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Supplement of

Potential sources and processes affecting speciated atmospheric mercury at Kejimkujik National Park, Canada: comparison of receptor models and data treatment methods

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Supplement

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Figure S7. Observation–prediction time series in 2009. a) Case 2009, b) Case 09+mean, c) Case 09+median, d) Case 09+RM, e) Case 09-RM, and f) Case 09ScaleRM, observed GOM and PBM have been scaled.

Figure S8. Observation–prediction time series in 2010. a) Case 2010, b) Case 10+mean, c) Case 10+median, d) Case 10+RM, e) Case 10-RM, and f) Case 10ScaleRM, observed GOM and PBM have been scaled.

Section 1. Selection of the member of PMF factors

The number of PMF factors needs to be chosen according to the understanding of the sources impacting the samples utilized. When the background information is not enough to determine the number of factors, several methods could be used to determine the range of the number of the factors. The maximum individual column mean (IM) and the maximum individual column standard deviation (IS) of the scaled residual matrix can be used to identify the range of the number of factors. IM and IS will show a drastic drop when the number of factors increase up to a critical value (Lee et al., 1999). The optimal number of factors should be no less than the critical value. The trend of dQ also provides useful information on deciding the number of factors. When dQ becomes small as the number of factor increases, there might be too many factors (Hopke, 2000; Brown et al., 2015). Runs with different numbers of factors in the range determined by IM, IS, and dQ should be conducted. The model performance and the interpretability of factors in each run should be evaluated. The optimal PMF solution should be a compromise of those indexes and the interpretability of the factor profiles and their comparability with those from the literature (Belis et al, 2015a, 2015b; Cesari et al., 2016).

In this study, the IM and IS were calculated to determine the number of factors. The IM and IS dropped dramatically in 2009 when the number of factor increased to 3 (Figure S1). In the line plot of Q(Robust) and Q(true) vs. the number of factors (Figure S2), no significant decreases were found when the number of factors is larger than 5 in 2009. Therefore, the PMF was run using the number of factors from 3 to 5 in 2009. In 2010, the decrease of IS value was gradual while the IM value experienced a drastic drop when the number of factors increased to 3 (Figure S3). The trend of the Q (Robust) and Q (True) in 2010 is similar to 2009 (Figure S4). Therefore, the PMF runs with the number of factors from 3 to 5 were also conducted in 2010. The number of the factors selected (4) is a compromise of the trends of these indexes and the physical meanings of the factors obtained following Cesari et al. (2016). A detailed comparison of the physical meanings of solutions with different number of factors can be found in Liao (2016).

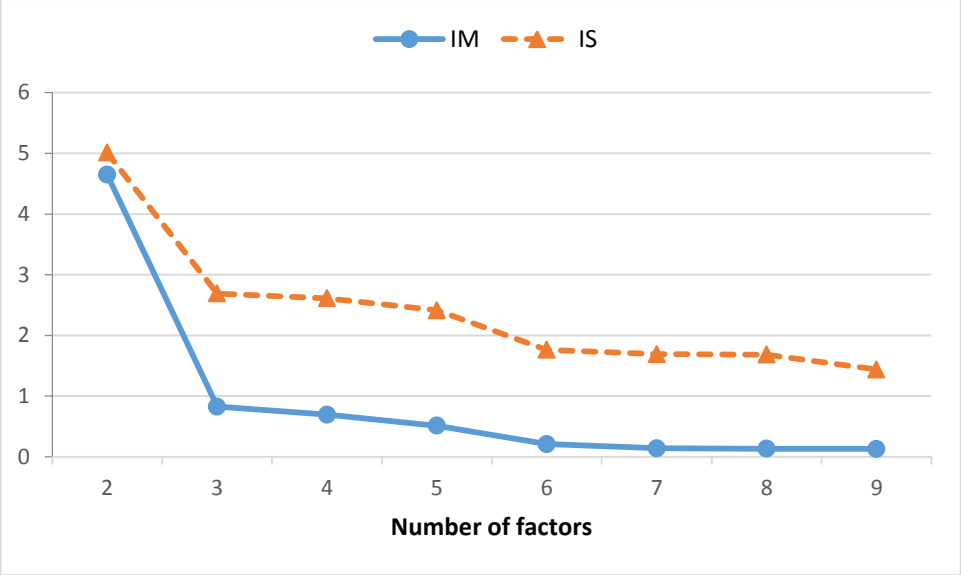


Figure S1. IM and IS vs number of PMF factors in 2009.

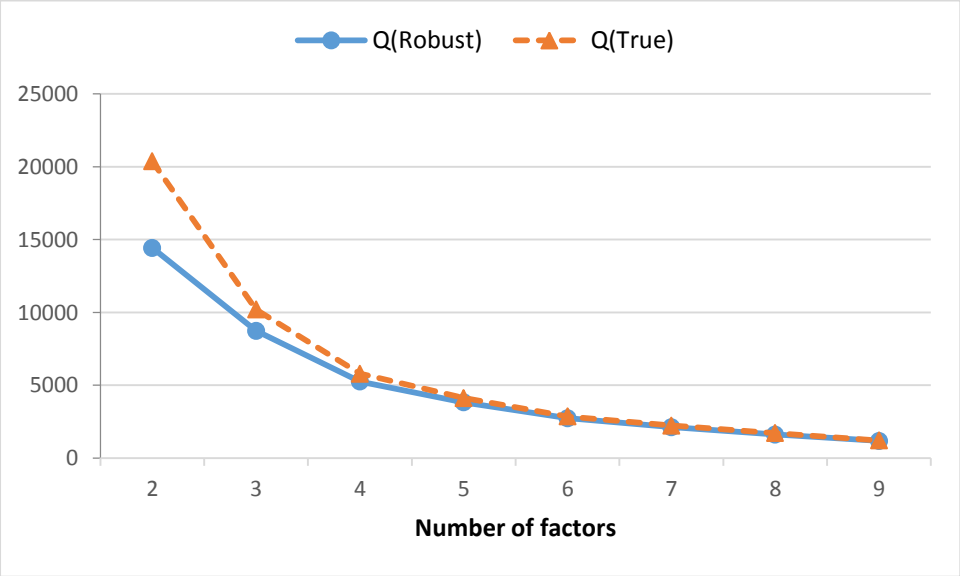


Figure S2. Q(Robust) and Q(true) vs number of PMF factors in 2009.

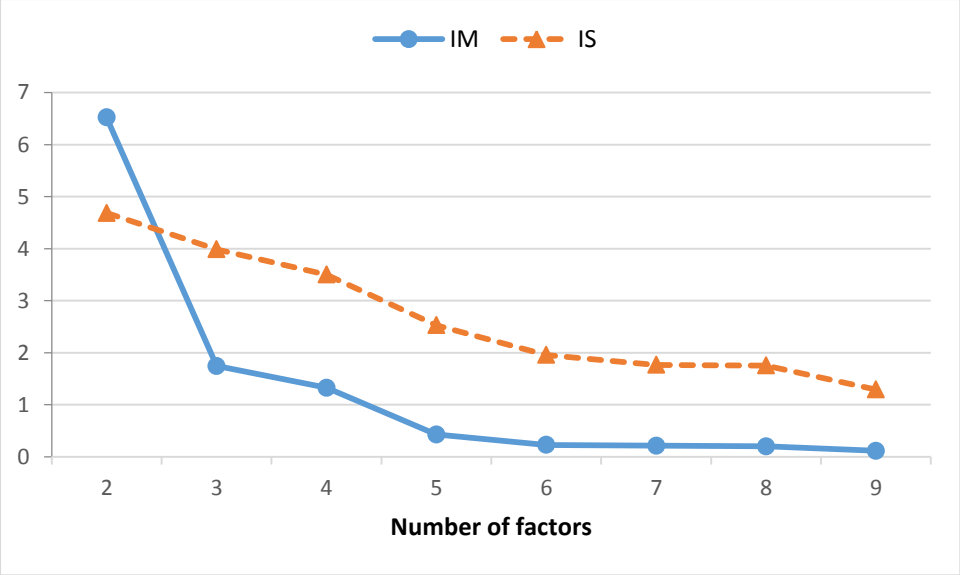


Figure S3. IM and IS vs number of PMF factors in 2010.

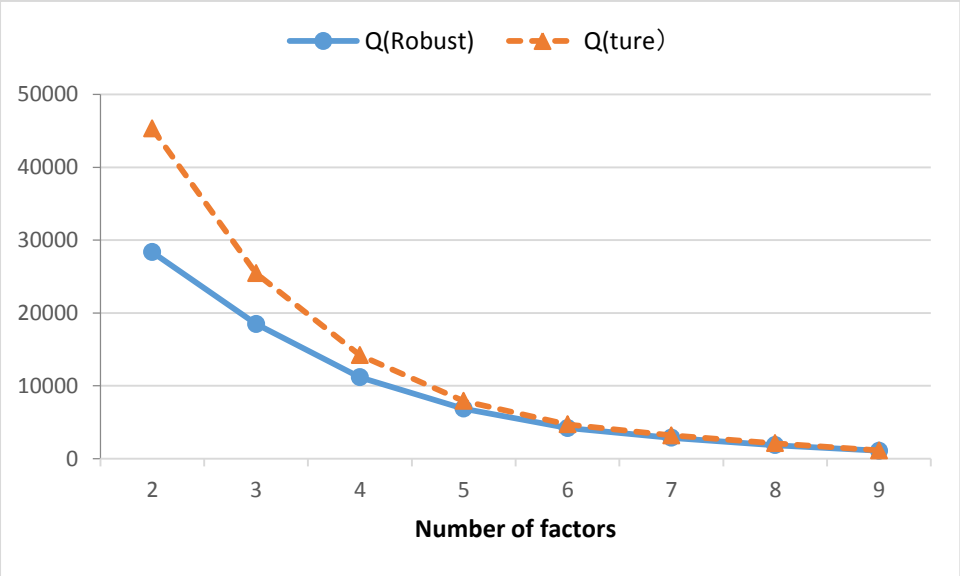


Figure S4. Q(Robust) and Q(true) vs number of PMF factors in 2010.

References

Belis, C. A., Pernigotti, D., Karagulian, F., Pirovano, G., Larsen, B. R., Gerboles, M., and Hopke, P. K.: A new methodology to assess the performance and uncertainty of source apportionment models in intercomparison exercises, Atmos. Environ., 119, 35-44, 10.1016/j.atmosenv.2015.08.002, 2015a.

Belis, C. A., Karagulian, F., Amato, F., Almeida, M., Artaxo, P., Beddows, D. C. S., Bernardoni, V., Bove, M. C., Carbone, S., Cesari, D., Contini, D., Cuccia, E., Diapouli, E., Eleftheriadis, K., Favez, O., El-Haddad, I., Harrison, R. M., Hellebust, S., Hovorka, J., Jang, E., Jorquera, H., Kammermeier, T., Karl, M., Lucarelli, F., Mooibroek, D., Nava, S., Nøjgaard, J. K., Paatero, P., Pandolfi,

M., Perrone, M. G., Petit, J. E., Pietrodangelo, A., Pokorná, P., Prati, P., Prevot, A. S. H., Quass, U., Querol, X., Saraga, D., Sciare, J., Sfetsos, A., Valli, G., Vecchi, R., Vestenius, M., Yubero, E., and Hopke, P. K.: A New Methodology to Assess the Performance and Uncertainty of Source Apportionment Models II: The Results of Two European Intercomparison Exercises, Atmos. Environ., 123, 240-250, 10.1016/j.atmosenv.2015.10.068, 2015b.

Brown, S. G., Eberly, S., Paatero, P., and Norris, G. A.: Methods for estimating uncertainty in PMF solutions: Examples with ambient air and water quality data and guidance on reporting PMF results, Sci. Total Environ., 518-519, 626-635, dx.doi.org/10.1016/j.scitotenv.2015.01.022, 2015.

Cesari, D., Amato, F., Pandolfi, M., Alastuey, A., Querol, X., and Contini, D.: An inter-comparison of PM10 source apportionment using PCA and PMF receptor models in three European sites, Environment Science and Pollution Research, 23, 15133-15148, 10.1007/s11356-016-6599-z, 2016.

Hopke, P. K.: A guide to positive matrix factorization, available at: https://www3.epa.gov/ttnamti1/files/ambient/pm25/workshop/laymen.pdf (last access: May 30, 2016), 2000.

Lee, E., Chan, C. K., & Paatero, P.: Application of positive matrix factorization in source apportionment of particulate pollutants in Hong Kong. Atmos. Environ., 33(19), 3201-3212, 10.1016/S1352-2310(99)00113-2, 1999.

Liao, Y.: Analysis of potential sources and processes affecting ambient speciated mercury concentrations at Kejimkujik National Park, Nova Scotia, Master's thesis, University of Windsor, Windsor, Ontria, Canada, 2016.

Section 2. Stability of PMF model runs

As pointed out in the PMF User's Guide (US EPA, 2014), when the Q (robust) values over several runs are highly variable, the stability of the result is poor. In this study, the differences of the Q (Robust) value between different runs were all smaller than 5 indicating that the results were quite stable in 2009 and 2010. More information can be found in Liao (2016).

References

Liao, Y.: Analysis of potential sources and processes affecting ambient speciated mercury concentrations at Kejimkujik National Park, Nova Scotia, Master's thesis, University of Windsor, Windsor, Ontria, Canada, 2016.

US Environmental Protection Agency (US EPA): EPA Positive Matrix Factorization (PMF) 5.0 Fundamentals and User Guide, available at: https://www.epa.gov/sites/production/files/2015-02/documents/pmf_5.0_user_guide.pdf (last access: May 30, 2016), 2014.

Table S1. Point source emissions of Hg and other pollutants reported in NPRI in the province of Nova Scotia (Data source: Environmental Canada, 2016). Bold facilities are shown in Figure 1.

Facility	Location (lat, long)	Distance to KEJ/direction	Hg (Kg)		SO_2 (Tonnes)		NO_2 (Tonnes)		NH_3 (Tonnes)	
			2009	2010	2009	2010	2009	2010	2009	2010
Brooklyn Power	Brooklyn (44.05°N, 64.70°W)	50 Km southeast	0	0	9.9	26	309	259	0	0
Michelin North America (Canada)- Bridgewater Plant	Bridgewater (44.39°N, 64.54°W)	53 Km east	0	0	195	184	68	63	0	0
High Liner Foods Inc.	Lunenburg (44.37°N,64.30°W)	72 Km east	0	0	27	27	0	0	0	0
Department of National Defence – 14 Wing Greenwood	Greenwood (44.98°N, 64.91°W)	75 Km north	0	0	55	68	19	18	0	0
Louisana Pacific Canada Ltd.	East River (44.58°N, 64.16°W)	88 Km northeast	0	0	122	102	100	99	0	0
Maple Leaf Foods – Larsen Packers Limited	Berwick (45.05°N, 64.75°W)	89 Km northeast	0	0	51	38	0	0	0	0
Michelin North America (Canada) - Waterville Plant	Waterville (45.05°N, 64.65°W)	92 Km northeast	0	0	162	182	57	62	0	0
Acadia University – Acadia Campus	Wolfville (45.08°N, 64.37°W)	108 Km northeast	0	0	77	73	27	26	0	0
CKF. Inc.	Hantsport (45.06°N, 64.17°W)	116 Km northeast	0	0	66	57	21	72	0	0
Minas Basin Pulp and Power	Hantsport (45.07°N, 64.17°W)	116 Km northeast	0	0	225	260	66	76	0	0
Mount Saint Vincent University	Halifax (44.67°N, 63.65°W)	129 Km northeast	0	0	27	13	7.2	3.9	0	0
Department of National Defence – Canadian Forces Ammunition Depot	Bedford (44.71°N, 63.63°W)	131 Km northeast	0	0	56	50	0	0	0	0

