

Supplementary Information

Potential sources and processes affecting speciated atmospheric mercury at Kejimikujik National Park, Canada

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Table S1. Emission of Hg and other pollutants reported in NPRI within 150 Km of the sampling site (Data source: Environmental Canada, 2016).

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°N, 64°W)	50 Km southeast	0	0	9.9	26	309	259	0	0
Michelin North America (Canada)	Bridgewater (44°N, 64°W)	53 Km east	0	0	195	184	68	63	0	0
High Liner Foods INC.	Lunenburg (44°N, 64°W)	72 Km east	0	0	27	27	0	0	0	0
Department of National Defence	Greenwood (44°N, 65°W)	75 Km north	0	0	55	68	19	18	0	0
Louisiana-Pacific Canada LTD.	East River (44°N, 64°W)	88 Km northeast	0	0	122	102	100	99	0	0
Maple Leaf Foods/Larsen Packers Limiter	Berwick (45°N, 64°W)	89 Km northeast	0	0	51	38	0	0	0	0
Michelin North America (Canada)	Waterville (45°N, 64°W)	92 Km northeast	0	0	162	182	57	62	0	0
Acadia University/Acadia Campus	Wolfville (45°N, 64°W)	108 Km northeast	0	0	77	73	27	26	0	0
CKF INC.	Hantsport (45°N, 64°W)	116 Km northeast	0	0	66	57	21	0	0	0
Minas Basin Pulp and Power Company Limited	Hantsport (45°N, 64°W)	116 Km northeast	0	0	225	260	66	76	0	0
Mount Saint Vincent University	Halifax (44°N, 63°W)	129 Km northeast	0	0	27	13	7.2	3.9	0	0
Department of National Defence – CFB Halifax – Canadian Forces Ammunition Depot Bedford	Bedford (44°N, 63°W)	131 Km northeast	0	0	56	50	0	0	0	0
Department of National Defence – CFB Halifax – Windsor Park	Halifax (44°N, 63°W)	132 Km northeast	0	0	59	44	36	30	0	0

Dalhousie University	Halifax (44°N, 63°W)	133 Km northeast	0.18	0.15	253	260	89	72	0	0
Department of National Defence/CFB Halifax -Stadacona/Dockyard	Halifax (44°N, 63°W)	133 Km northeast	0	0	211	177	58	51	0	0
Capital Health – Camp Hill Site Central Heating Plant	Halifax (44°N, 63°W)	133 Km northeast	0	0	15	12	14	20	0	0
Saint Mary's University - Halifax	Halifax (44N, 63W)	133 Km northeast	0	0	1.2	0	3	0	0	0
Oland Brewery/Main Plant	Halifax (44°N, 63°W)	133 Km northeast	0	0	31	0	0	0	0	0
Nova Scotia Power Incorporated/Tufts Cove Generating Station	Dartmouth (44°N, 63°W)	134 Km northeast	0	0	2205	2205	3054	3054	0	0
Nova Scotia Power Incorporated – Burnside Combustion Turbines	Dartmouth (44°N, 63°W)	134 Km northeast	0	0	0	0	60	40	0	0
Maritime Paper Products LTD.	Dartmouth (44°N, 63°W)	134 Km northeast	0	0	7.2	0.868	3.1	2.1	0	0
Capital Health – Victoria General Hospital Central Heating Plant	Halifax (44°N, 63°W)	134 Km northeast	0	0	215	7.6	60	19	0	0
Capital Health – Nova Scotia Hospital Central Heating Plant	Halifax (44°N, 63°W)	136 Km northeast	0	0	3.3	1.1	9.3	8.7	0	0
Imperial oil-Darhmouth Refinery	Dartmouth (44°N, 63°W)	137Km northeast	2.6	2.9	4231	3073	1543	1251	0.593	2.2
Department of National Defence – 12 Wing Shearwater	Shearwater (44°N, 63°W)	138 Km northeast	0	0	150	127	43	38	0	0
Total emission			2.78	3.05	8,455	6,988	5,647	5,193	0.593	2.2
Provincial total emission			147.5	90.3	108,961	70,336	22,235	22,157	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 predicted 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 predicted to observed annual mean:					0.97
GOM (%)	Min	0	0	0	0
	Max	100	0	100	0
	Average	25	0	70	0
	Median	19	0	78	0
Ratio of predicted to observed annual mean:					0.86
PBM (%)	Min	0	0	0	0
	Max	0	93	100	92
	Average	0	21	69	9
	Median	0	14	74	5
Ratio of predicted 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 predicted 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 predicted 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 predicted 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 predicted 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 predicted 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 predicted 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	
	Max	16	54	98	
	Average	3	5	78	
	Median	2	2	83	
Ratio of predicted to observed annual mean:					0.97
RM (%)	Min	0	0	0	
	Max	37	83	100	
	Average	10	16	73	
	Median	7	11	78	
Ratio of predicted 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	
	Max	34	15	98	
	Average	7	1	79	
	Median	4	1	84	
Ratio of predicted 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	
	Max	0	65	100	
	Average	0	7	78	
	Median	0	4	83	
Ratio of predicted to observed annual mean:					0.97
GOM (%)	Min	0	0	0	
	Max	97	0	100	
	Average	23	0	75	
	Median	18	0	81	
Ratio of predicted to observed annual mean:					0.75

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

Table S5. PMF factor contributions to speciated Hg and ratios of predicted 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 predicted 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 predicted 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 predicted 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	28	70	98	100
	Average	3	6	83	8
	Median	2	3	88	4
Ratio of predicted to observed annual mean:					0.96
GOM (%)	Min	0	0	0	0
	Max	22	100	100	0
	Average	2	28	70	0
	Median	1	22	76	0
Ratio of predicted to observed annual mean:					1.35
PBM (%)	Min	0	0	0	0
	Max	0	85	100	99
	Average	0	4	93	2
	Median	0	2	97	1
Ratio of predicted 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	0	100	100
	Average	3	0	88	8
	Median	2	0	93	4

Ratio of predicted to observed annual mean:					0.97	
GOM (%)	Factor	Min	0	0	0	0
	Max	0	100	100	100	0
	Average	0	36	64	64	0
	Median	0	38	62	62	0
Ratio of predicted to observed annual mean:					1.32	
PBM (%)	Factor	Min	0	0	0	0
	Max	0	0	100	100	100
	Average	0	0	97	97	3
	Median	0	0	99	99	1
Ratio of predicted to observed annual mean:					0.88	

d) Case 10+RM

Ratio of predicted to observed annual mean:						0.98
GEM (%)	Factor	Min	0	0	0	0
	Max	100	9	99	99	100
	Average	11	1	79	79	9
	Median	7	1	85	85	4
Ratio of predicted to observed annual mean:						0.98
RM (%)	Factor	Min	0	0	0	0
	Max	100	47	98	98	96
	Average	9	8	80	80	3
	Median	5	6	86	86	1
Ratio of predicted to observed annual mean:						1.16

e) Case 10-RM

Ratio of predicted to observed annual mean:						0.98
GEM (%)	Factor	Min	0	0	0	0
	Max	100	10	99	99	100
	Average	11	1	78	78	9
	Median	7	1	85	85	4
Ratio of predicted to observed annual mean:						0.98

f) Case 10ScaleRM

Ratio of predicted to observed annual mean:						0.98
GEM (%)	Factor	Min	0	0	0	0
	Max	100	10	99	99	100
	Average	11	1	78	78	9
	Median	7	1	85	85	4
Ratio of predicted to observed annual mean:						0.98
GOM (%)	Factor	Min	0	0	0	0
	Max	100	69	99	99	75
	Average	8	14	77	77	1
	Median	4	11	80	80	0
Ratio of predicted to observed annual mean:						1.23

