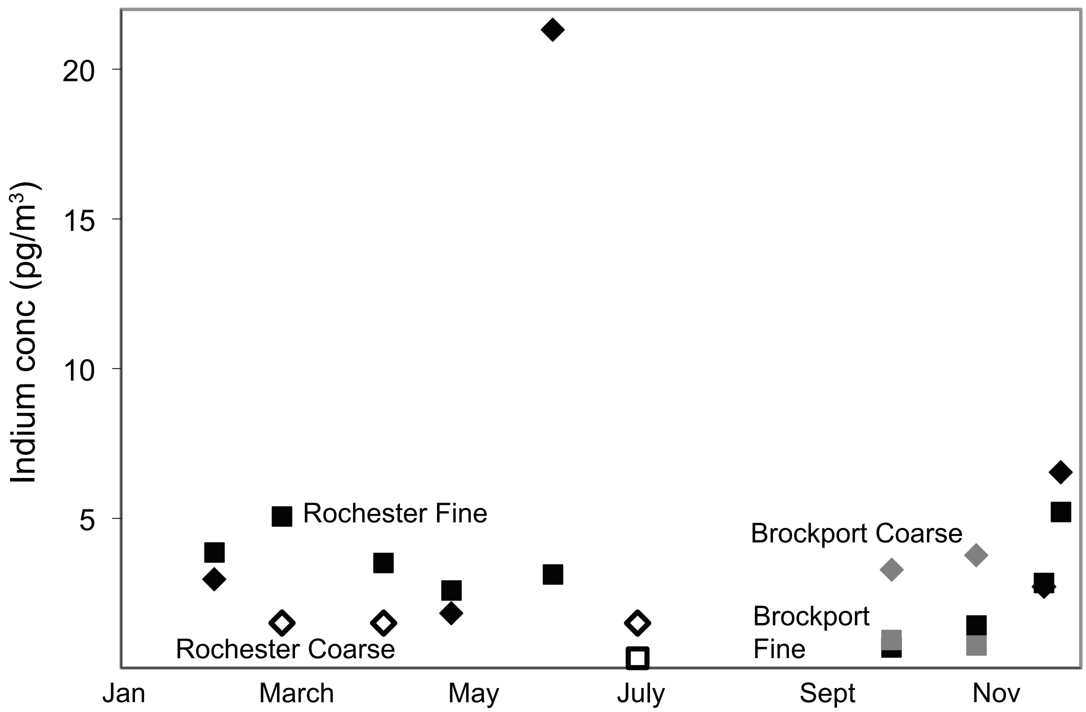

Figure S5: Indium concentrations contributed by coarse particles (PM_{3-10}) are similar to concentrations contributed by fine particles (PM_3). Hollow data points were below detection limit. Black squares = Rochester PM_3; Black diamonds = Rochester PM_{3-10}; Grey squares = Brockport PM_3; Grey diamonds = Brockport PM_{3-10}.