Table S1 – Continued 1

Facility	Location (lat, long)	Distance to KEJ/direction	Hg (Kg)		SO_2 (Tonnes)		NO_2 (Tonnes)		NH_3 (Tonnes)	
			2009	2010	2009	2010	2009	2010	2009	2010
Department of National Defence - Windsor Park	Halifax (44.66°N, 63.61°W)	132 Km northeast	0	0	59	44	36	30	0	0
Department of National Defence – Stadacona/Dockyard	Halifax (44.66°N, 63.58°W)	133 Km northeast	0	0	211	177	58	51	0	0
Capital Health – Camp Hill Site Heating Plant	Halifax (44.64°N, 63.59°W)	133 Km northeast	0	0	15	12	14	20	0	0
DalHousie University	Halifax (44.64°N, 63.59°W)	133 Km northeast	0.18	0.15	253	260	89	72	0	0
Saint Mary's University	Halifax (44.63°N, 63.58°W)	133 Km northeast	0	0	1.2	0	3	0	0	0
Oland Brewery	Halifax (44.66°N, 63.60°W)	133 Km Northeast	0	0	31	0	0	0	0	0
Nova Scotia Power – Tufts Cove Generating Station	Dartmouth (44.67°N, 63.60°W)	134 Km northeast	0	0	2,205	2,205	3,054	3,054	0	0
Capital Health-Victoria General Hospital Central Heating Plant	Halifax (44.64°N, 63.58°W)	134 Km northeast	0	0	215	7.6	60	19	0	0
Maritime Paper Products Ltd.	Dartmouth (44.70°N, 63.60°W)	134 Km northeast	0	0	7.2	0.868	3.1	2.1	0	0
Nova Scotia Power –Burnside Combustion Turbines	Dartmouth (44.71°N, 63.61°W)	134 Km northeast	0	0	0	0	60	40	0	0
Capital Health – Nova Scotia Hospital Central Heating Plant	Dartmouth (44.65°N, 63.55°W)	136 Km northeast	0	0	3.3	1.1	9.3	8.7	0	0
Imperial Oil – Dartmouth Refinery	Dartmouth (44.64°N, 63.54°W)	137 Km northeast	2.6	2.9	4,231	3,073	1,543	1,251	0.593	2.2
Department of National Defence – 12 Wing Shearwater	Shearwater (44.63°N, 63.51°W)	138 Km northeast	0	0	150	127	43	38	0	0
Martells Contracting	Elmsdale (44.96°N, 63.48°W)	154 Km northeast	0	0	28	17	4.5	2.8	0	0

Table S1 – Continued 2

Facility	Location (lat, long)	Distance to KEJ/direction	Hg (Kg)		SO_2 (Tonnes)		NO_2 (Tonnes)		NH_3 (Tonnes)	
			2009	2010	2009	2010	2009	2010	2009	2010
The Shaw Group Ltd.	Hardwoodlands (45.07°N, 63.52°W)	160 Km northeast	0	0	0	0	27	19	0	0
Lafarge Canada Inc. – Brookfield Cement Plant	Brookfield (45.24°N, 63.33°W)	180 Km northeast	5	5.9	562	667	498	591	0	0
Polycello	Amherst (45.82°N, 64.23°W)	183 Km northeast	0	0	0.003	0.002	0.462	0.335	0	0
Enligna Canada Inc.	Middle Musquodoboit (45.13°N, 62.95°W)	188 Km northeast	0	0	2.8	2.9	25	26	0	0
Oxford Frozen Foods	Oxford (45.73°N, 63.85°W)	188 Km northeast	0	0	66	59	0	0	0.9	0
Municipality of the county of Colchester – Wastewater Treatment Facility	Truro (45.37°N, 63.34°W)	188 Km northeast	0	0	0	0	0	0	2	0.08
Crossley Carpet Mills Limited	Truro (45.35°N, 63.29°W)	189 Km northeast	0	0	40	32	12	11	0	0
Rothsay	Truro (45.36°N, 63.31°W)	189 Km northeast	0	0	77	60	0	0	0	0
Stanfield's Ltd.	Truro (45.37°N, 63.28°W)	191 Km northeast	0	0	21	21	0	0	0	0
Stella-Jones Inc.	Truro (45.38°N, 63.27°W)	192 Km northeast	0	0	12	19	2.9	4.2	0	0
The Canadian Salt Company Limited – Pugwash Mine and Refinery	Pugwash (45.84°N, 63.66°W)	209 Km northeast	0	0	168	153	32	31	0	0
Michelin North America (Canada) – Pictou County Plant	New Glasgow (45.62°N, 62.74°W)	245 Km northeast	0	0	209	229	72	78	0	0
Maritime Steel and Foundries Limited	New Glasgow (45.58°N, 62.64°W)	245 Km northeast	0	0	0.25	0	0.875	0	0	0

Table S1 – Continued 3

Facility	Location (lat, long)	Distance to KEJ/direction	Hg (Kg)		SO_2 (Tonnes)		NO_2 (Tonnes)		NH_3 (Tonnes)	
			2009	2010	2009	2010	2009	2010	2009	2010
Nova Scotia Power – Trenton Generating Station	Trenton (45.62°N, 62.64°W)	248 Km northeast	33	19	30,429	19,257	5,126	5,577	0	0
Nova Forge Corporation	Trenton (45.62°N, 62.64°W)	248 Km northeast	0	0	3.1	0	0	0	0	0
Northern Pulp Nova Scotia Corporation	New Glasgow (45.65°N, 62.72°W)	266 Km northeast	0	0	246	89	688	676	42	46
St. Francis Xavier University	Antigonish (45.62°N, 61.99°W)	291 Km northwast	0	0	41	36	25	17	0	0
Exxonmobil Canada Properties – Goldboro Gas Plant	Goldboro (45.17°N, 61.61°W)	300 Km northeast	0	0	0	0	521	415	0	0
Nova Scotia Power – Point Tupper Generating Station	Port Hawkesbury (45.58°N, 61.35°W)	335 Km northeast	12	9.5	9,394	5,721	1,952	1,952	0	0
Newpage Port Hawkesbury Corp.	Port Hawkesbury (45.60°N, 61.36°W)	355 Km northeast	0	0	294	85	404	306	0.23	0.23
Exxonmobil Canada Properties – Point Tupper Fractionation Plant	Port Hawkesbury (45.58°N, 61.34°W)	335 Km northeast	0	0	0	0	48	23	0	0
Exxonmobil Canada Properties – Thebaud Platform	Offshore (43.01°N, 59.98°W)	402 Km east	0	0	0	0	135	126	0	0
Exxonmobil Canada Properties – North Triumph Platform	North Triumph Platform (43.01°N, 58.98°W)	433 Km east	0	0	0	0	26	29	0	0
Nova Scotia Power – Point Aconi Generating Station	Point Aconi (46.32°N, 60.30°W)	442 Km northeast	2.7	2.8	3,627	3,365	1,759	1,747	0	0
Exxonmobil Canada Properties – Venture Platform	Venture Platform (44.06°N, 59.58°W)	450 Km east	0	0	18	0	54	51	0	0

Table S1 – Continued 4

Facility	Location (lat, long)	Distance to KEJ/direction	Hg (Kg)		SO_2 (Tonnes)		NO_2 (Tonnes)		NH_3 (Tonnes)	
			2009	2010	2009	2010	2009	2010	2009	2010
Cape Breton University	Sydney (46.17°N, 60.09°W)	450 Km northeast	0	0	60	57	0	0	0	0
Nova Scotia Power – Lingan Generating Station	Lingan (46.23°N, 60.04°W)	457 Km northeast	92	50	55,208	33,479	5,106	5,219	0	0
Provincial total emission			147.5	90.3	108,961	70,336	22,165	22,166	45.7	48.5

Table S2. Coefficients of cross-correlation among all variables in 2009 (bold numbers are significant at $p<0.05$).

	GOM	PBM	PM	O_3	SO_2	HNO_3	Ca^{2+}	K^+	Na^+	Mg^{2+}	Cl^-	NO_3^-	NH_4^+	SO_4^{2-}	Temperature	Relative humidity	Wind speed	Precipitation
GEM	**0.37**	**0.28**	0.15	**0.48**	0.11	**0.18**	0.13	0.01	0.06	0.07	-0.01	0.14	**0.18**	**0.24**	0.03	0.06	**0.24**	**0.17**
GOM		0.10	**0.31**	**0.27**	**0.21**	**0.45**	**0.39**	**0.17**	-0.09	-0.02	**-0.18**	**0.17**	**0.28**	**0.27**	**0.46**	**-0.38**	-0.01	-0.09
PBM			**0.47**	**0.56**	**0.63**	**0.42**	**0.28**	0.11	**0.20**	**0.23**	0.06	**0.50**	**0.53**	**0.54**	**-0.43**	**-0.38**	-0.09	-0.15
PM				**0.52**	**0.64**	**0.70**	**0.68**	**0.43**	**0.37**	**0.44**	**0.19**	**0.66**	**0.80**	**0.79**	**0.17**	**-0.36**	-0.05	**-0.18**
O_3					**0.49**	**0.52**	**0.51**	0.05	**0.21**	**0.26**	0.09	**0.36**	**0.50**	**0.58**	**-0.17**	**-0.49**	**0.20**	-0.09
SO_2						**0.80**	**0.52**	**0.29**	**0.29**	**0.34**	0.09	**0.71**	**0.74**	**0.70**	**-0.21**	**-0.31**	-0.10	**-0.19**
HNO_3							**0.67**	**0.33**	0.16	**0.23**	-0.05	**0.61**	**0.80**	**0.77**	**0.16**	**-0.37**	-0.13	**-0.16**
Ca^{2+}								**0.47**	**0.44**	**0.53**	**0.31**	**0.59**	**0.57**	**0.58**	**0.23**	**-0.37**	0.05	-0.09
K^+									**0.64**	**0.66**	**0.57**	**0.53**	**0.34**	**0.41**	**0.37**	-0.04	0.14	0.03
Na^+										**0.99**	**0.96**	**0.61**	**0.24**	**0.39**	-0.10	0.00	**0.25**	-0.10
Mg^{2+}											**0.93**	**0.64**	**0.30**	**0.45**	-0.06	-0.06	**0.25**	-0.11
Cl^-												**0.38**	0.02	**0.18**	-0.12	0.07	**0.29**	-0.07
NO_3^-													**0.72**	**0.68**	-0.02	**-0.17**	-0.01	-0.13
NH_4^+														**0.94**	0.12	**-0.31**	-0.09	-0.13
SO_4^{2-}															0.08	**-0.31**	0.01	-0.13
Temperature																-0.11	0.11	0.09
Relative humidity																	**0.26**	**0.39**
Wind speed																		**0.39**

Table S3. Coefficients of cross-correlation among all variables in 2010 (bold numbers are significant at $p<0.05$).

	GOM	PBM	O_3	SO_2	HNO_3	Ca^{2+}	K^+	Na^+	Mg^{2+}	Cl^-	NO_3^-	NH_4^+	SO_4^{2-}	Temperature	Relative humidity	Wind speed	Precipitation
GEM	**0.31**	0.11	**0.70**	0.02	-0.11	-0.01	**-0.13**	0.09	0.08	0.10	-0.01	-0.10	-0.11	**-0.48**	-0.02	**0.38**	**0.18**
GOM		**0.29**	**0.55**	**0.30**	**0.24**	0.07	0.07	-0.03	-0.01	-0.10	**0.18**	0.06	0.06	-0.04	**-0.66**	-0.06	**-0.18**
PBM			**0.32**	0.06	0.04	-0.02	-0.03	-0.07	-0.07	-0.08	0.03	0.01	0.00	**-0.16**	**-0.39**	**-0.15**	**-0.22**
O_3				**0.18**	0.11	0.02	-0.07	0.05	0.05	0.01	0.06	0.00	0.02	**-0.29**	**-0.39**	**0.31**	0.01
SO_2					**0.63**	**0.13**	**0.16**	0.05	0.09	-0.04	**0.25**	**0.26**	**0.31**	-0.03	**-0.31**	-0.10	**-0.13**
HNO_3						**0.25**	**0.34**	-0.11	0.00	**-0.24**	**0.28**	**0.48**	**0.53**	**0.33**	**-0.25**	**-0.23**	**-0.13**
Ca^{2+}							**0.57**	0.01	**0.38**	0.00	**0.70**	**0.78**	**0.71**	**0.19**	-0.10	-0.05	-0.02
K^+								0.09	**0.33**	0.06	**0.55**	**0.68**	**0.67**	**0.32**	**-0.16**	-0.08	-0.07
Na^+									**0.92**	**0.96**	**0.21**	-0.09	0.02	**-0.16**	0.08	**0.28**	-0.01
Mg^{2+}										**0.89**	**0.45**	**0.21**	**0.28**	-0.08	0.04	**0.24**	-0.01
Cl^-											0.08	**-0.13**	-0.06	**-0.20**	**0.15**	**0.32**	0.03
NO_3^-												**0.68**	**0.64**	0.10	**-0.23**	-0.05	-0.11
NH_4^+													**0.97**	**0.28**	**-0.15**	**-0.14**	-0.09
SO_4^{2-}														**0.29**	**-0.15**	**-0.13**	-0.10
Temperature															-0.10	**-0.19**	0.03
Relative humidity																**0.24**	**0.41**
Wind speed																	**0.47**

Table S4. PMF factor contributions to speciated Hg and ratios of reproduced to observed annual Hg concentrations in 2009.

a) Case 2009

Factor		Combustion emission	Industrial sulfur	Photochemistry & re-emission	Sea salt
GEM (%)	Min	0	0	0	0
	Max	20	56	97	97
	Average	4	6	77	14
	Median	2	3	83	9
Ratio of reproduced to observed annual mean:					0.97
GOM (%)	Min	0	0	0	0
	Max	100	0	100	0
	Average	26	0	73	0
	Median	21	0	79	0
Ratio of reproduced to observed annual mean:					0.86
PBM (%)	Min	0	0	0	0
	Max	0	93	100	92
	Average	0	22	69	9
	Median	0	14	74	5
Ratio of reproduced to observed annual mean:					1.03

b) Case 09+mean

Factor		Combustion emission	Industrial sulfur	Photochemistry & re-emission	Sea salt
GEM (%)	Min	0	0	0	0
	Max	79	34	98	99
	Average	5	4	78	13
	Median	3	2	83	7
Ratio of reproduced to observed annual mean:					0.94
GOM (%)	Min	0	0	0	0
	Max	97	0	100	0
	Average	17	0	83	0
	Median	12	0	88	0
Ratio of reproduced to observed annual mean:					1.19
PBM (%)	Min	0	0	0	0
	Max	0	87	100	94
	Average	0	23	67	10
	Median	0	19	71	5
Ratio of reproduced to observed annual mean:					1.19

c) Case 09+median

Factor		Combustion emission	Industrial sulfur	Photochemistry & re-emission	Sea salt
GEM (%)	Min	0	0	0	0
	Max	89	28	98	99
	Average	6	3	79	13
	Median	3	1	83	7

Ratio of reproduced to observed annual mean:					0.93
GOM (%)	Min	0	0	0	0
	Max	95	100	100	0
	Average	14	1	85	0
	Median	10	0	90	0
Ratio of reproduced to observed annual mean:					1.20
PBM (%)	Min	0	0	0	0
	Max	0	86	100	96
	Average	0	20	70	10
	Median	0	15	75	6
Ratio of reproduced to observed annual mean:					1.14

d) Case 09+RM

Factor		Combustion emission	Industrial sulfur	Photochemistry & re-emission	Sea salt
GEM (%)	Min	0	0	0	0
	Max	16	54	98	97
	Average	3	5	78	13
	Median	2	2	83	9
Ratio of reproduced to observed annual mean:					0.97
RM (%)	Min	0	0	0	0
	Max	37	83	100	18
	Average	10	16	73	1
	Median	7	11	78	0
Ratio of reproduced to observed annual mean:					1.04

e) Case 09-RM

Factor		Combustion emission	Industrial sulfur	Photochemistry & re-emission	Sea salt
GEM (%)	Min	0	0	0	0
	Max	34	15	98	96
	Average	7	1	79	13
	Median	4	1	84	9
Ratio of reproduced to observed annual mean:					0.97

f) Case 09ScaleRM

Factor name		Combustion emission	Industrial sulfur	Photochemistry & re-emission	Sea salt
GEM (%)	Min	0	0	0	0
	Max	0	65	100	99
	Average	0	7	78	15
	Median	0	4	83	10
Ratio of reproduced to observed annual mean:					0.97
GOM (%)	Min	0	0	0	0
	Max	97	0	100	67
	Average	23	0	75	2
	Median	18	0	81	1
Ratio of reproduced to observed annual mean:					0.75

PBM (%)	Min	0	0	0	0
	Max	0	88	100	96
	Average	0	16	74	10
	Median	0	10	80	6
Ratio of reproduced to observed annual mean:					0.94

Table S5. PMF factor contributions to speciated Hg and ratios of reproduced to observed annual Hg concentrations in 2010.