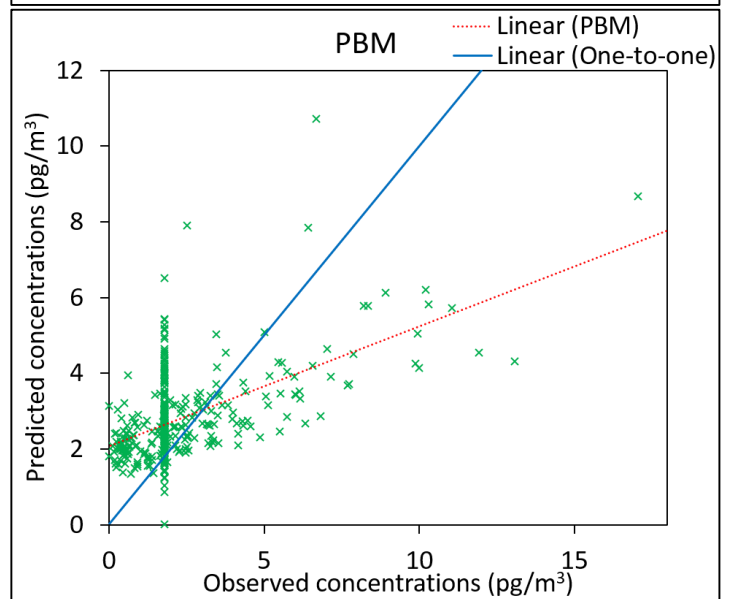
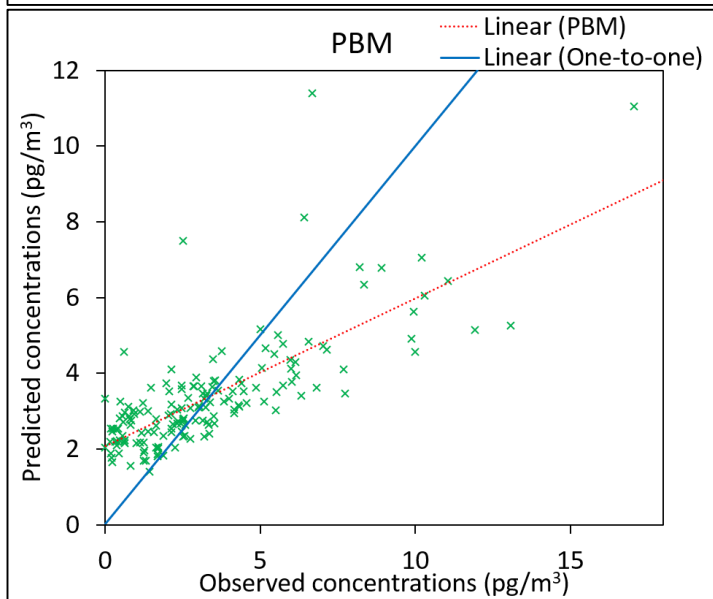
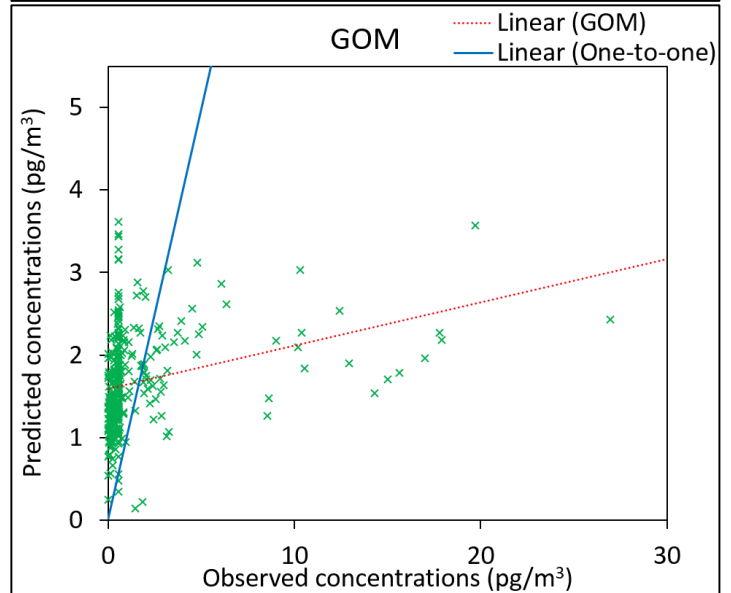
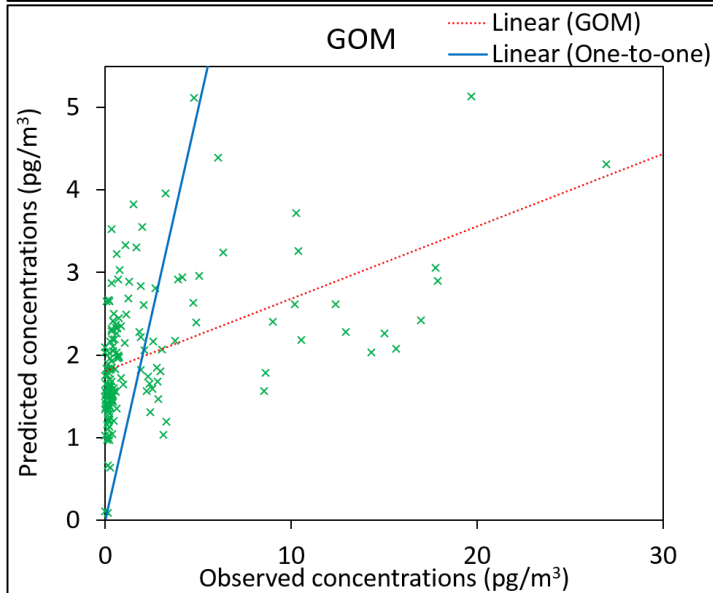
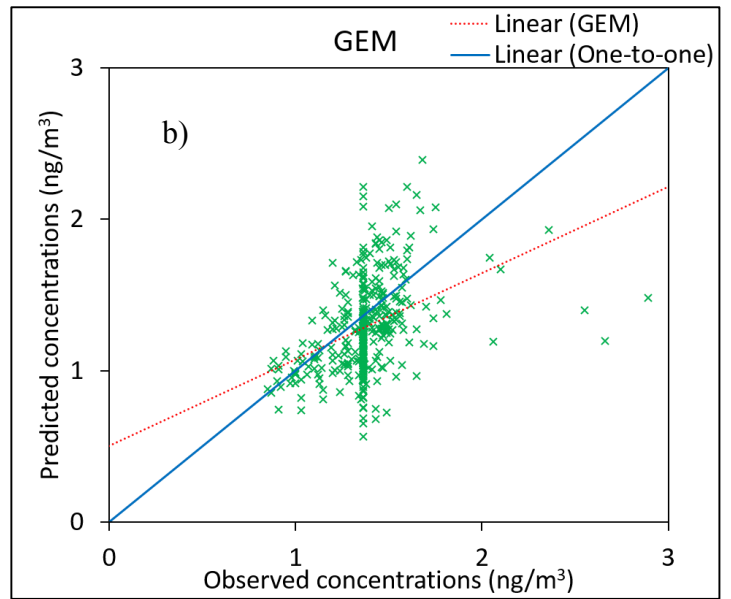
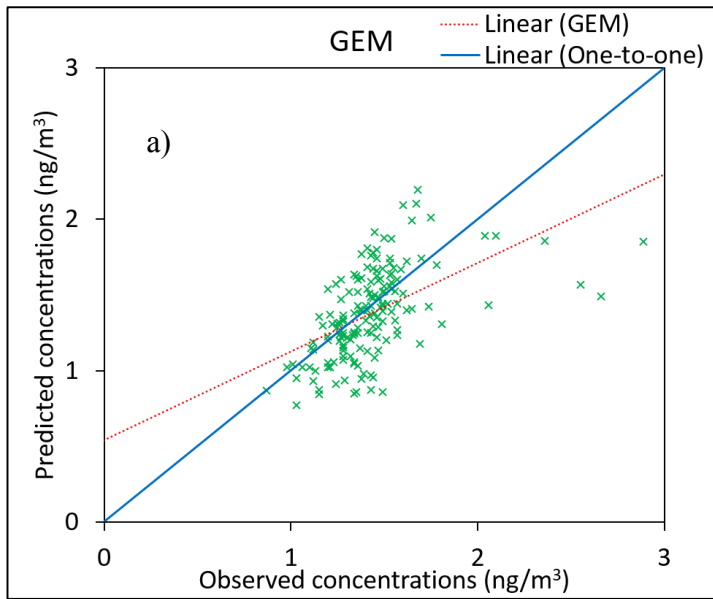
	Min	0	0	0	0
PBM	Max	100	40	97	98
(%)	Average	12	6	76	6
	Median	7	4	82	2
Ratio of predicted to observed annual mean:					0.88