a) Case 2010

Factor		Combustion emission	Industrial sulfur	Photochemistry & re-emission	Sea salt
GEM (%)	Min	0	0	0	0
	Max	100	9	99	100
	Average	11	1	79	9
	Median	7	1	85	4
Ratio of reproduced to observed annual mean:					0.98
GOM (%)	Min	0	0	0	0
	Max	100	100	100	0
	Average	5	29	67	0
	Median	2	28	68	0
Ratio of reproduced to observed annual mean:					1.34
PBM (%)	Min	0	0	0	0
	Max	100	28	98	99
	Average	11	4	80	5
	Median	6	3	86	2
Ratio of reproduced to observed annual mean:					1.00

b) Case 10+mean

Factor		Combustion emission	Industrial sulfur	Photochemistry & re-emission	Sea salt
GEM (%)	Min	0	0	0	0
	Max	29	70	99	100
	Average	3	6	83	8
	Median	2	3	89	4
Ratio of reproduced to observed annual mean:					0.96
GOM (%)	Min	0	0	0	0
	Max	32	99	100	0
	Average	2	27	70	0
	Median	2	21	76	0
Ratio of reproduced to observed annual mean:					1.35
PBM (%)	Min	0	0	0	0
	Max	0	84	100	98
	Average	0	4	93	2
	Median	0	2	97	1
Ratio of reproduced to observed annual mean:					0.87

c) Case 10+median

Factor		Combustion emission	Industrial sulfur	Photochemistry & re-emission	Sea salt
GEM (%)	Min	0	0	0	0
	Max	39	1	100	100
	Average	3	0	88	8
	Median	2	0	93	4

Ratio of reproduced to observed annual mean:					0.97
GOM (%)	Min	0	0	0	0
	Max	5	100	100	0
	Average	0	36	64	0
	Median	0	36	63	0
Ratio of reproduced to observed annual mean:					1.32
PBM (%)	Min	0	0	0	0
	Max	0	0	100	100
	Average	0	0	97	3
	Median	0	0	99	1
Ratio of reproduced to observed annual mean:					0.88

d) Case 10+RM

Factor		Combustion emission	Industrial sulfur	Photochemistry & re-emission	Sea salt
GEM (%)	Min	0	0	0	0
	Max	100	9	99	100
	Average	11	1	79	9
	Median	7	1	85	4
Ratio of reproduced to observed annual mean:					0.98
RM (%)	Min	0	0	0	0
	Max	100	47	98	96
	Average	9	8	80	3
	Median	5	6	86	1
Ratio of reproduced to observed annual mean:					1.16

e) Case 10-RM

Factor		Combustion emission	Industrial sulfur	Photochemistry & re-emission	Sea salt
GEM (%)	Min	0	0	0	0
	Max	100	10	99	100
	Average	11	1	78	9
	Median	7	1	85	4
Ratio of reproduced to observed annual mean:					0.98

f) Case 10ScaleRM

Factor		Combustion emission	Industrial sulfur	Photochemistry & re-emission	Sea salt
GEM (%)	Min	0	0	0	0
	Max	100	10	99	100
	Average	11	1	78	9
	Median	7	1	85	4
Ratio of reproduced to observed annual mean:					0.98
GOM (%)	Min	0	0	0	0
	Max	100	69	99	75
	Average	8	14	77	1
	Median	4	11	80	0
Ratio of reproduced to observed annual mean:					1.23

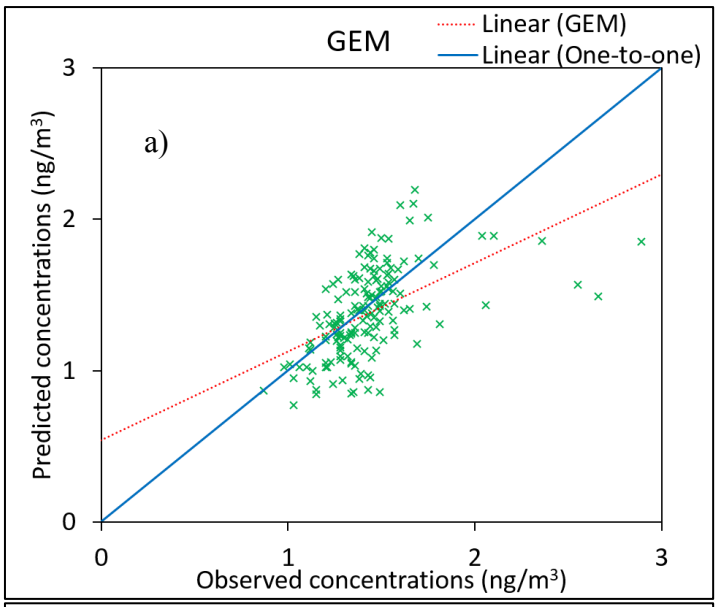
PBM (%)	Min	0	0	0	0
	Max	100	40	97	98
	Average	12	6	76	6
	Median	7	4	82	2
Ratio of reproduced to observed annual mean:					0.88

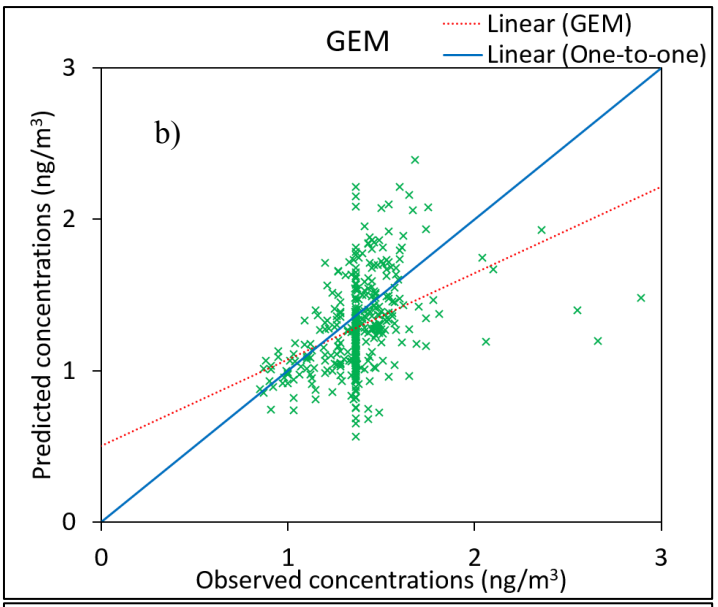
Table S6. Pearson correlation coefficients between Hg forms and other compounds in Case 2009, Case 09+mean, and Case 09+median (bold numbers are significant at $p<0.05$).

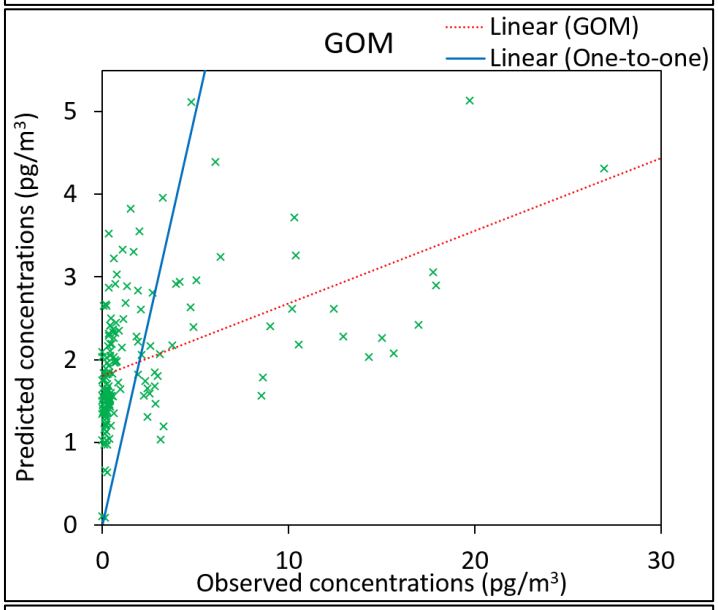
	GEM2009	GEM09+mean	GEM09+medain	GOM2009	GOM09+mean	GOM09+median	PBM2009	PBM09+mean	PBM09+median
GEM	1.00	1.00	1.00	**0.37**	**0.37**	**0.35**	**0.28**	**0.35**	**0.35**
GOM	**0.37**	**0.37**	**0.35**	1.00	1.00	1.00	0.11	**0.20**	**0.19**
PBM	**0.28**	**0.35**	**0.35**	0.11	**0.20**	**0.19**	1.00	1.00	1.00
PM	0.15	**0.11**	**0.12**	**0.31**	**0.19**	**0.18**	**0.48**	**0.30**	**0.31**
O_3	**0.48**	**0.52**	**0.50**	**0.28**	**0.24**	**0.25**	**0.56**	**0.44**	**0.45**
SO_2	0.11	**0.16**	**0.16**	**0.21**	**0.19**	**0.19**	**0.63**	**0.51**	**0.52**
HNO_3	**0.18**	**0.14**	**0.16**	**0.45**	**0.27**	**0.27**	**0.42**	**0.27**	**0.29**
Ca^{2+}	0.13	0.10	0.09	**0.39**	**0.36**	**0.36**	**0.28**	**0.26**	**0.26**
K^+	0.01	0.03	0.02	**0.17**	**0.17**	**0.17**	0.11	**0.14**	**0.13**
Na^+	0.06	**0.12**	0.10	-0.09	-0.03	-0.02	**0.20**	**0.22**	**0.21**
Mg^{2+}	0.07	**0.12**	**0.10**	-0.01	0.03	0.04	**0.23**	**0.24**	**0.23**
Cl^-	-0.01	0.06	0.04	**-0.18**	-0.09	-0.08	0.06	**0.11**	0.10
NO_3^-	0.14	**0.15**	**0.13**	**0.17**	**0.15**	**0.15**	**0.49**	**0.41**	**0.41**
NH_4^+	**0.18**	**0.12**	**0.14**	**0.28**	0.10	0.09	**0.53**	**0.22**	**0.24**
SO_4^{2-}	**0.24**	**0.13**	**0.15**	**0.27**	0.06	0.05	**0.53**	**0.17**	**0.19**

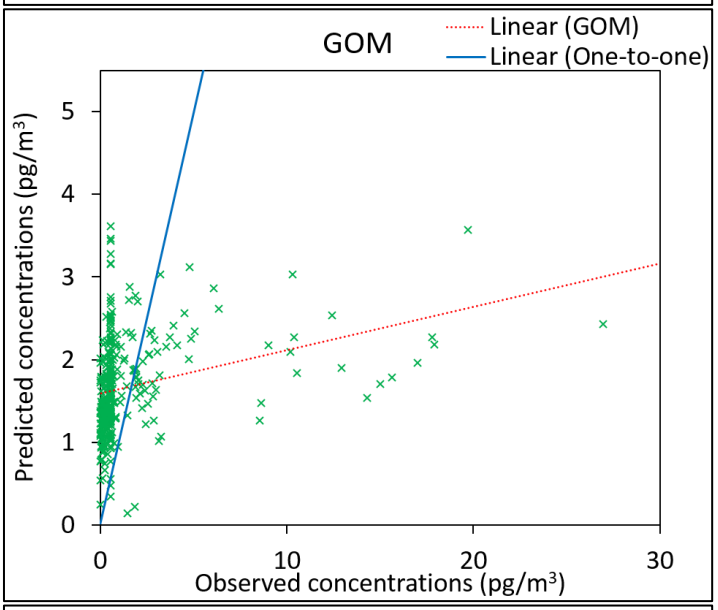
Table S7. Pearson correlation coefficients between Hg forms and other compounds in Case 2010, Case 10+mean, and Case 10+median (bold numbers are significant at $p<0.05$).

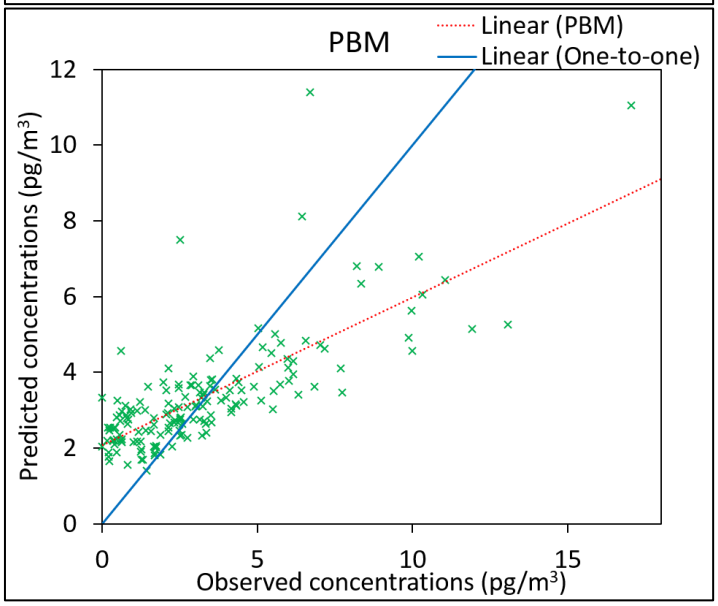
	GEM2010	GEM10+mean	GEM10+median	GOM2010	GOM10+mean	GOM10+median	PBM2010	PBM10+mean	PBM10+median
GEM	1.00	1.00	1.00	**0.32**	**0.29**	**0.29**	0.11	**0.19**	**0.19**
GOM	**0.32**	**0.29**	**0.29**	1.00	1.00	1.00	**0.29**	**0.15**	**0.15**
PBM	0.11	**0.19**	**0.19**	**0.29**	**0.15**	**0.15**	1.00	1.00	1.00
O_3	**0.70**	**0.68**	**0.68**	**0.56**	**0.51**	**0.51**	**0.32**	**0.29**	**0.29**
SO_2	0.01	0.00	0.00	**0.29**	**0.29**	**0.29**	0.05	-0.04	-0.03
HNO_3	**-0.12**	**-0.11**	**-0.11**	**0.23**	**0.24**	**0.24**	0.04	-0.04	-0.04
Ca^{2+}	-0.01	-0.02	-0.02	0.07	0.08	0.09	-0.02	-0.05	-0.05
K^+	**-0.13**	**-0.12**	**-0.13**	0.07	0.09	0.10	-0.03	-0.07	-0.08
Na^+	0.08	0.07	0.06	-0.03	0.00	-0.01	-0.07	**-0.10**	-0.09
Mg^{2+}	0.07	0.06	0.06	-0.01	0.02	0.02	-0.07	-0.10	-0.09
Cl^-	0.09	0.07	0.07	-0.10	-0.07	-0.07	-0.07	**-0.11**	**-0.11**
NO_3^-	-0.02	-0.02	-0.03	**0.18**	**0.19**	**0.20**	0.03	-0.03	-0.04
NH_4^+	-0.11	-0.10	**-0.10**	0.06	0.08	0.08	0.01	-0.04	-0.04
SO_4^{2-}	-0.11	-0.10	**-0.11**	0.06	0.08	0.08	0.00	-0.05	-0.05

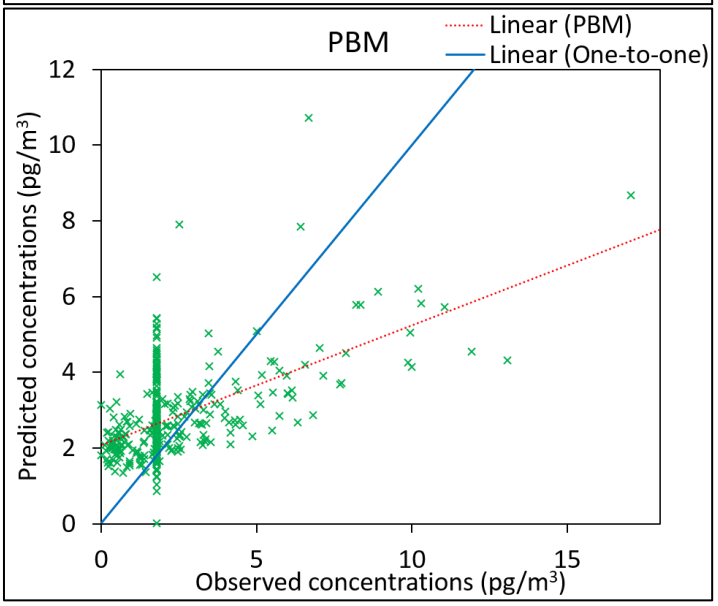

GEM
Linear (GEM)
Linear (One-to-one)
a)
Predicted concentrations (ng/m³)
Observed concentrations (ng/m³)
0
1
2
3

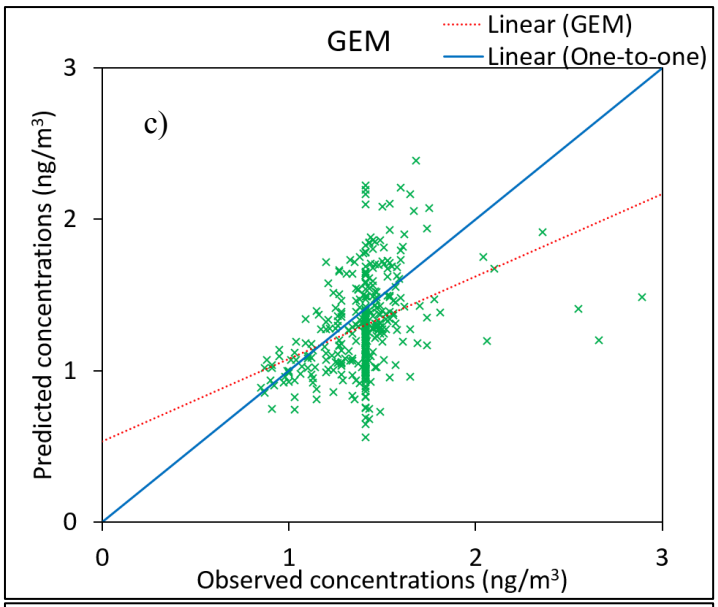

GEM
Linear (GEM)
Linear (One-to-one)
b)
Predicted concentrations (ng/m³)
Observed concentrations (ng/m³)
0
1
2
3

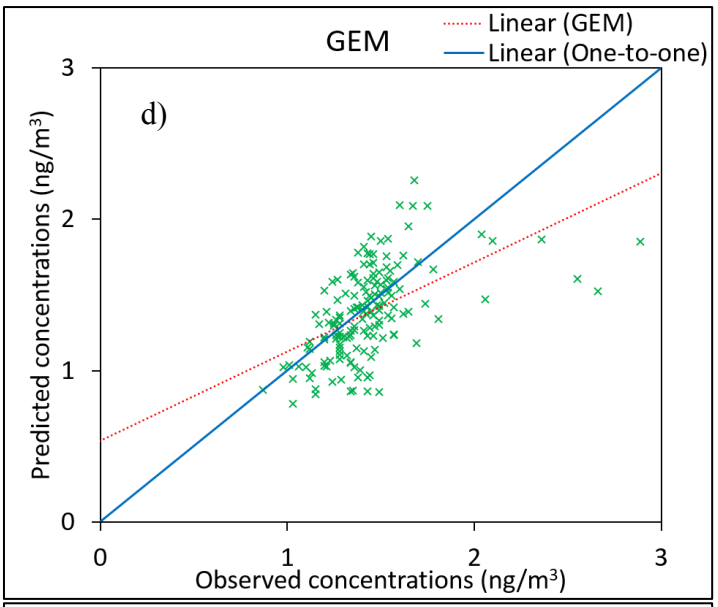

GOM
Linear (GOM)
Linear (One-to-one)
Predicted concentrations (pg/m³)
Observed concentrations (pg/m³)
0
10
20
30

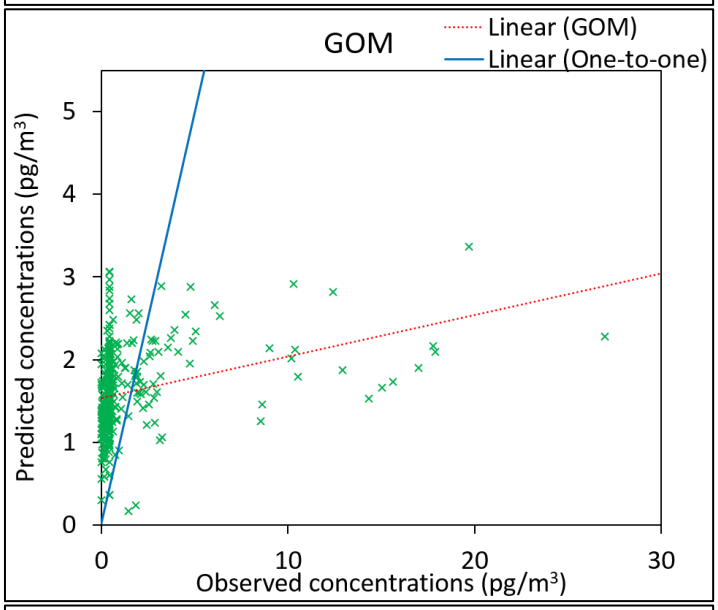

GOM
Linear (GOM)
Linear (One-to-one)
Predicted concentrations (pg/m³)
Observed concentrations (pg/m³)
0
10
20
30

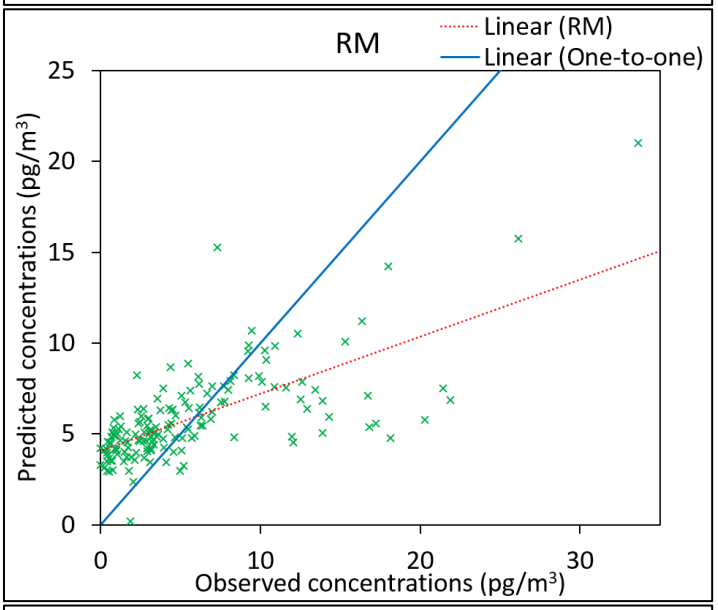

PBM
Linear (PBM)
Linear (One-to-one)
Predicted concentrations (pg/m³)
Observed concentrations (pg/m³)
0
5
10
15

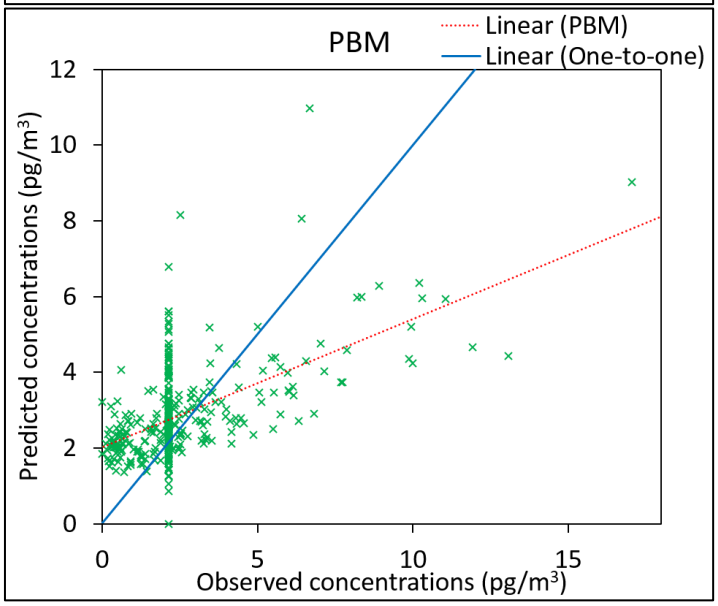

PBM
Linear (PBM)
Linear (One-to-one)
Predicted concentrations (pg/m³)
Observed concentrations (pg/m³)
0
5
10
15

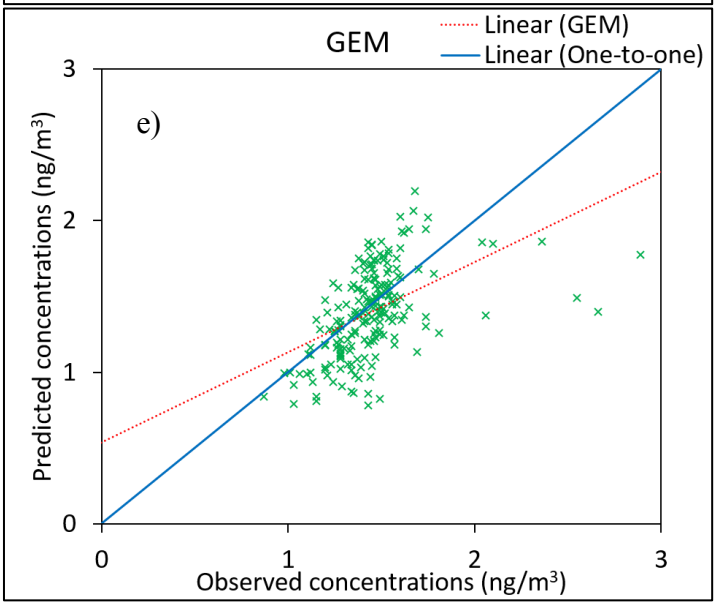

GEM
Linear (GEM)
Linear (One-to-one)
c)
Predicted concentrations (ng/m³)
Observed concentrations (ng/m³)
0
1
2
3


GEM
Linear (GEM)
Linear (One-to-one)
d)
Predicted concentrations (ng/m³)
Observed concentrations (ng/m³)
0
1
2
3


GOM
Linear (GOM)
Linear (One-to-one)
Predicted concentrations (pg/m³)
Observed concentrations (pg/m³)
0
10
20
30


RM
Linear (RM)
Linear (One-to-one)
Predicted concentrations (pg/m³)
Observed concentrations (pg/m³)
0
10
20
30


PBM
Linear (PBM)
Linear (One-to-one)
Predicted concentrations (pg/m³)
Observed concentrations (pg/m³)
0
5
10
15


GEM
Linear (GEM)
Linear (One-to-one)
e)
Predicted concentrations (ng/m³)
Observed concentrations (ng/m³)
0
1
2
3

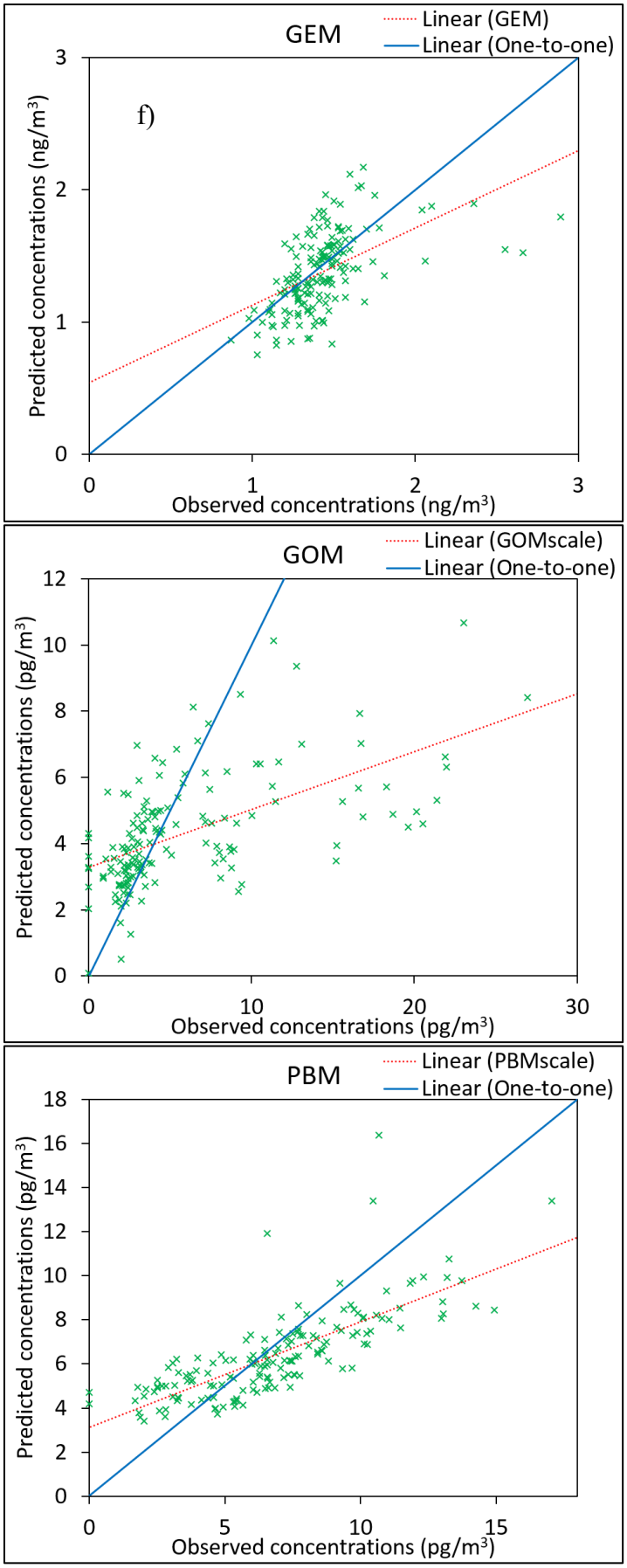
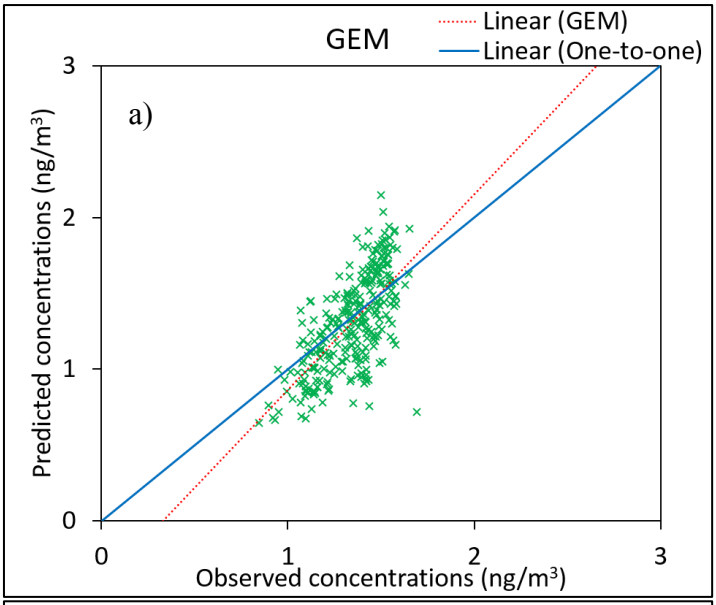
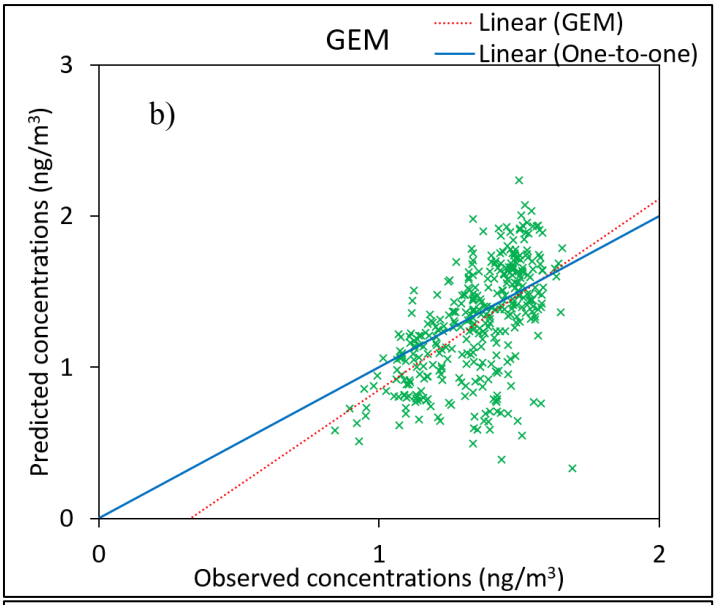
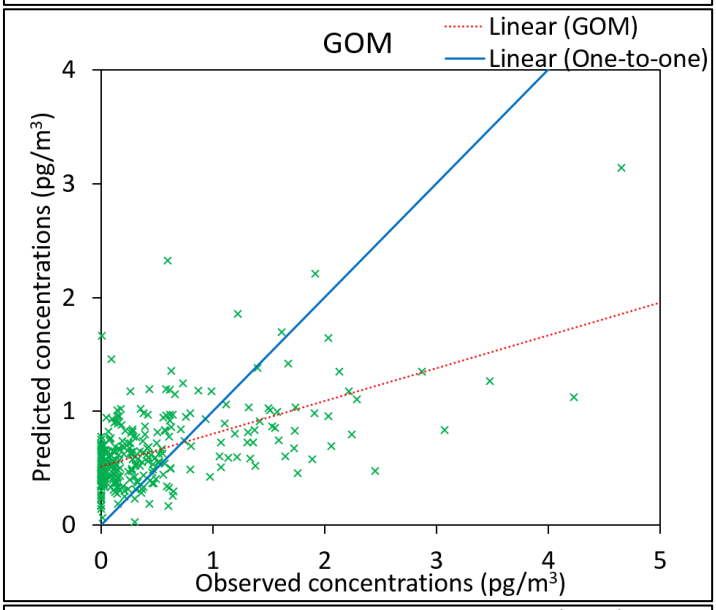
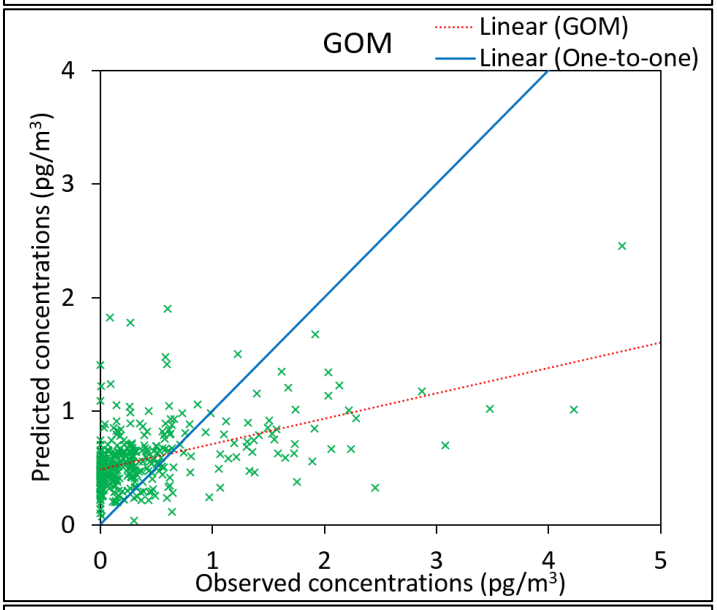