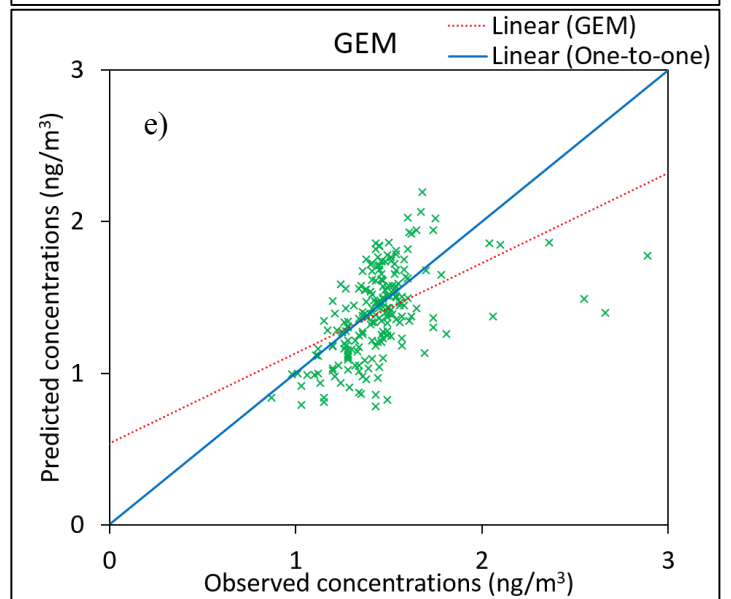
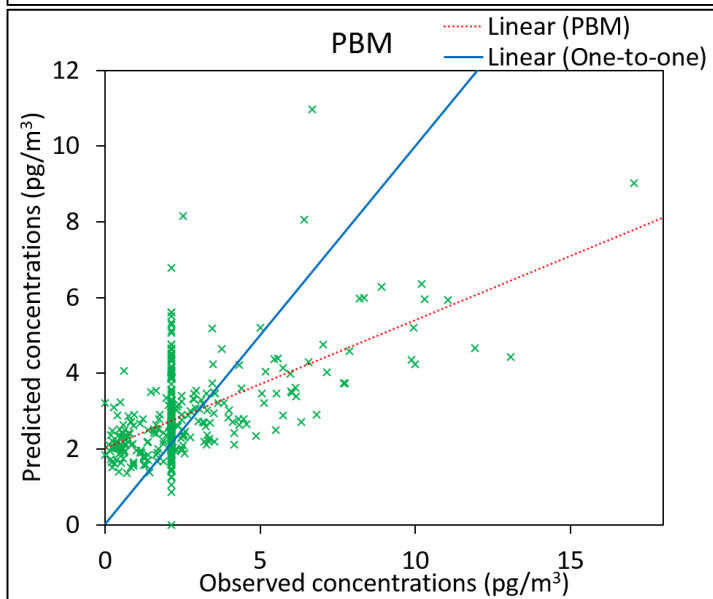
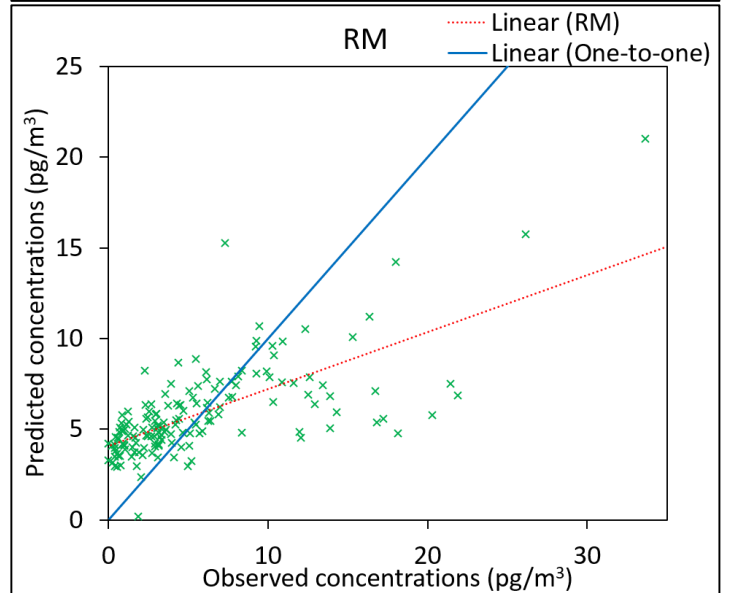
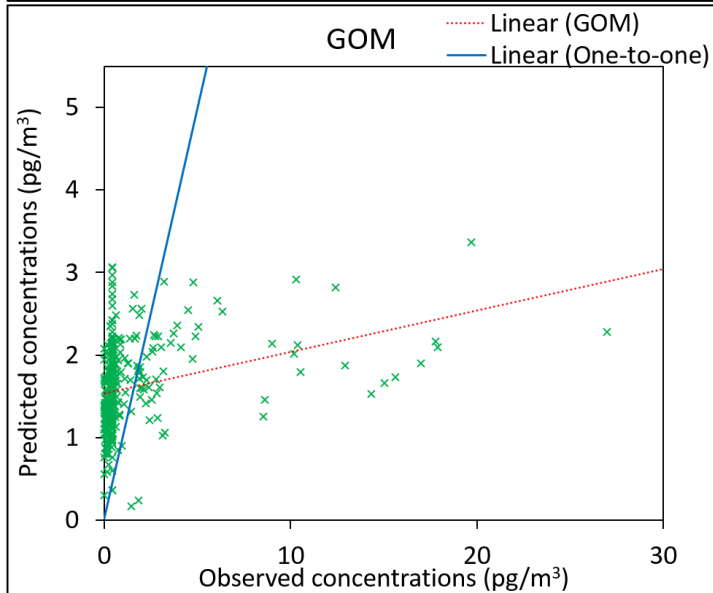
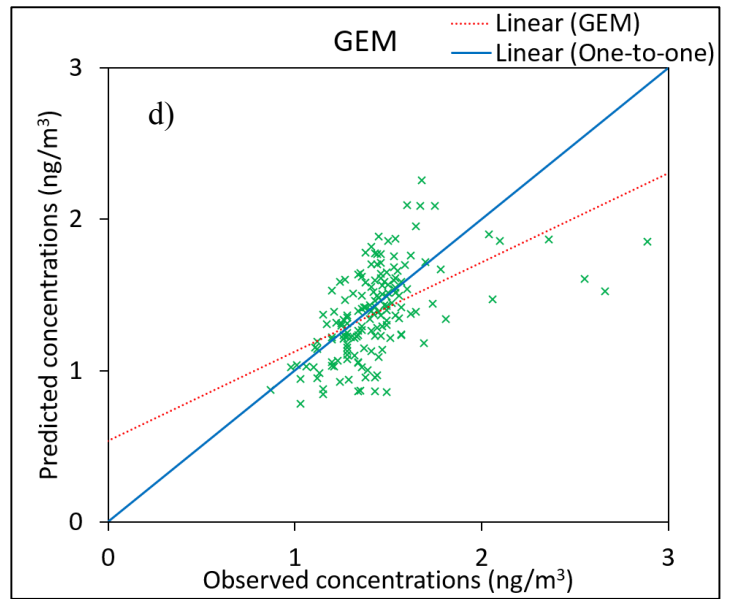
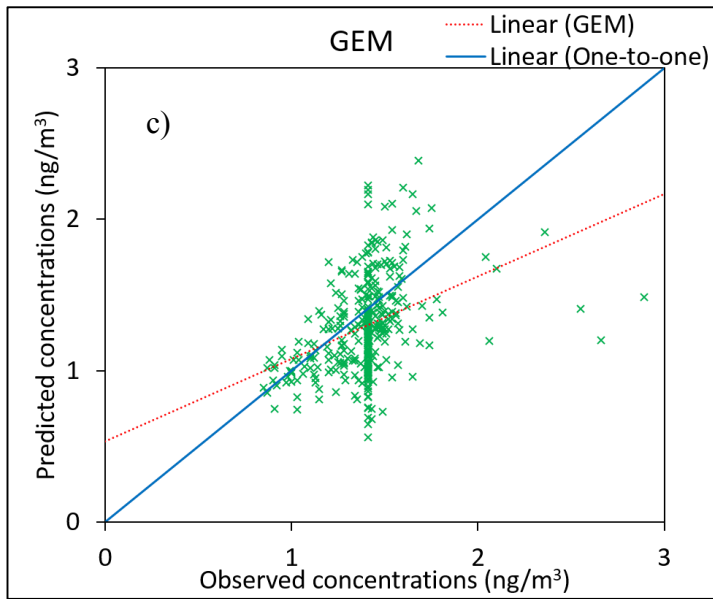
Table S6. Pearson correlation coefficients between Hg forms and other compounds in Case 2009, Case 09+mean, and Case 09+median (bold numbers are significant at <0.05).

	GEM200 9	GEM09+ mean	GEM09+ median	GOM200 9	GOM09+ mean	GOM09+ median	PBM200 9	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 <0.05).

	GEM201 0	GEM10+ mean	GEM10+ median	GOM201 0	GOM10+ mean	GOM10+ median	PBM201 0	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





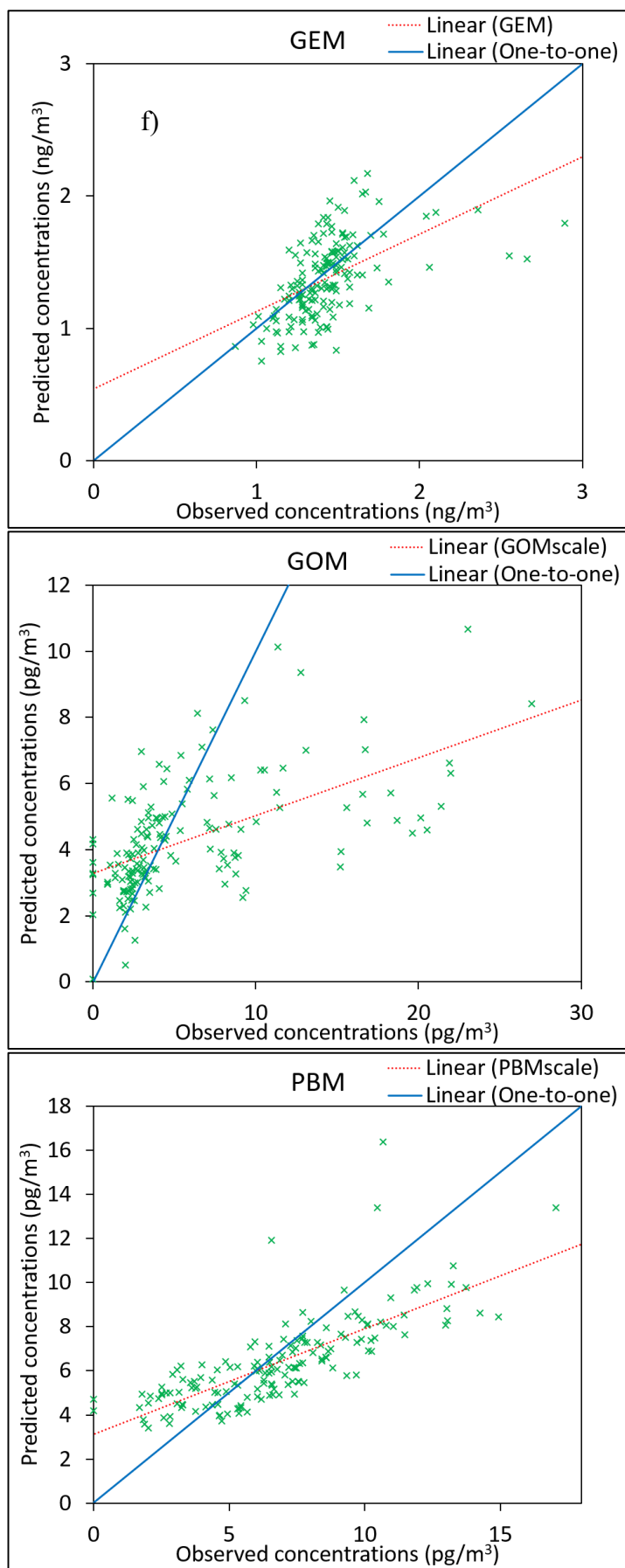
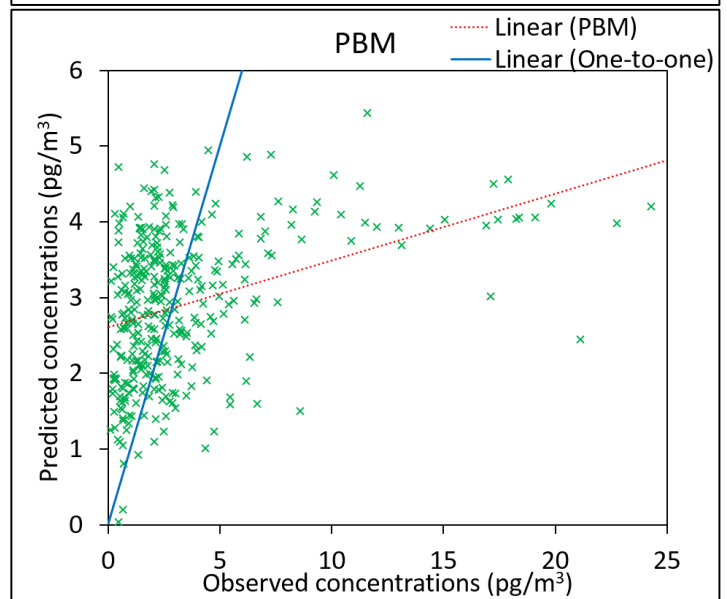
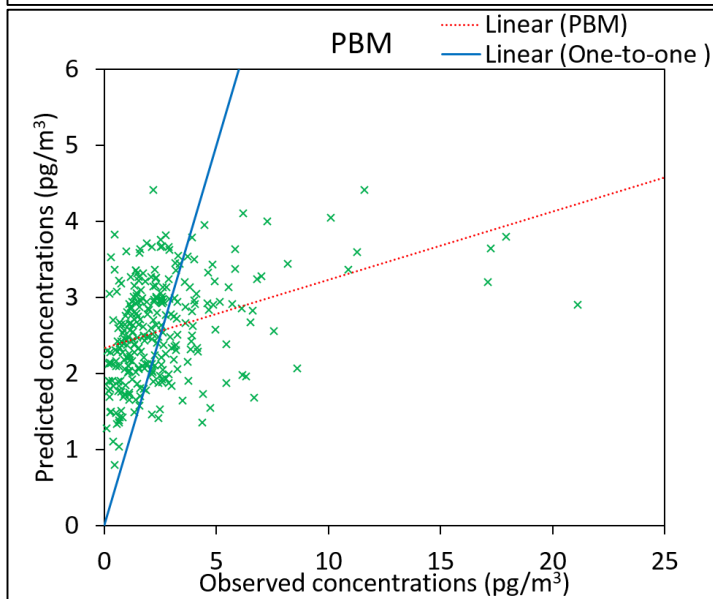
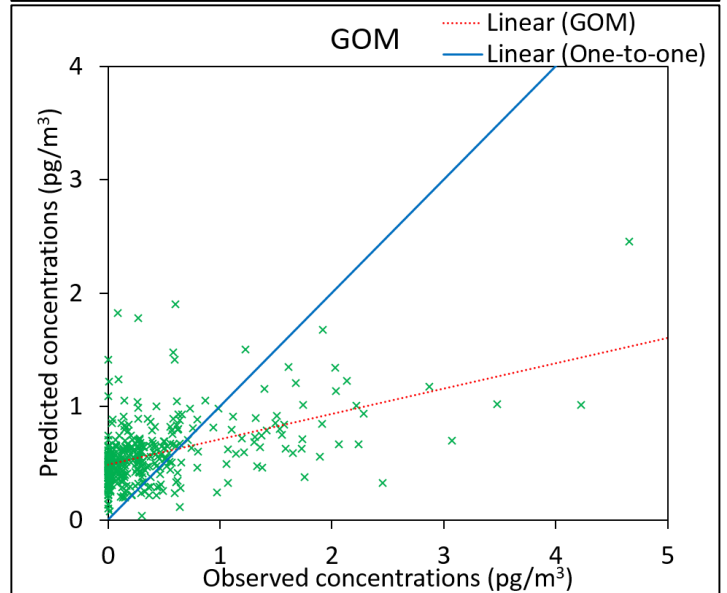
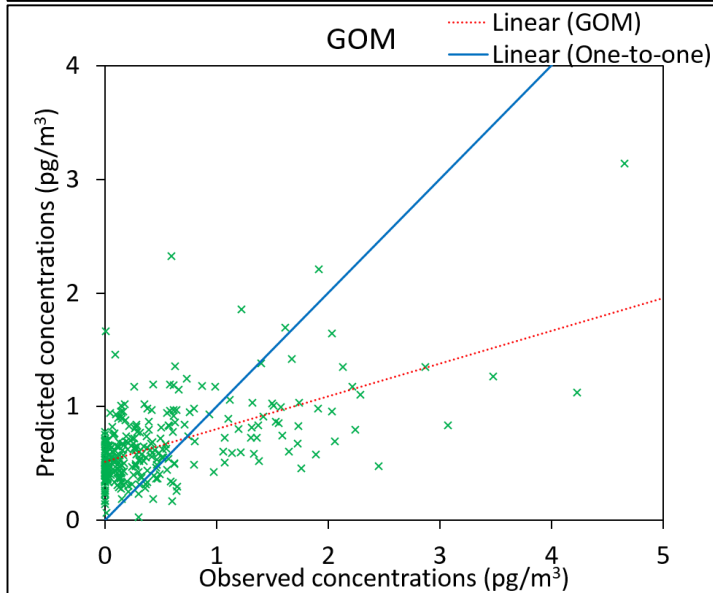
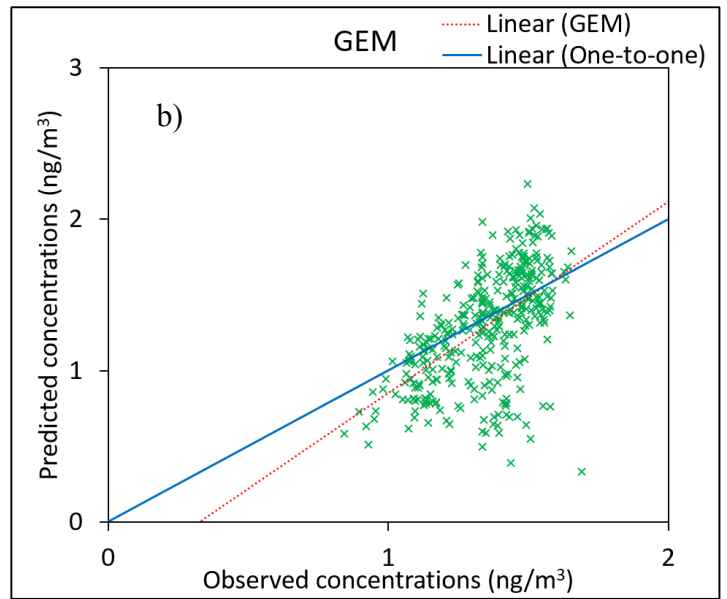
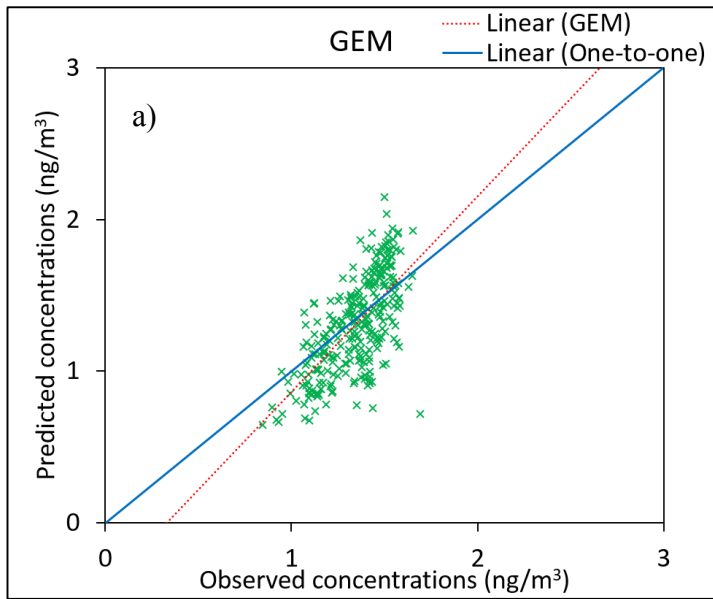