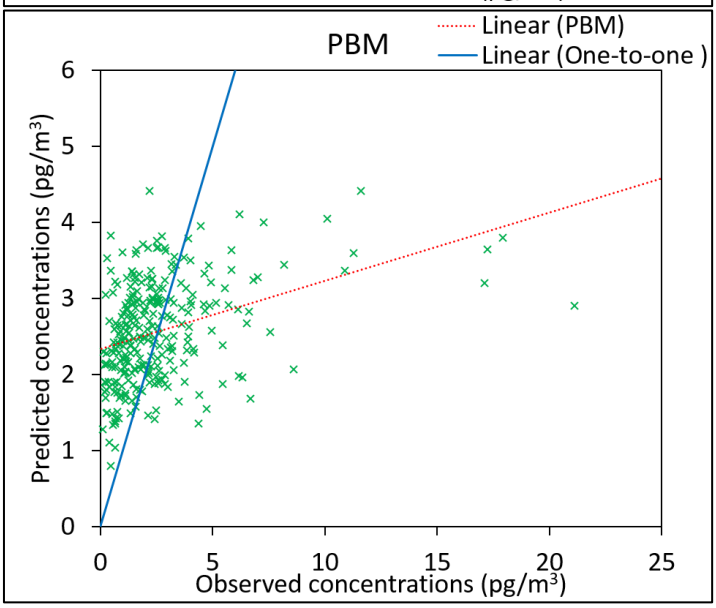
Figure S5. Observation–prediction scatter plot in 2009. a) Case 2009, b) Case 09+mean, c) Case 09+median, d) Case 09+RM, e) Case 09-RM, and f) Case 09ScaleRM, observed GOM and PBM have been scaled.

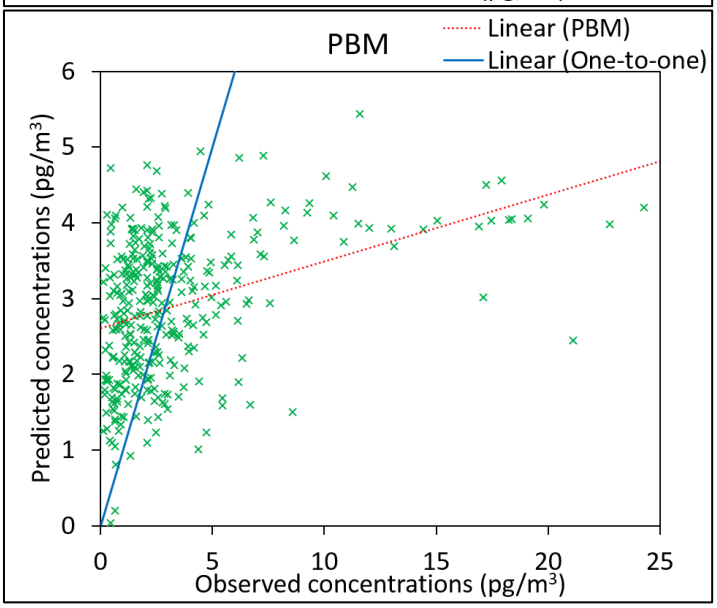

GEM
Linear (GEM)
Linear (One-to-one)
a)
Predicted concentrations (ng/m³)
Observed concentrations (ng/m³)
0
1
2
3

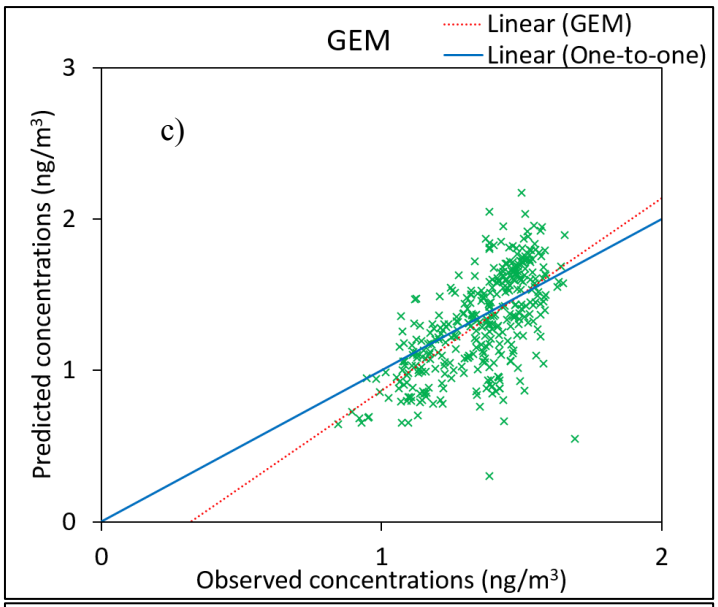

GEM
Linear (GEM)
Linear (One-to-one)
b)
Predicted concentrations (ng/m³)
Observed concentrations (ng/m³)
0
1
2
3

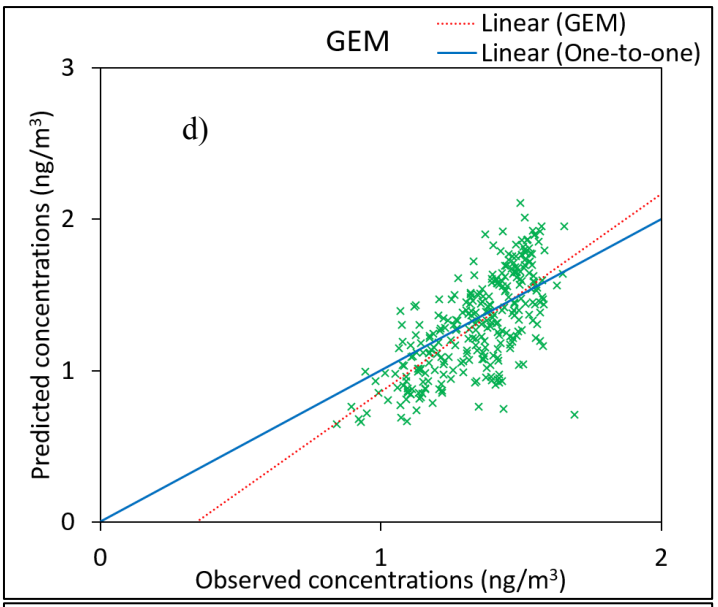

GOM
Linear (GOM)
Linear (One-to-one)
Predicted concentrations (pg/m³)
Observed concentrations (pg/m³)
0
1
2
3
4
5

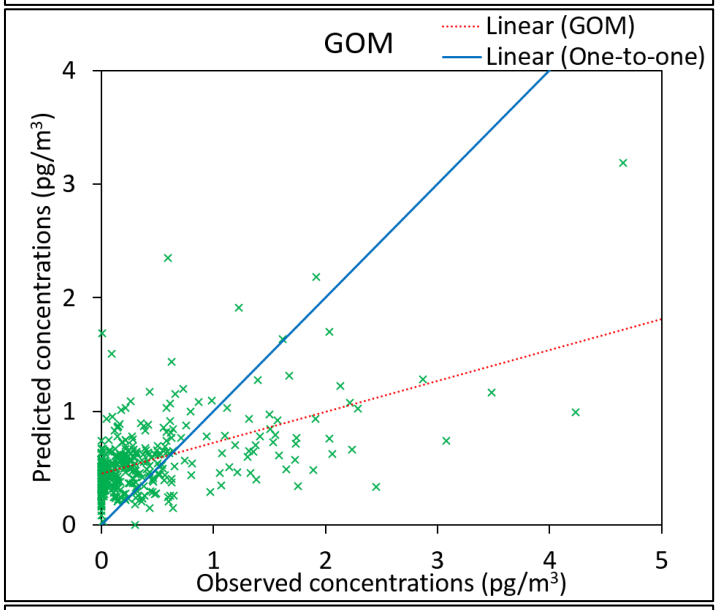

GOM
Linear (GOM)
Linear (One-to-one)
Predicted concentrations (pg/m³)
Observed concentrations (pg/m³)
0
1
2
3
4
5

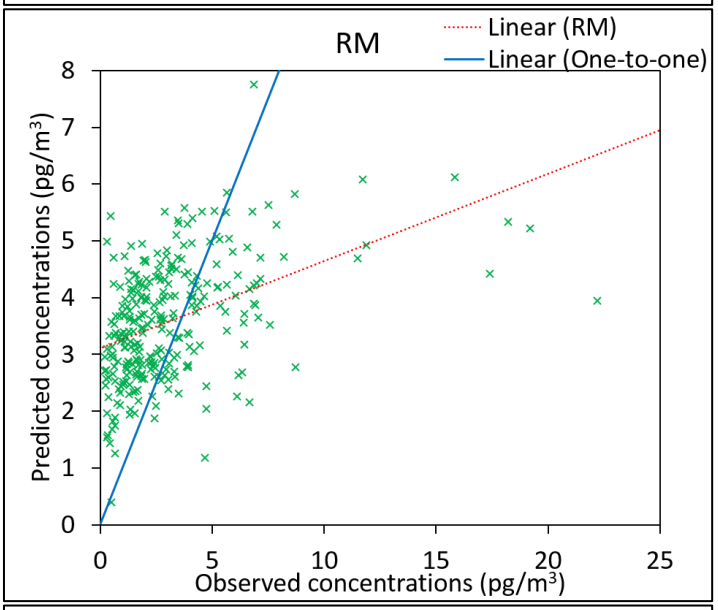

PBM
Linear (PBM)
Linear (One-to-one)
Predicted concentrations (pg/m³)
Observed concentrations (pg/m³)
0
5
10
15
20
25

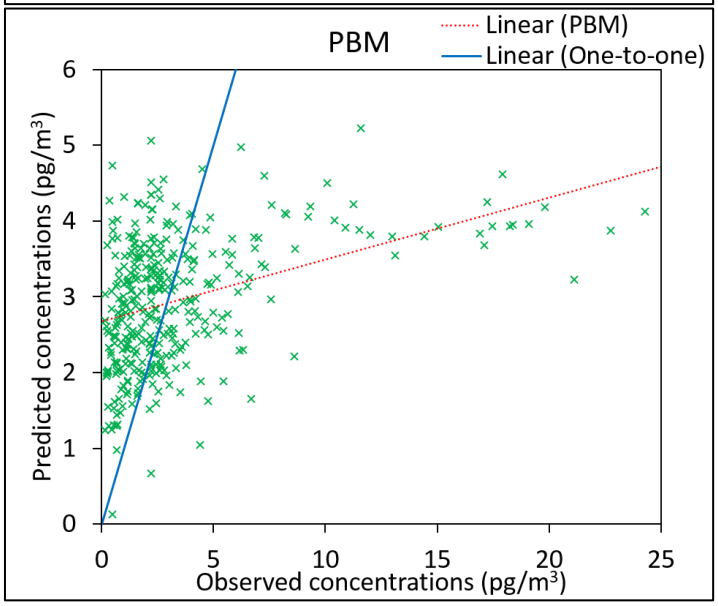

PBM
Linear (PBM)
Linear (One-to-one)
Predicted concentrations (pg/m³)
Observed concentrations (pg/m³)
0
5
10
15
20
25

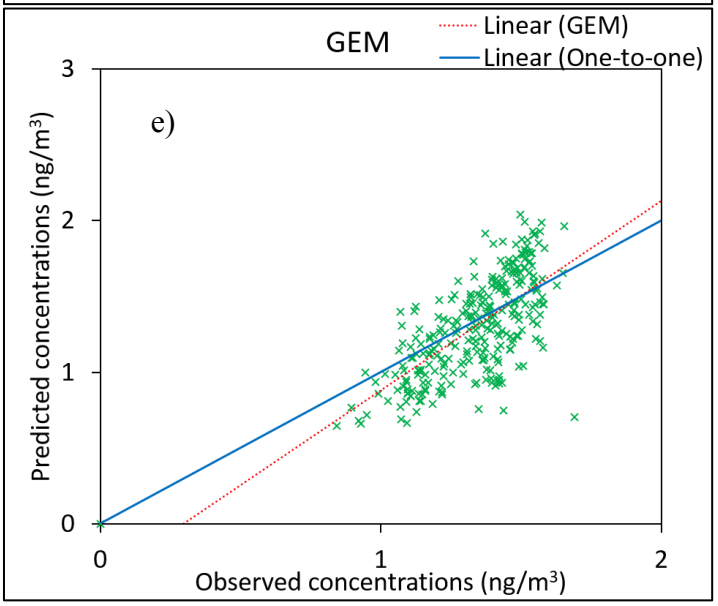

GEM
Linear (GEM)
Linear (One-to-one)
c)
Predicted concentrations (ng/m³)
Observed concentrations (ng/m³)
0
1
2
3


GEM
Linear (GEM)
Linear (One-to-one)
d)
Predicted concentrations (ng/m³)
Observed concentrations (ng/m³)
0
1
2
3


GOM
Linear (GOM)
Linear (One-to-one)
Predicted concentrations (pg/m³)
Observed concentrations (pg/m³)
0
1
2
3
4
5


RM
Linear (RM)
Linear (One-to-one)
Predicted concentrations (pg/m³)
Observed concentrations (pg/m³)
0
5
10
15
20
25


PBM
Linear (PBM)
Linear (One-to-one)
Predicted concentrations (pg/m³)
Observed concentrations (pg/m³)
0
5
10
15
20
25


GEM
Linear (GEM)
Linear (One-to-one)
e)
Predicted concentrations (ng/m³)
Observed concentrations (ng/m³)
0
1
2
3