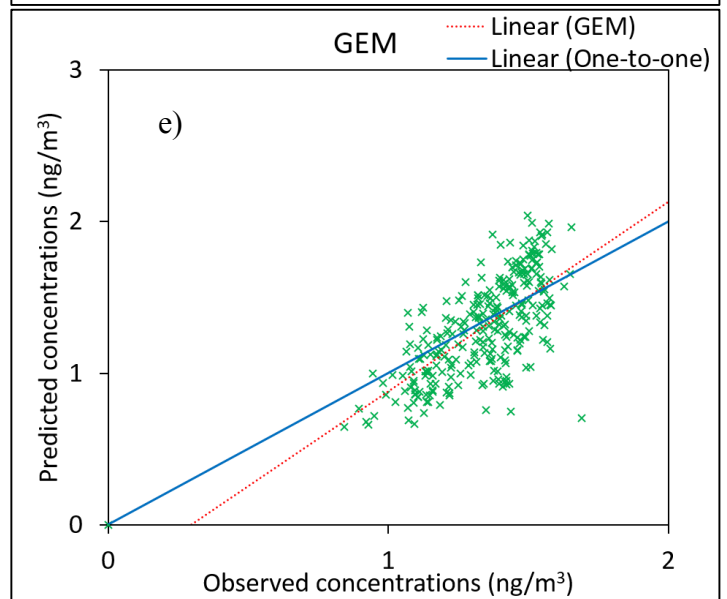
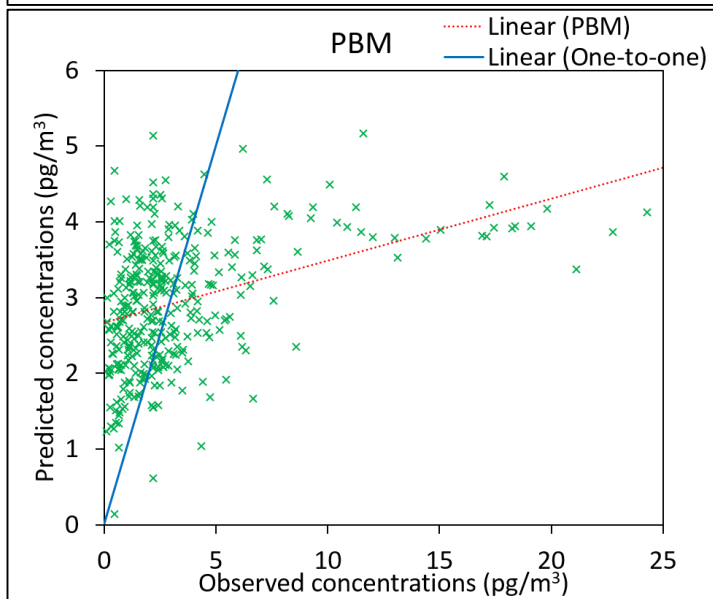
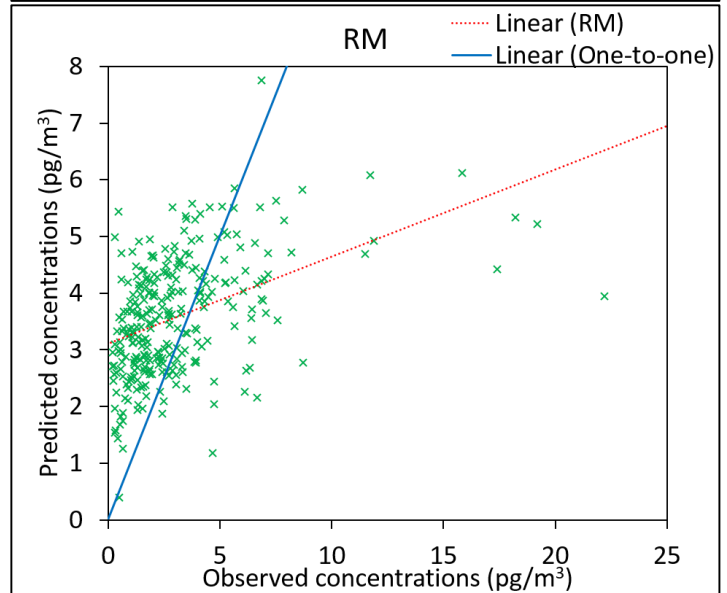
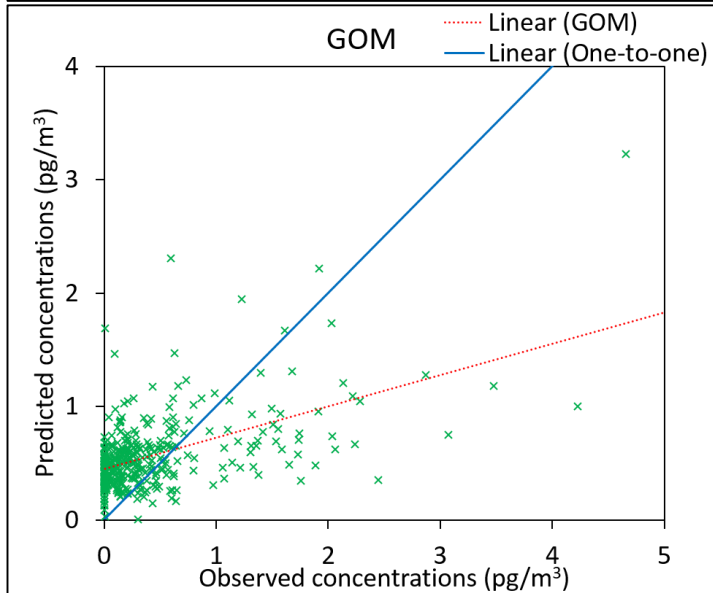
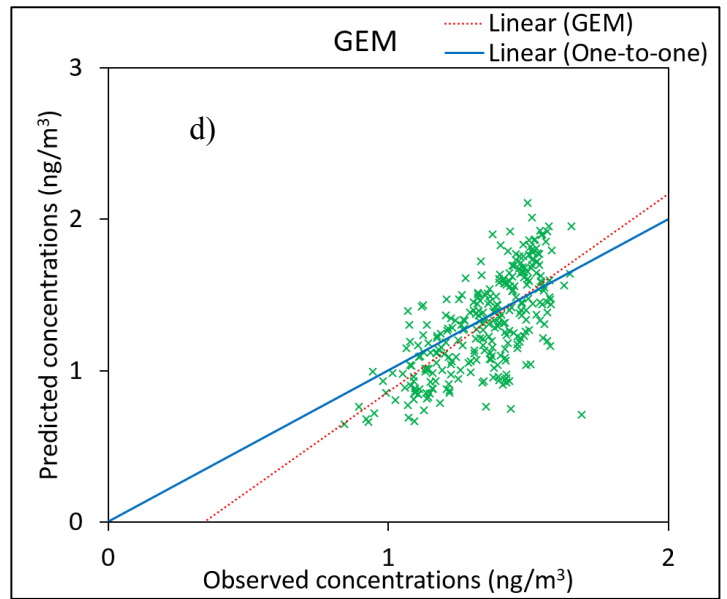
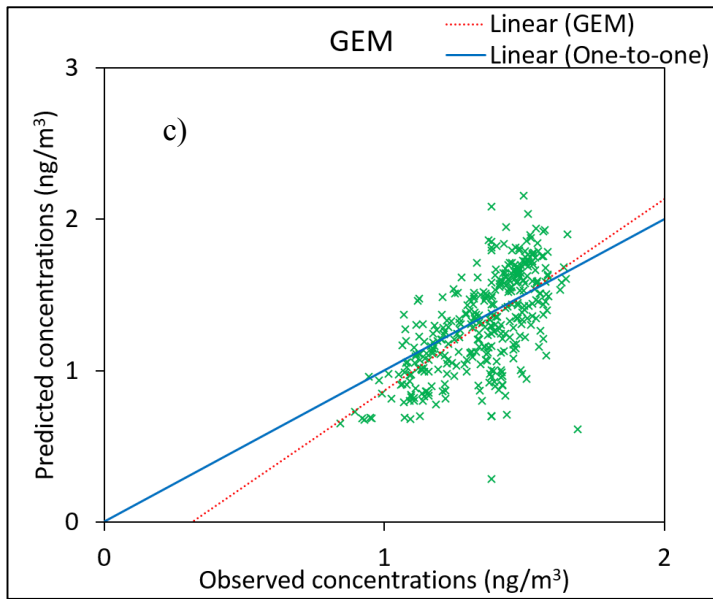


Figure S1. Obs/Pred scatter plot in 2009. a) Case 2009, b) Case 09+mean, c) Case 09+median, d) Case 09+RM, e) Case 09-RM, and f) Case 09ScaleRM, observed GOM and PBM have been scaled.





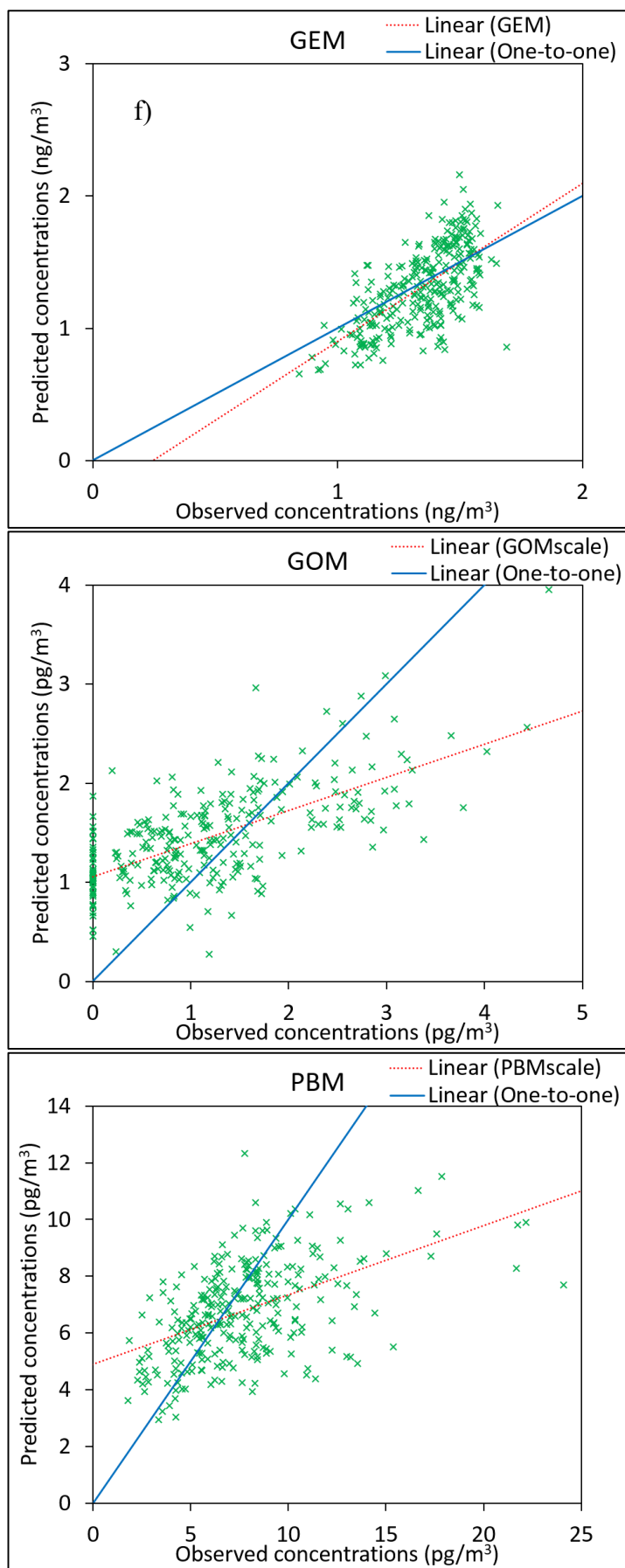
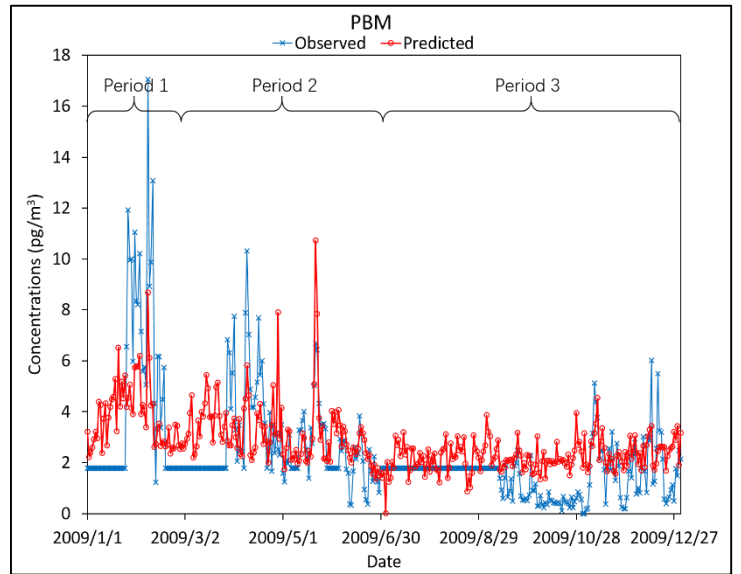
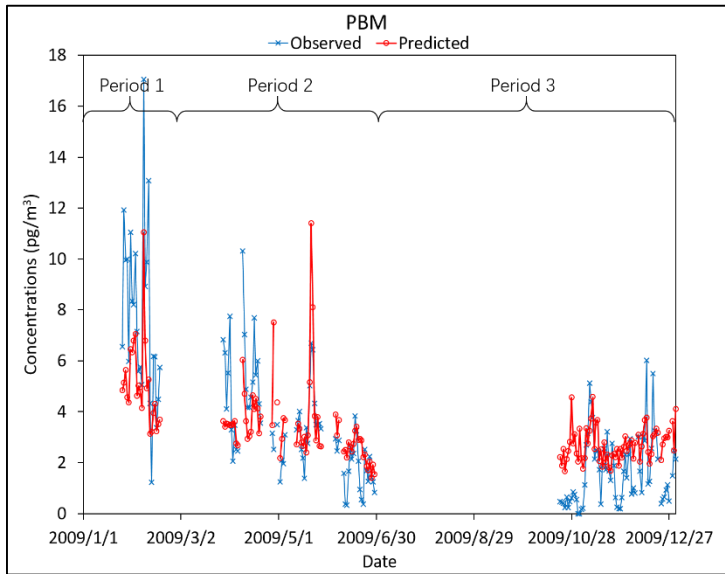
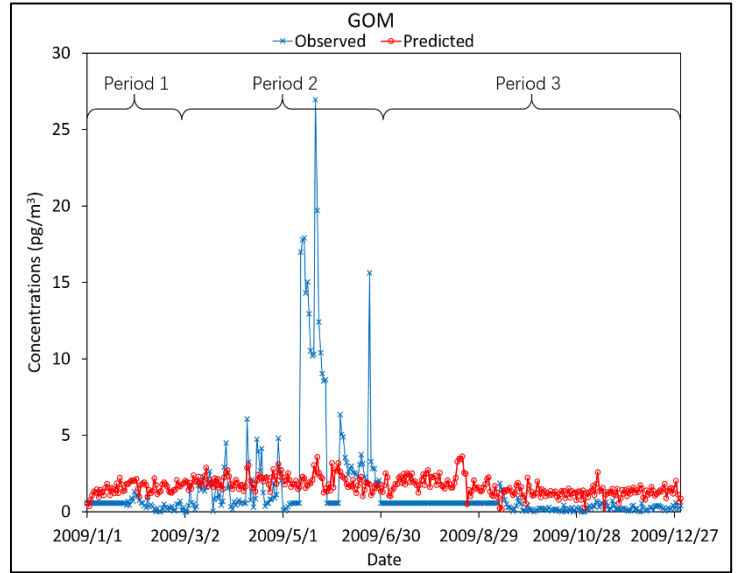