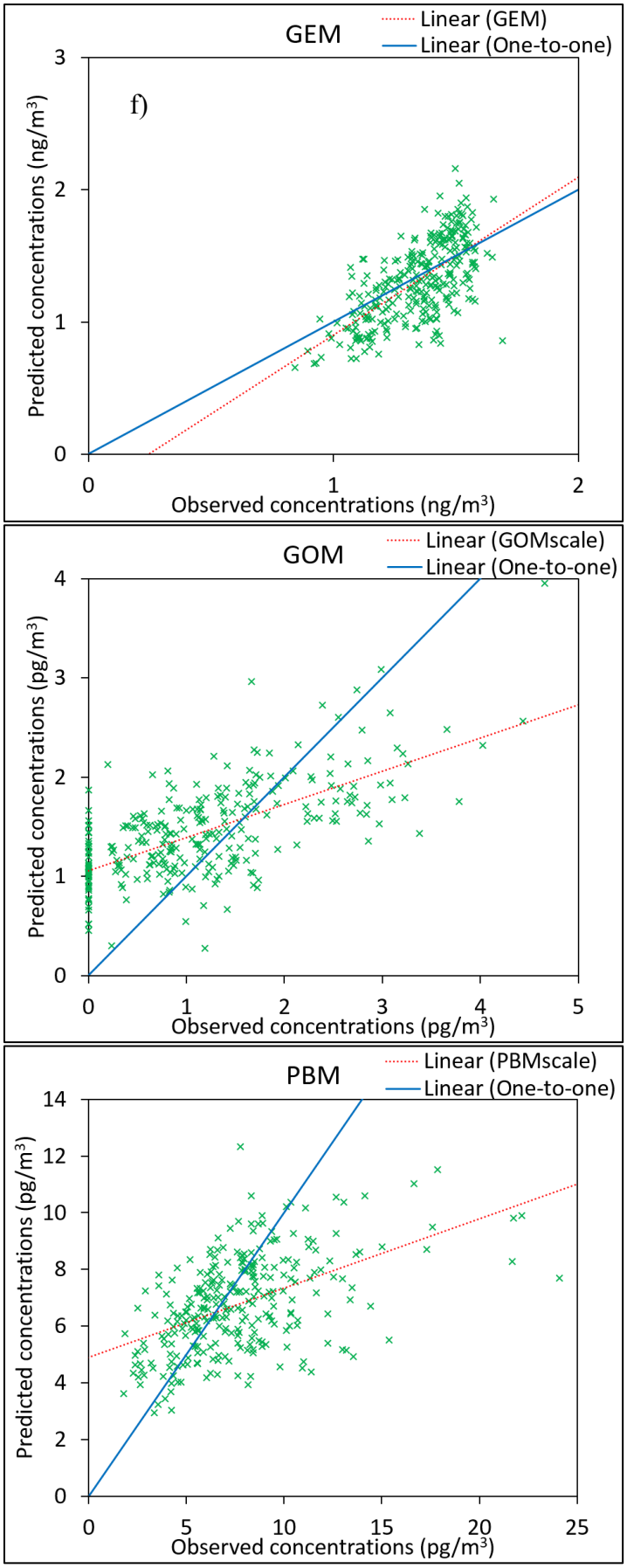
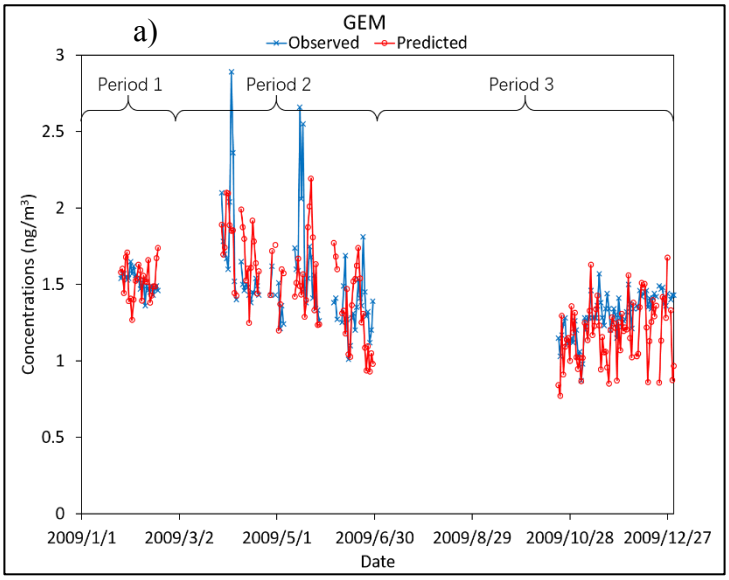
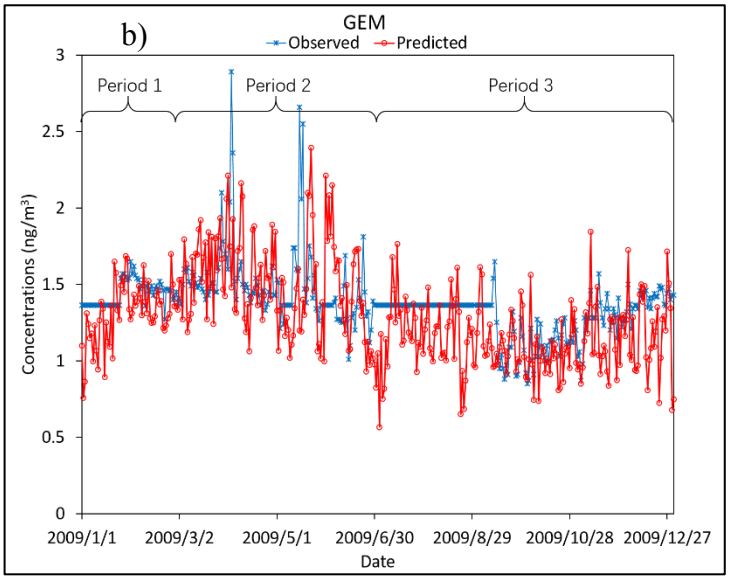
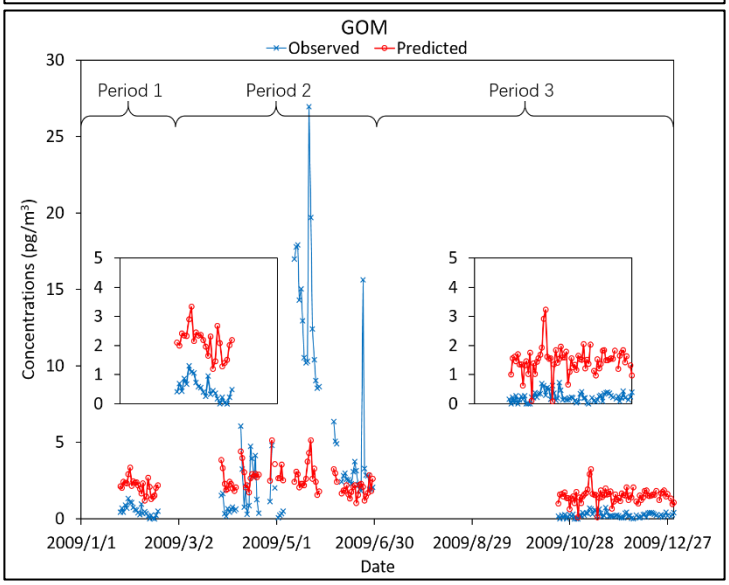
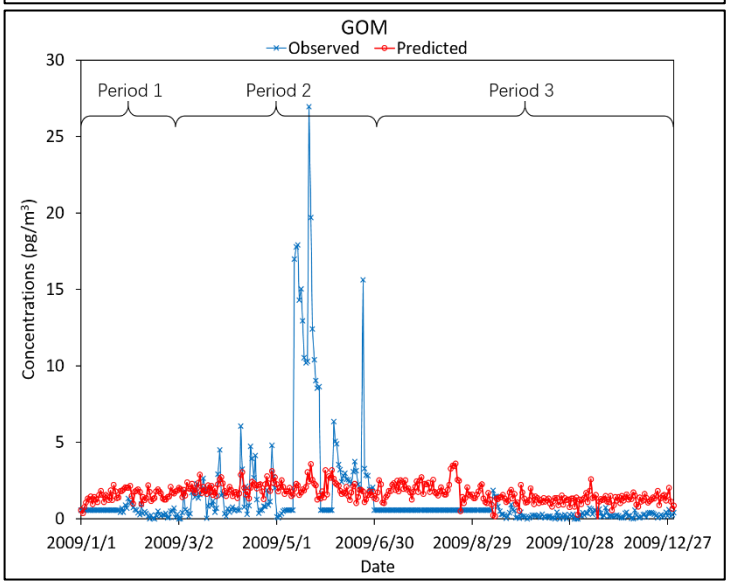


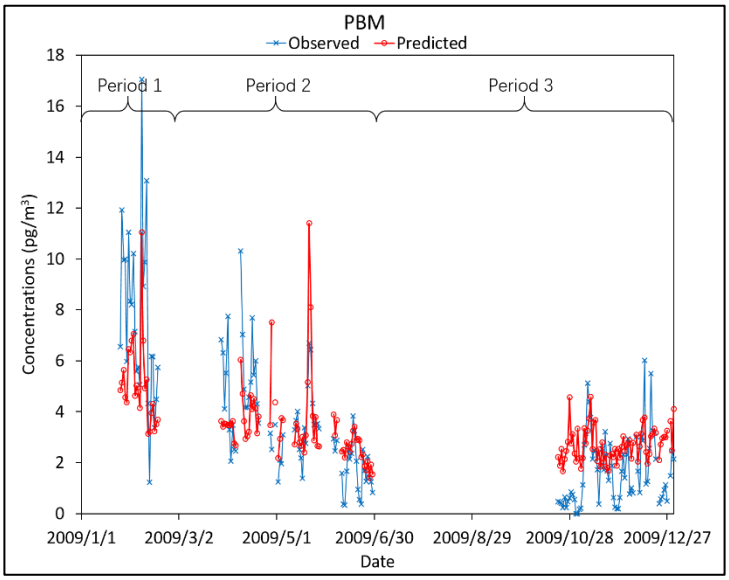
Figure S6. Observation–prediction scatter plot in 2010. a) Case 2010, b) Case 10+mean, c) Case 10+median, d) Case 10+RM, e) Case 10-RM, and f) Case 10ScaleRM, observed GOM and PBM have been scaled.

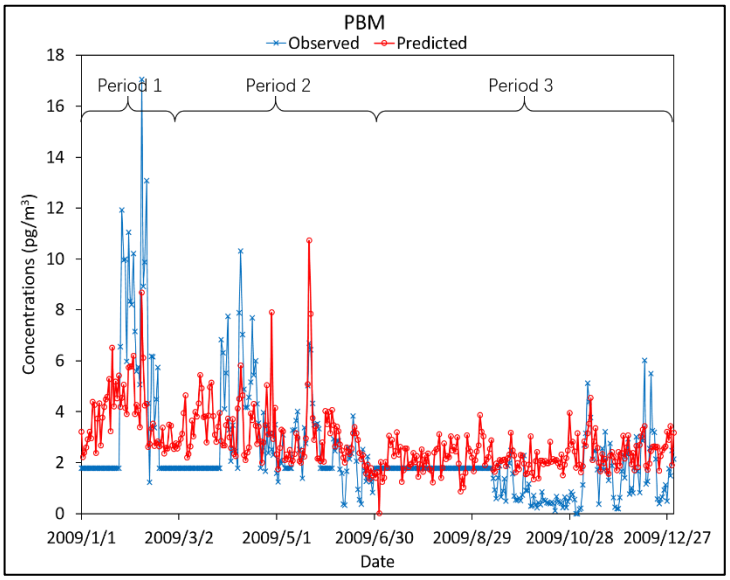

a)
GEM
Observed
Predicted
Period 1
Period 2
Period 3
Concentrations (ng/m³)
Date
2009/1/1
2009/3/2
2009/5/1
2009/6/30
2009/8/29
2009/10/28
2009/12/27

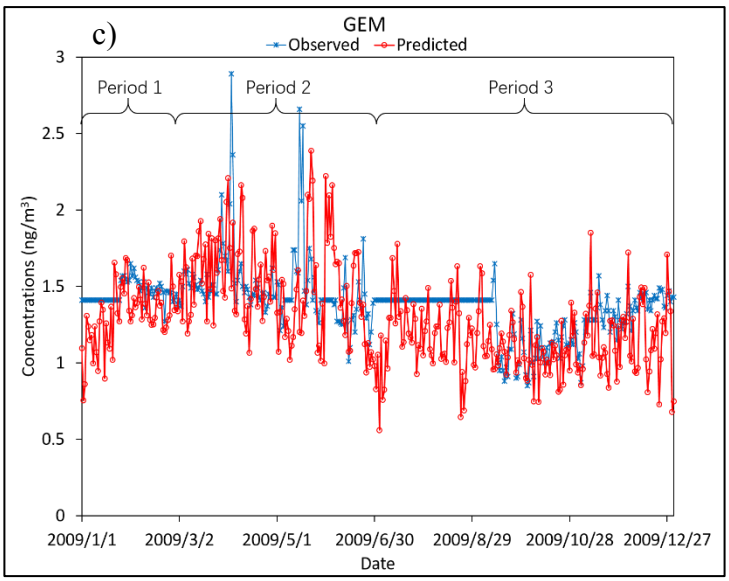

b)
GEM
Observed
Predicted
Period 1
Period 2
Period 3
Concentrations (ng/m³)
Date
2009/1/1
2009/3/2
2009/5/1
2009/6/30
2009/8/29
2009/10/28
2009/12/27

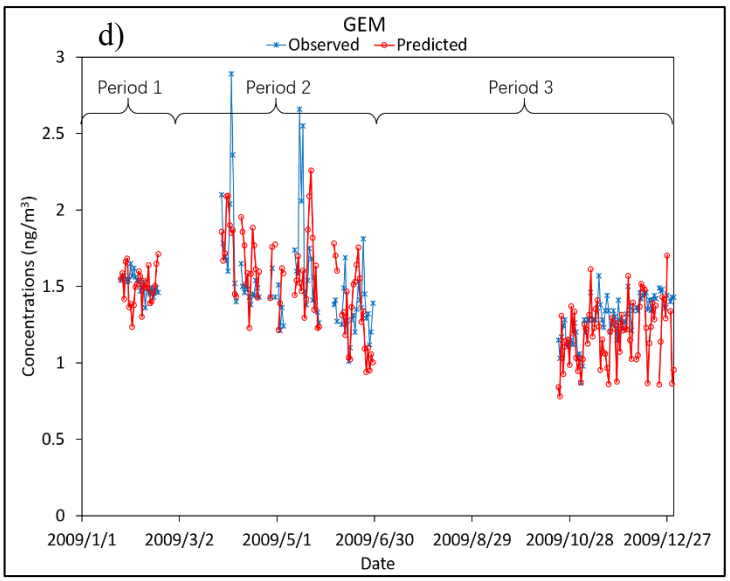

GOM
Observed
Predicted
Period 1
Period 2
Period 3
Concentrations (pg/m³)
Date
2009/1/1
2009/3/2
2009/5/1
2009/6/30
2009/8/29
2009/10/28
2009/12/27

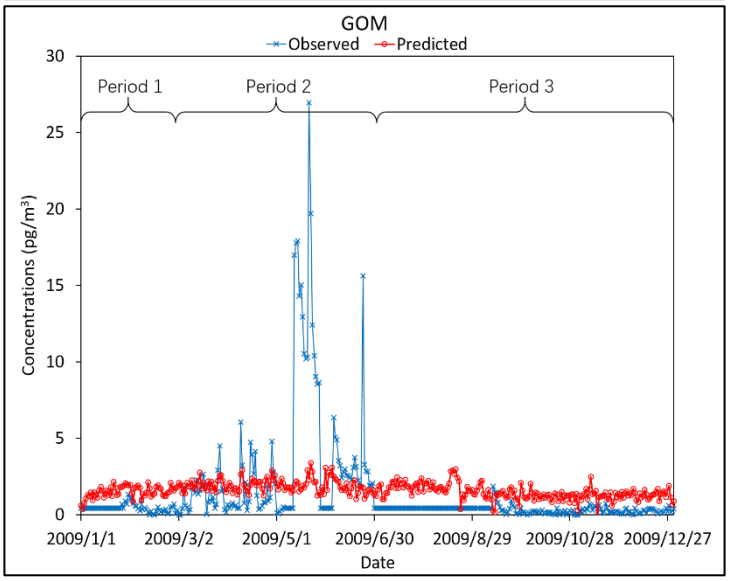

GOM
Observed
Predicted
Period 1
Period 2
Period 3
Concentrations (pg/m³)
Date
2009/1/1
2009/3/2
2009/5/1
2009/6/30
2009/8/29
2009/10/28
2009/12/27