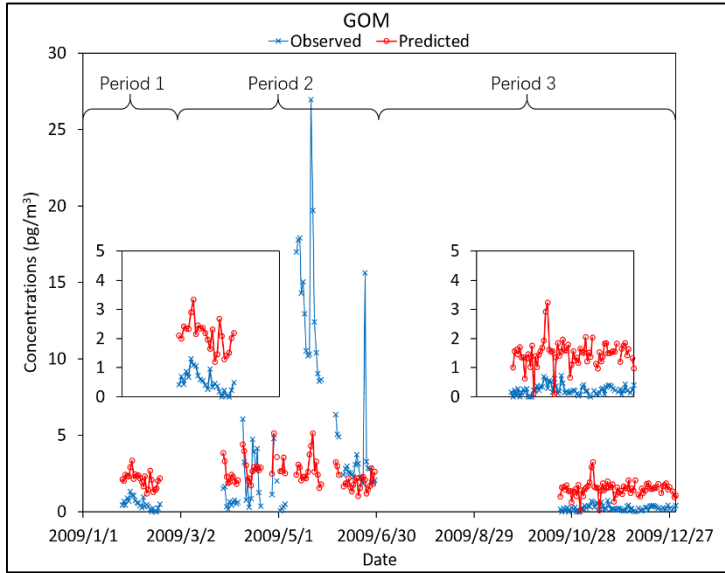
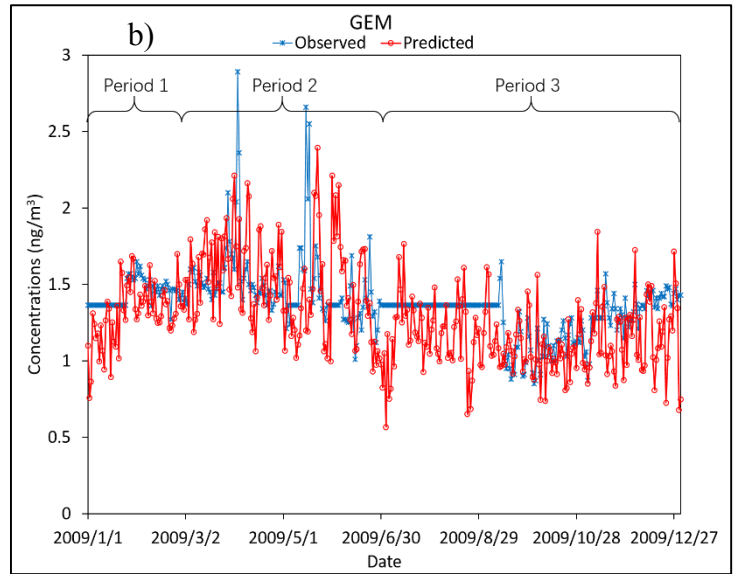
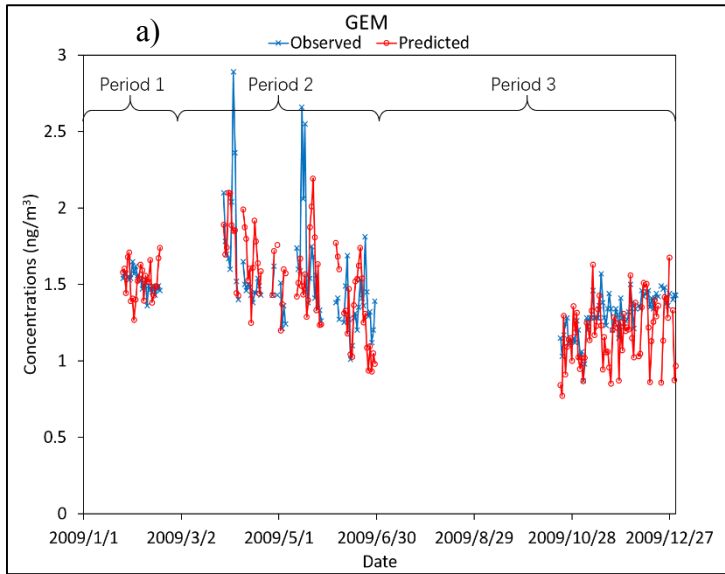
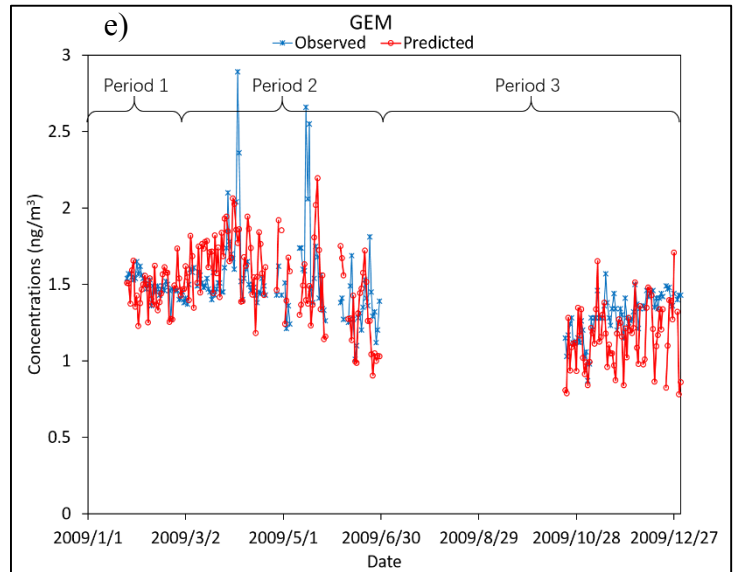
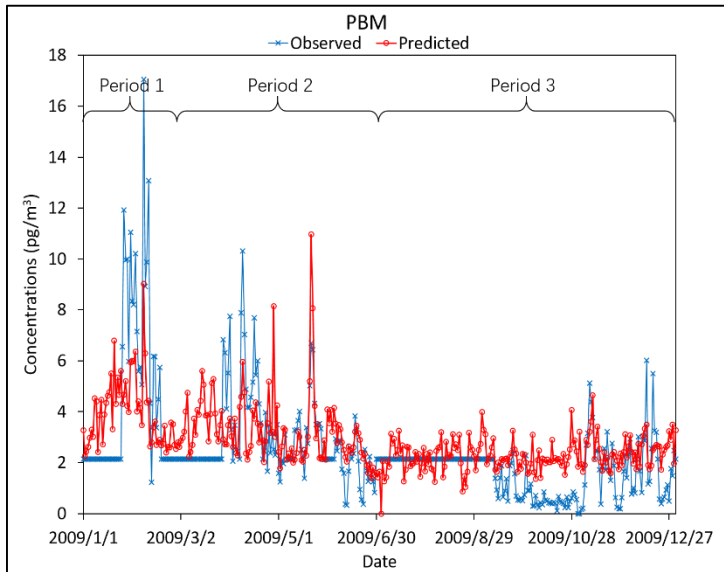
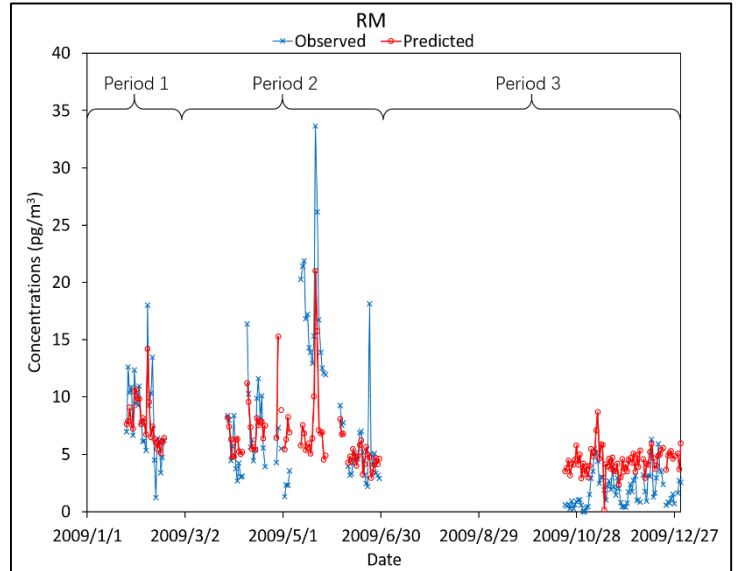
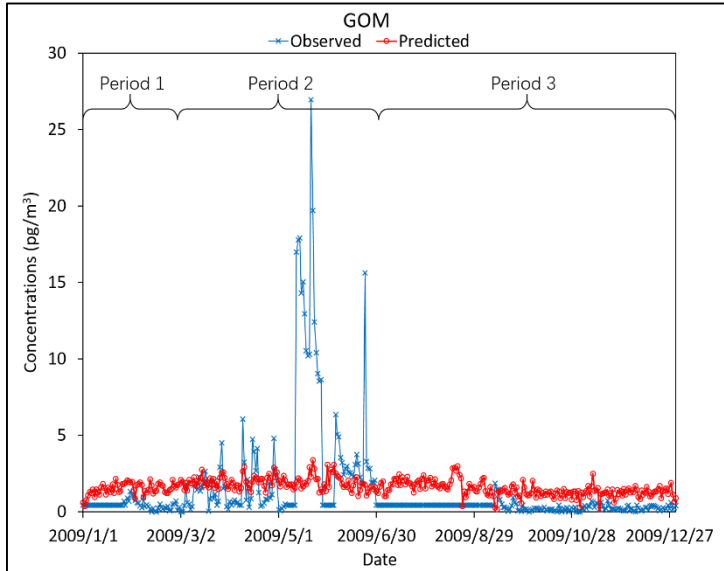
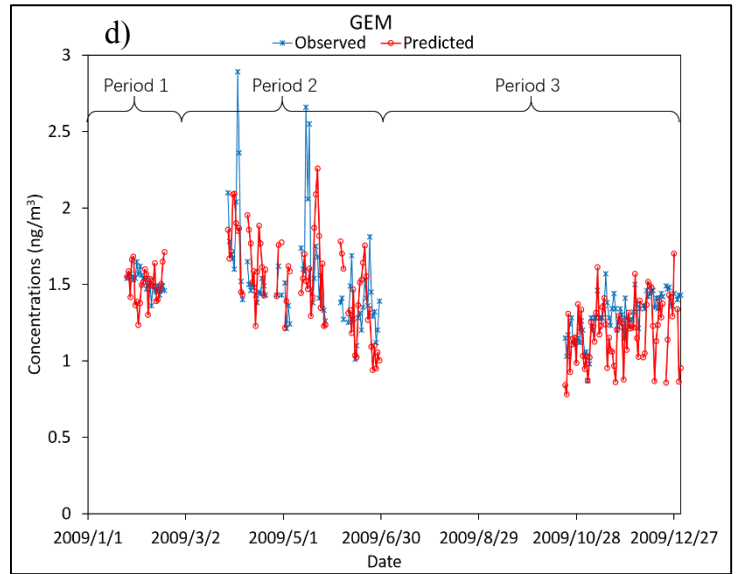
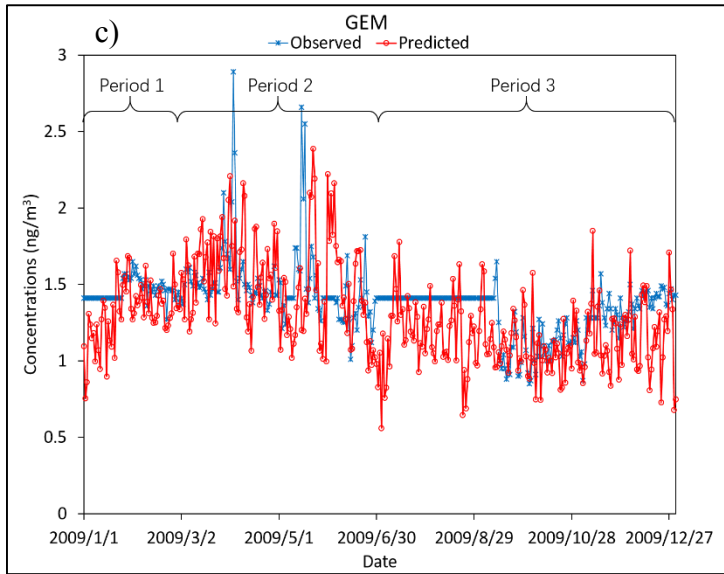