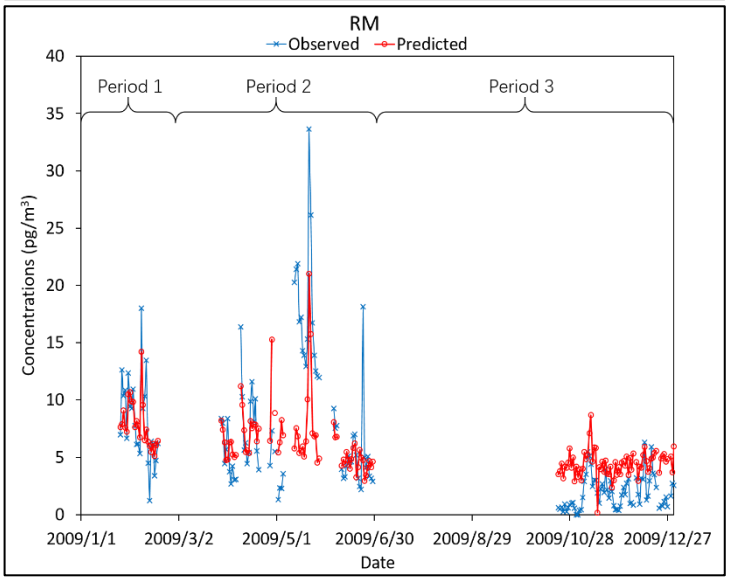

PBM
Observed
Predicted
Period 1
Period 2
Period 3
Concentrations (pg/m³)
Date
2009/1/1
2009/3/2
2009/5/1
2009/6/30
2009/8/29
2009/10/28
2009/12/27

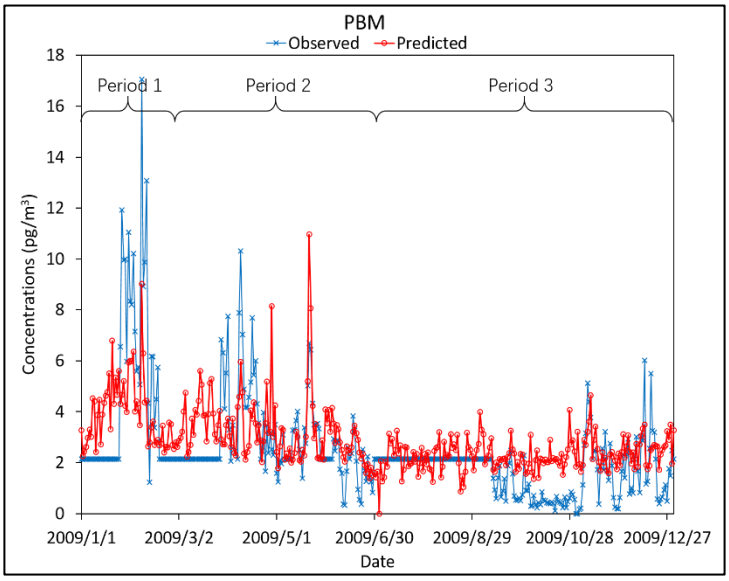

PBM
Observed
Predicted
Period 1
Period 2
Period 3
Concentrations (pg/m³)
Date
2009/1/1
2009/3/2
2009/5/1
2009/6/30
2009/8/29
2009/10/28
2009/12/27

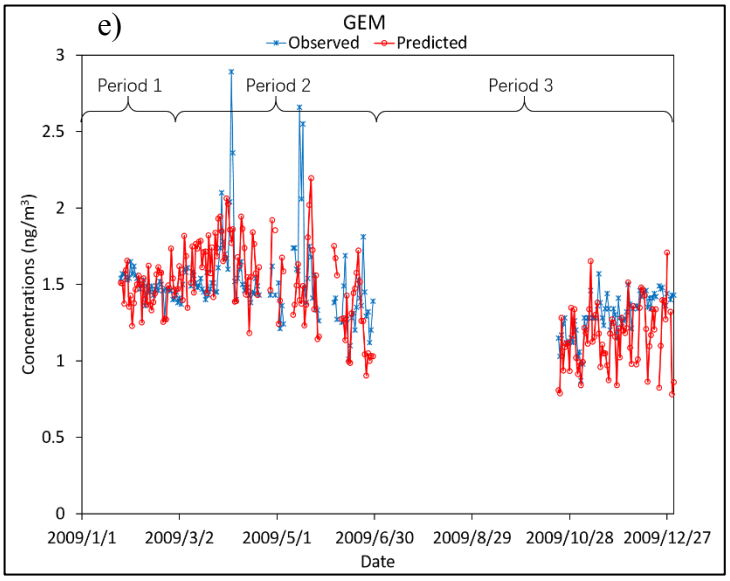

c)
GEM
Observed
Predicted
Period 1
Period 2
Period 3
Concentrations (ng/m³)
Date
2009/1/1
2009/3/2
2009/5/1
2009/6/30
2009/8/29
2009/10/28
2009/12/27


d)
GEM
Observed
Predicted
Period 1
Period 2
Period 3
Concentrations (ng/m³)
Date
2009/1/1
2009/3/2
2009/5/1
2009/6/30
2009/8/29
2009/10/28
2009/12/27


GOM
Observed
Predicted
Period 1
Period 2
Period 3
Concentrations (pg/m³)
Date
2009/1/1
2009/3/2
2009/5/1
2009/6/30
2009/8/29
2009/10/28
2009/12/27


RM
Observed
Predicted
Period 1
Period 2
Period 3
Concentrations (pg/m³)
Date
2009/1/1
2009/3/2
2009/5/1
2009/6/30
2009/8/29
2009/10/28
2009/12/27


PBM
Observed
Predicted
Period 1
Period 2
Period 3
Concentrations (pg/m³)
Date
2009/1/1
2009/3/2
2009/5/1
2009/6/30
2009/8/29
2009/10/28
2009/12/27


e)
GEM
Observed
Predicted
Period 1
Period 2
Period 3
Concentrations (ng/m³)
Date
2009/1/1
2009/3/2
2009/5/1
2009/6/30
2009/8/29
2009/10/28
2009/12/27

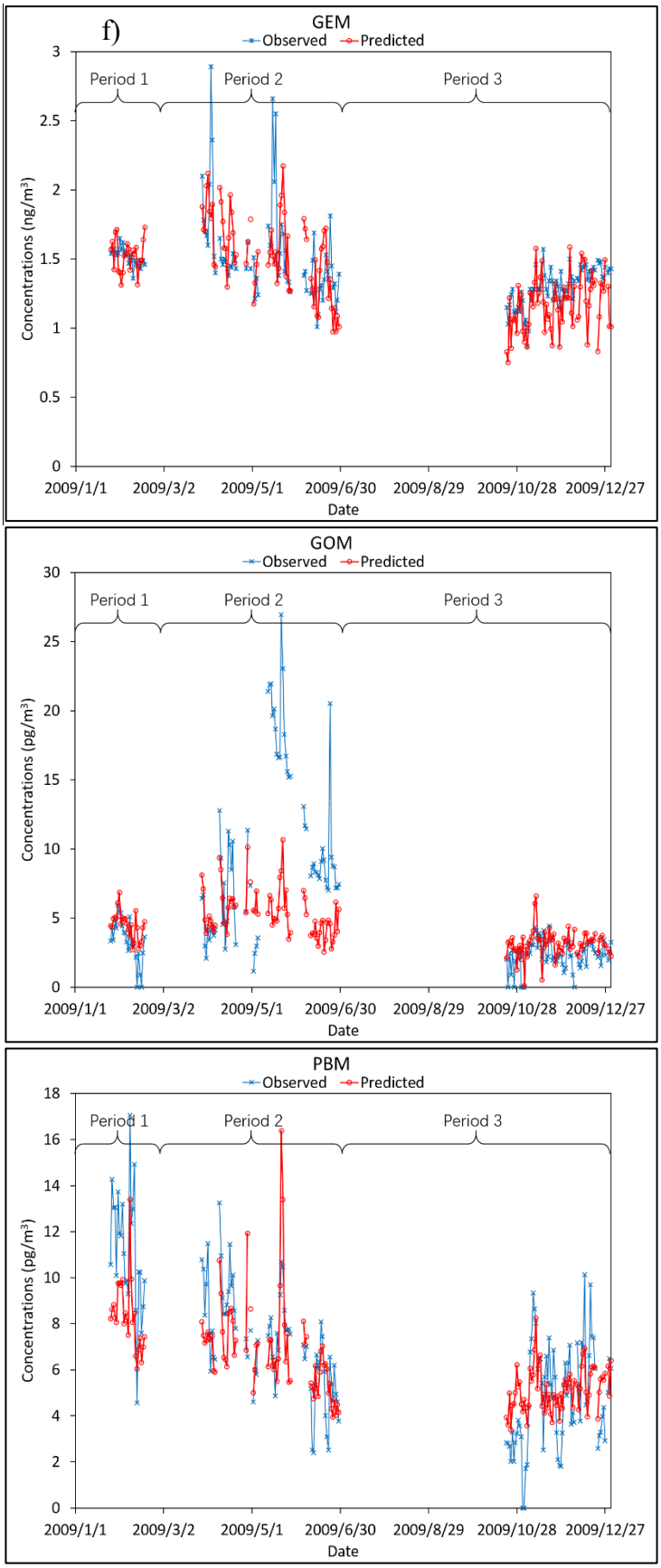
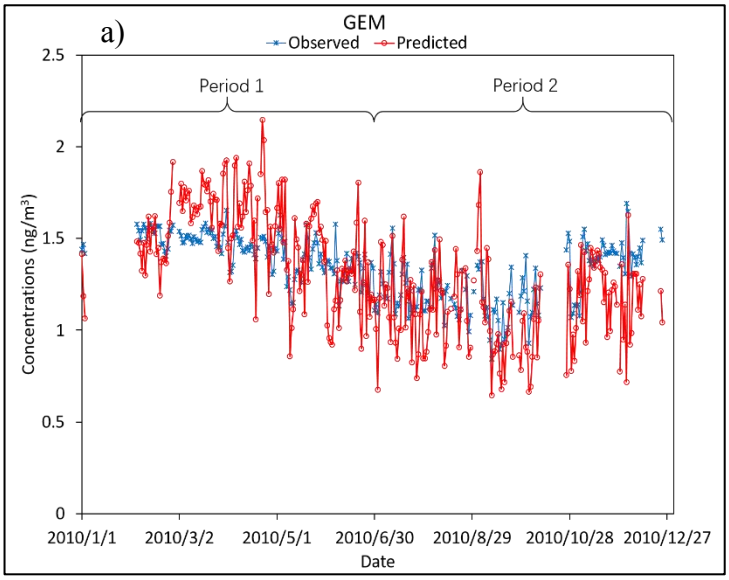
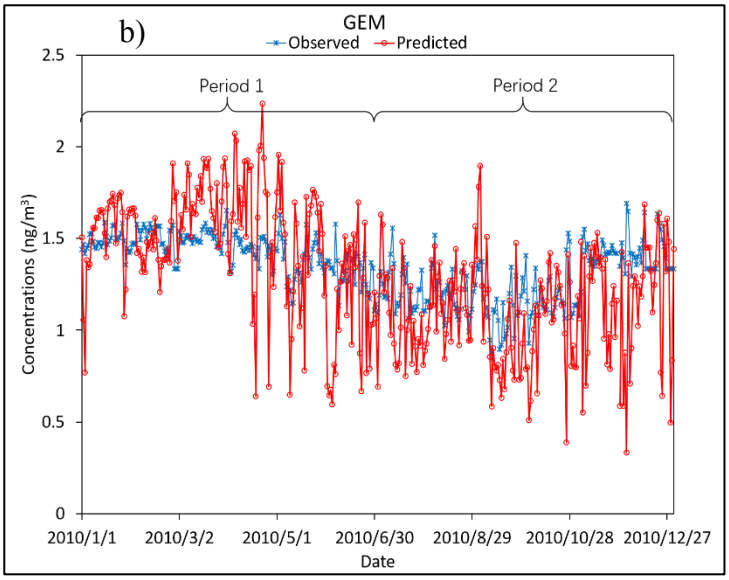
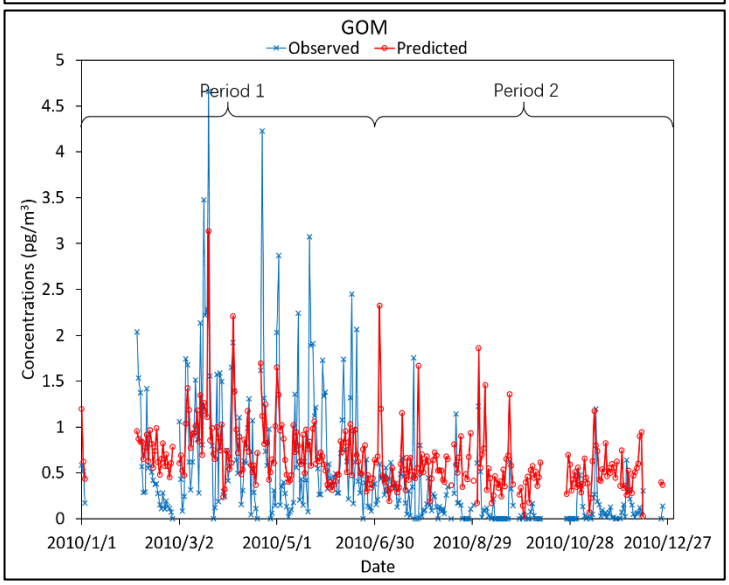
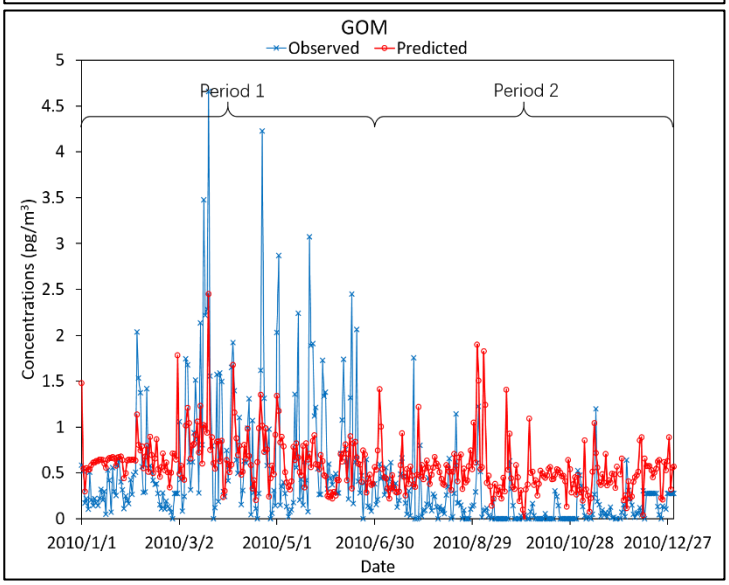


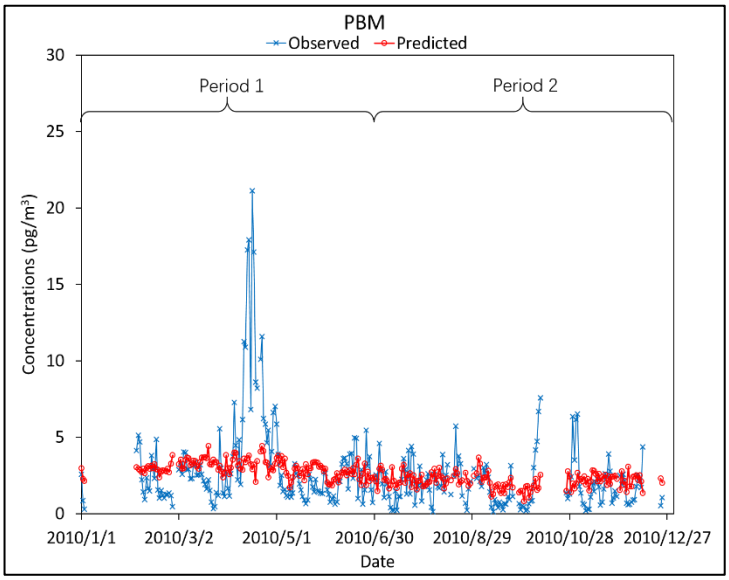
Figure S7. Observation–prediction time series in 2009. a) Case 2009, b) Case 09+mean, c) Case 09+median, d) Case 09+RM, e) Case 09-RM, and f) Case 09ScaleRM, observed GOM and PBM have been scaled.

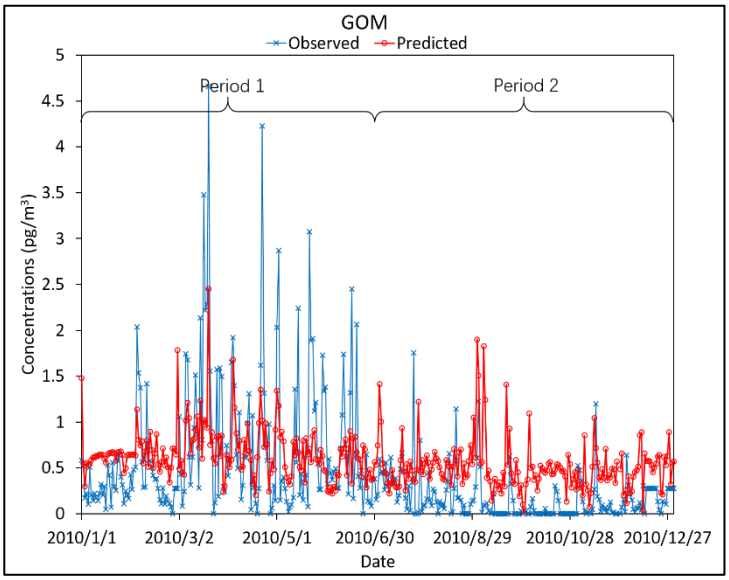

a)
GEM
Observed
Predicted
Period 1
Period 2
Concentrations (ng/m³)
Date
2010/1/1
2010/3/2
2010/5/1
2010/6/30
2010/8/29
2010/10/28
2010/12/27

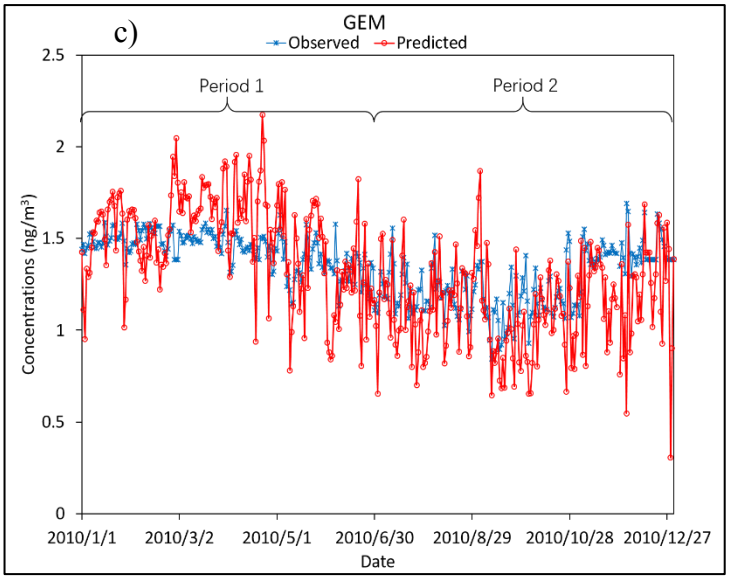

b)
GEM
Observed
Predicted
Period 1
Period 2
Concentrations (ng/m³)
Date
2010/1/1
2010/3/2
2010/5/1
2010/6/30
2010/8/29
2010/10/28
2010/12/27

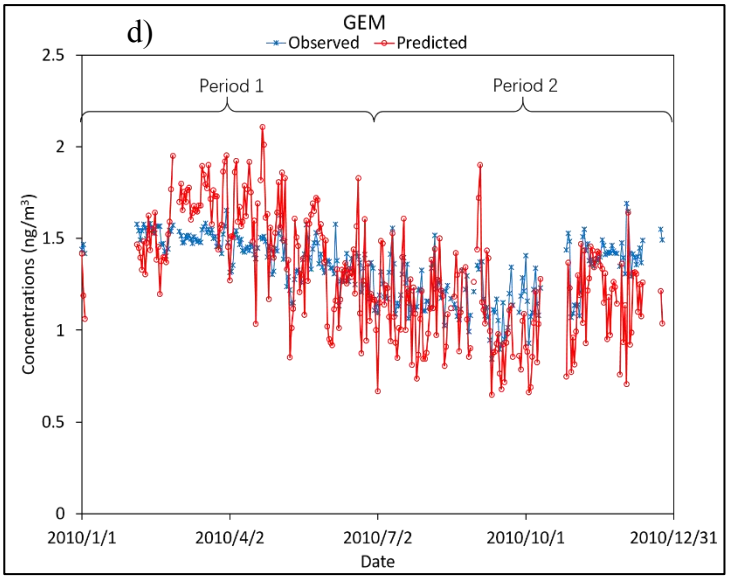

GOM
Observed
Predicted
Period 1
Period 2
Concentrations (pg/m³)
Date
2010/1/1
2010/3/2
2010/5/1
2010/6/30
2010/8/29
2010/10/28
2010/12/27

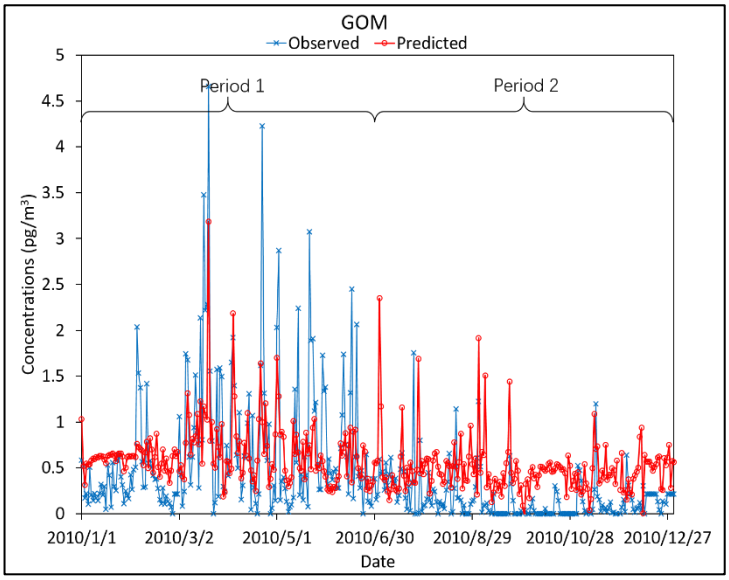

GOM
Observed
Predicted
Period 1
Period 2
Concentrations (pg/m³)
Date
2010/1/1
2010/3/2
2010/5/1
2010/6/30
2010/8/29
2010/10/28
2010/12/27

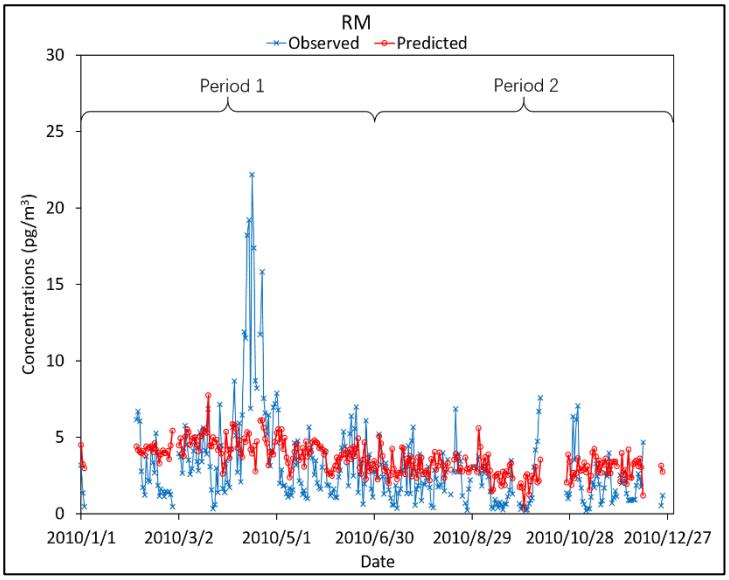

PBM
Observed
Predicted
Period 1
Period 2
Concentrations (pg/m³)
Date
2010/1/1
2010/3/2
2010/5/1
2010/6/30
2010/8/29
2010/10/28
2010/12/27

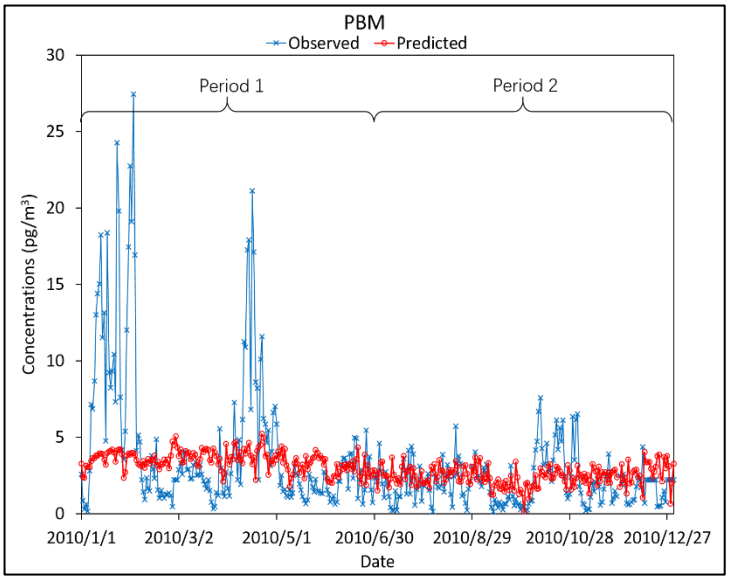

GOM
Observed
Predicted
Period 1
Period 2
Concentrations (pg/m³)
Date
2010/1/1
2010/3/2
2010/5/1
2010/6/30
2010/8/29
2010/10/28
2010/12/27

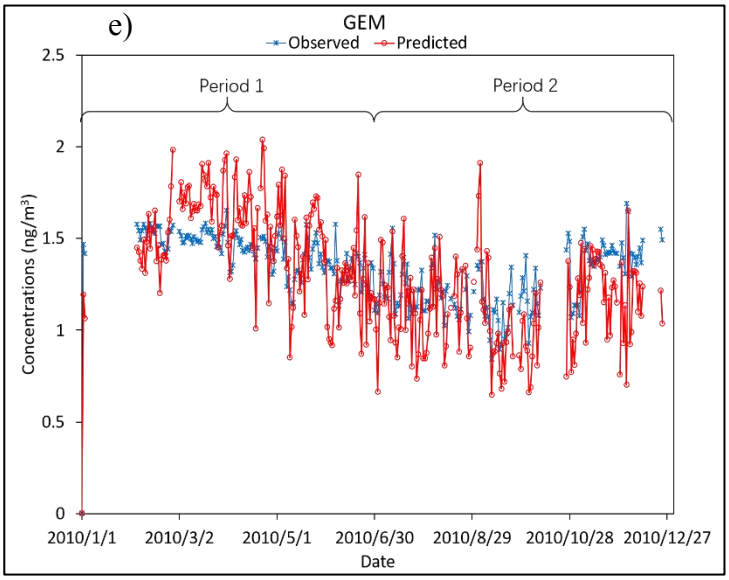

c)
GEM
Observed
Predicted
Period 1
Period 2
Concentrations (ng/m³)
2.5
2
1.5
1
0.5
0
2010/1/1
2010/3/2
2010/5/1
2010/6/30
2010/8/29
2010/10/28
2010/12/27
Date


d)
GEM
Observed
Predicted
Period 1
Period 2
Concentrations (ng/m³)
2.5
2
1.5
1
0.5
0
2010/1/1
2010/4/2
2010/7/2
2010/10/1
2010/12/31
Date


GOM
Observed
Predicted
Period 1
Period 2
Concentrations (pg/m³)
5
4.5
4
3.5
3
2.5
2
1.5
1
0.5
0
2010/1/1
2010/3/2
2010/5/1
2010/6/30
2010/8/29
2010/10/28
2010/12/27
Date


RM
Observed
Predicted
Period 1
Period 2
Concentrations (pg/m³)
30
25
20
15
10
5
0
2010/1/1
2010/3/2
2010/5/1
2010/6/30
2010/8/29
2010/10/28
2010/12/27
Date


PBM
Observed
Predicted
Period 1
Period 2
Concentrations (pg/m³)
30
25
20
15
10
5
0
2010/1/1
2010/3/2
2010/5/1
2010/6/30
2010/8/29
2010/10/28
2010/12/27
Date


e)
GEM
Observed
Predicted
Period 1
Period 2
Concentrations (ng/m³)
2.5
2
1.5
1
0.5
0
2010/1/1
2010/3/2
2010/5/1
2010/6/30
2010/8/29
2010/10/28
2010/12/27
Date

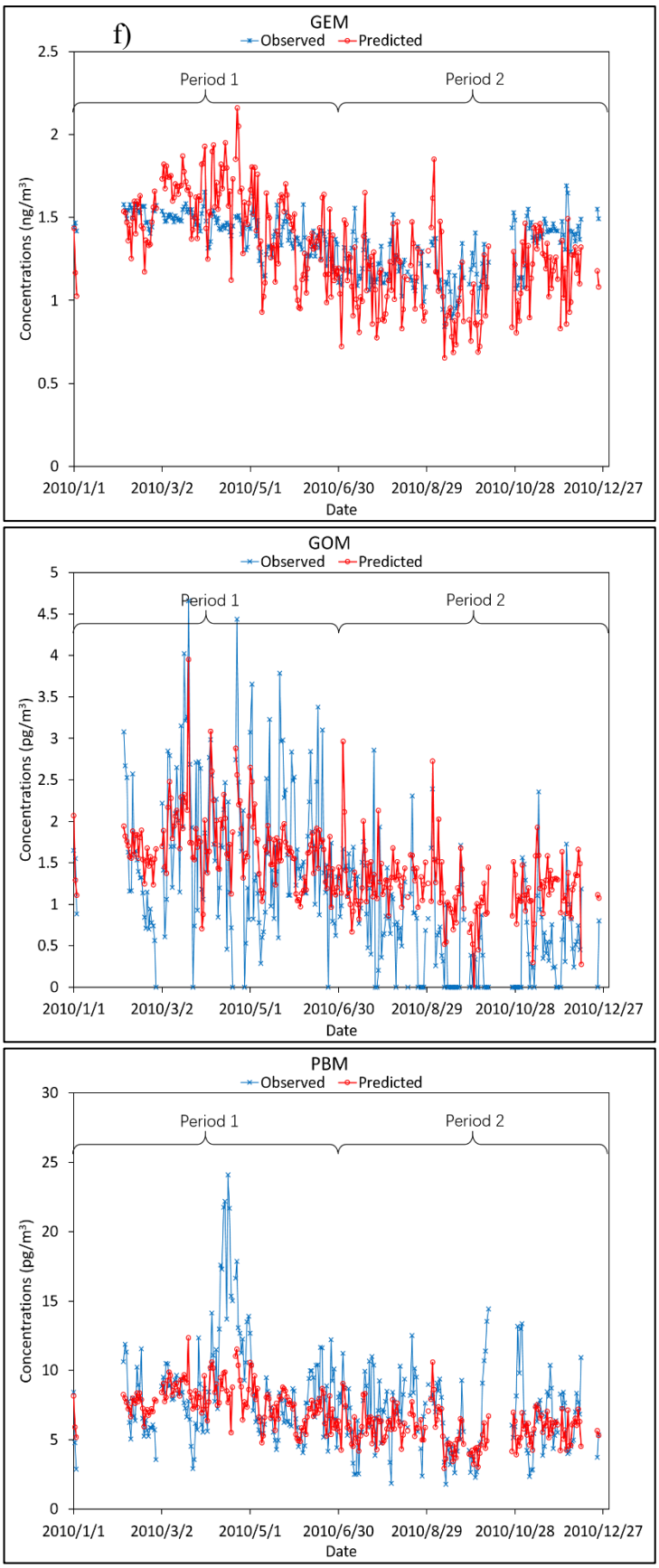


Figure S8. Observation–prediction time series in 2010. a) Case 2010, b) Case 10+mean, c) Case 10+median, d) Case 10+RM, e) Case 10-RM, and f) Case 10ScaleRM, observed GOM and PBM have been scaled.