Figure S2. Obs/Pred scatter plot in 2010. a) Case 2010, b) Case 10+mean, c) Case 10+median, d) Case 10+RM, e) Case 10-RM, and f) Case 10ScaleRM, observed GOM and PBM have been scaled.





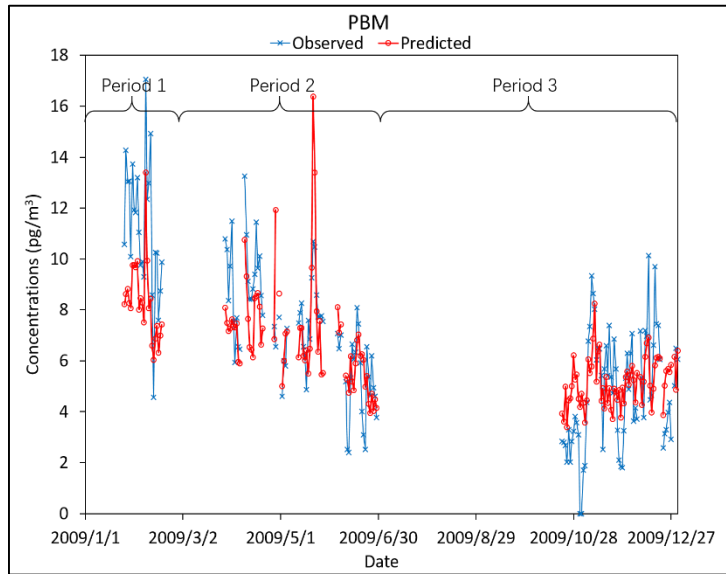
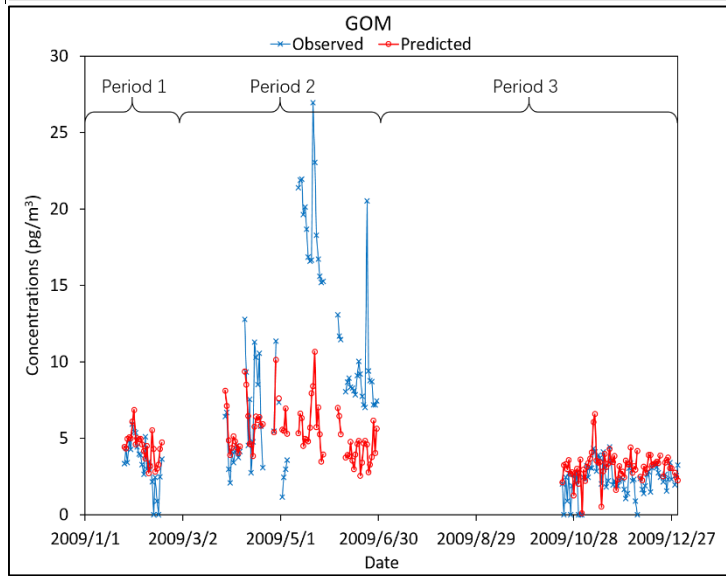
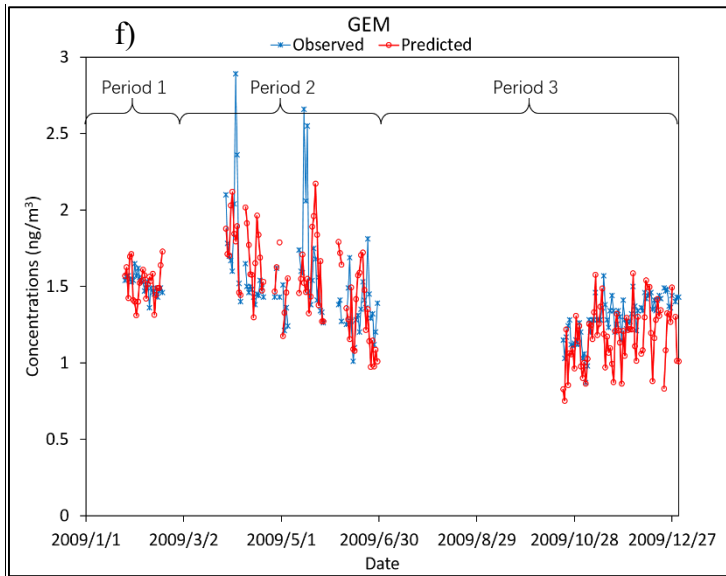
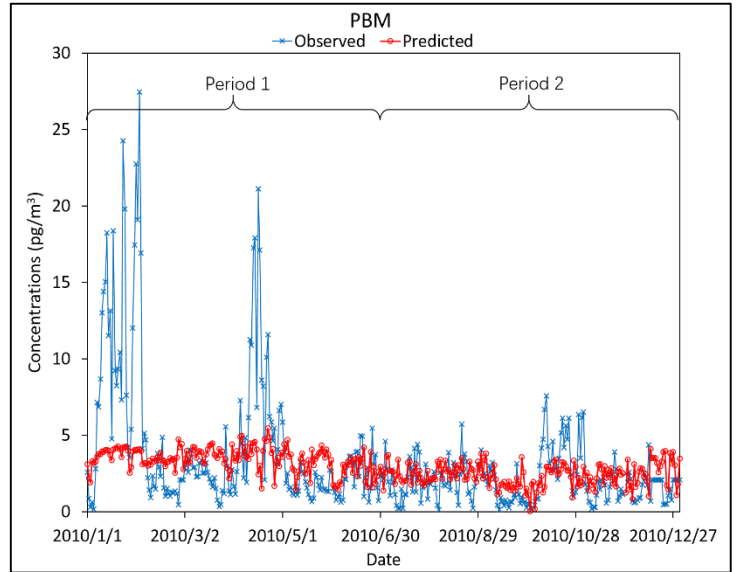
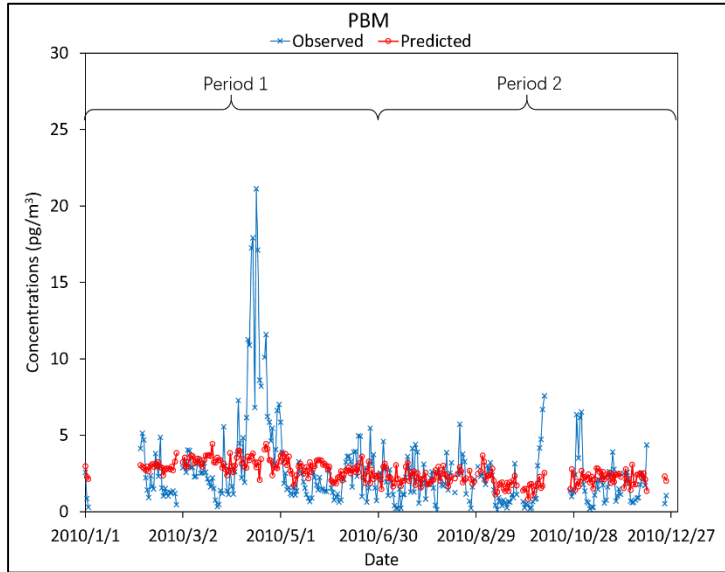
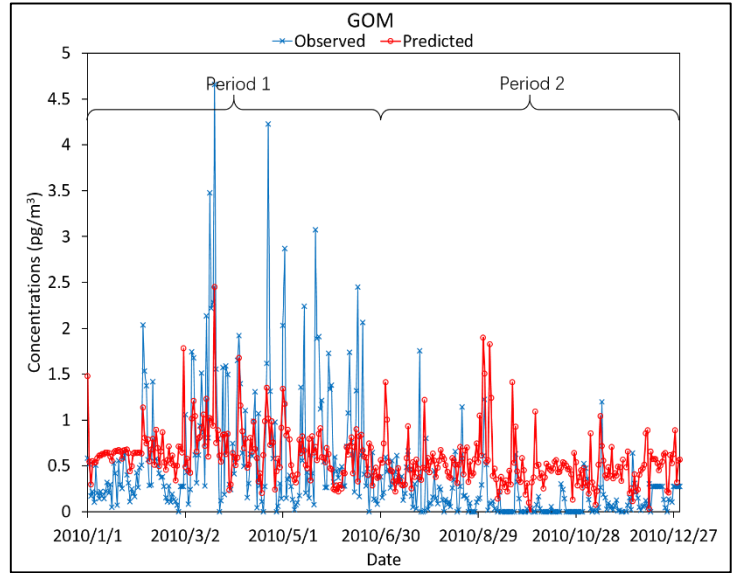
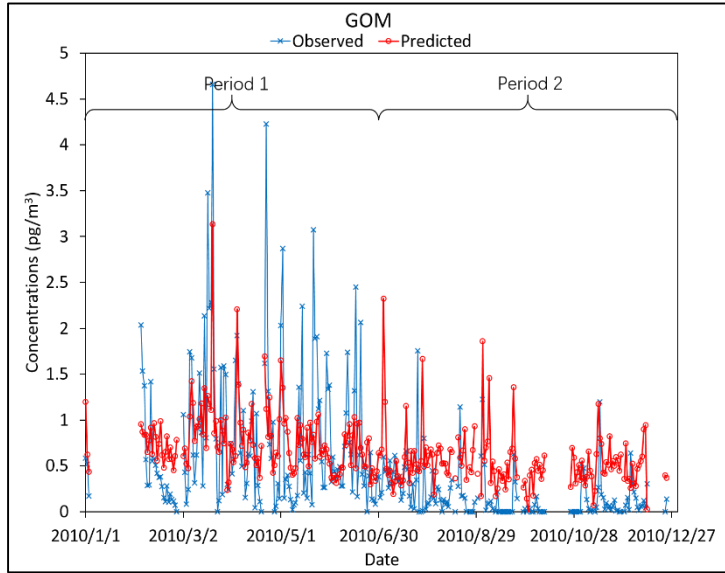
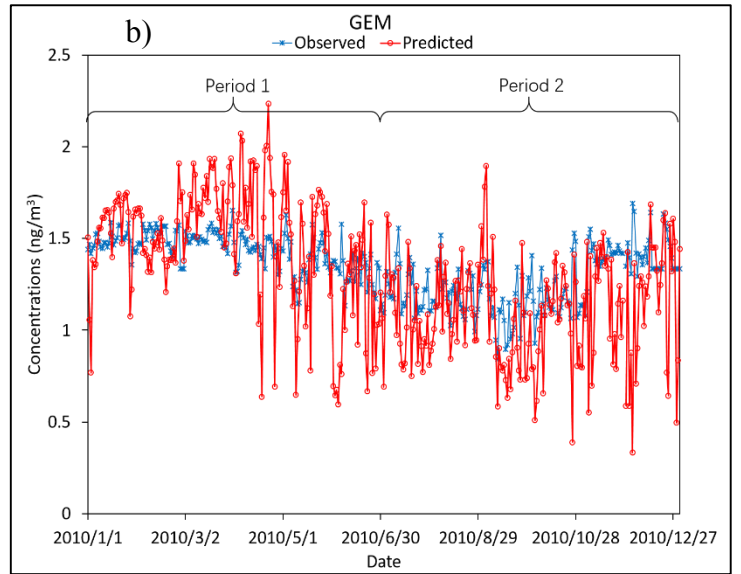
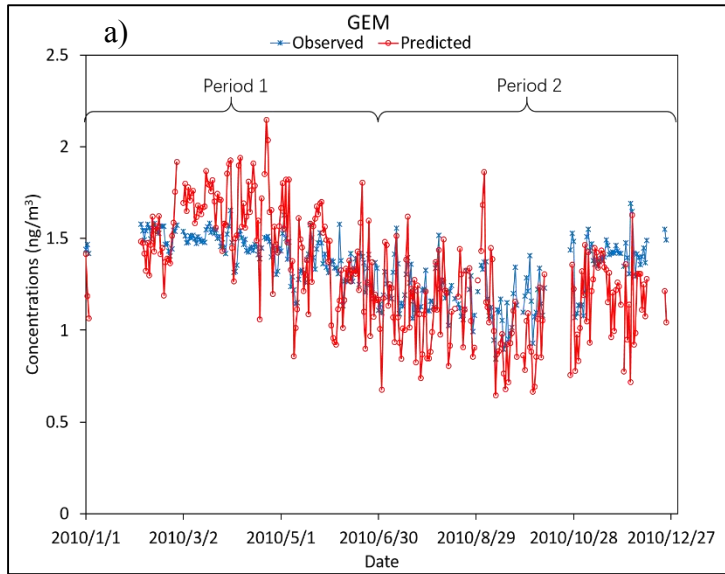
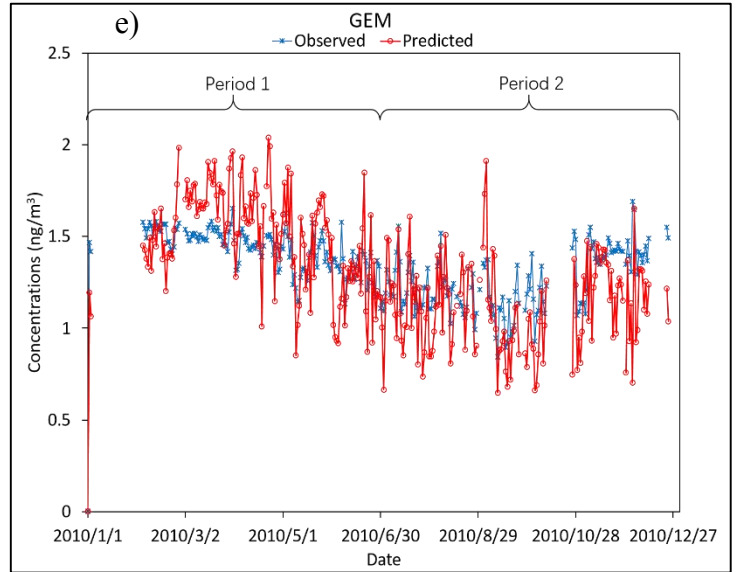
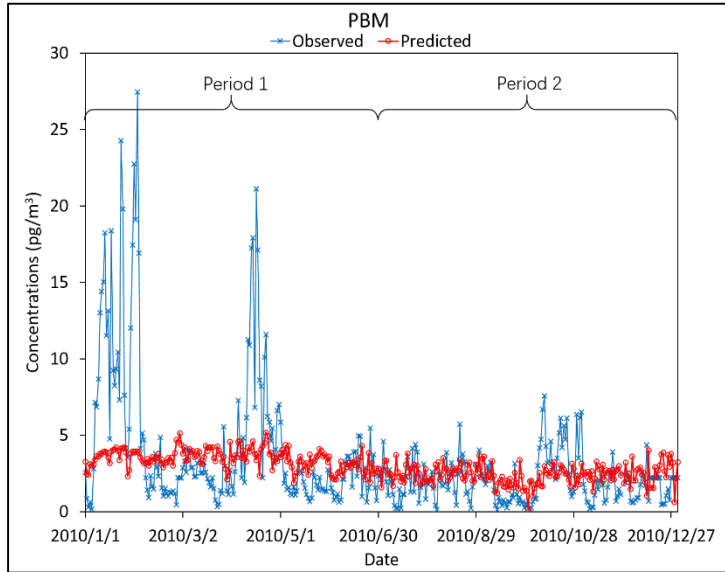
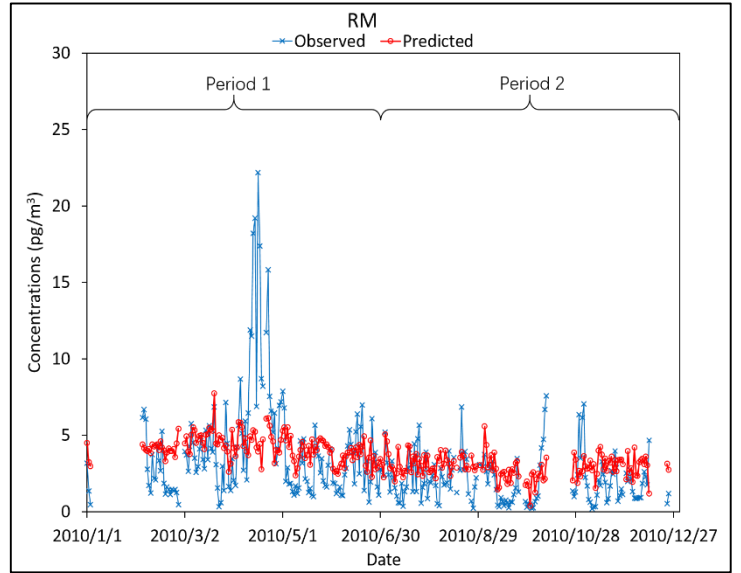
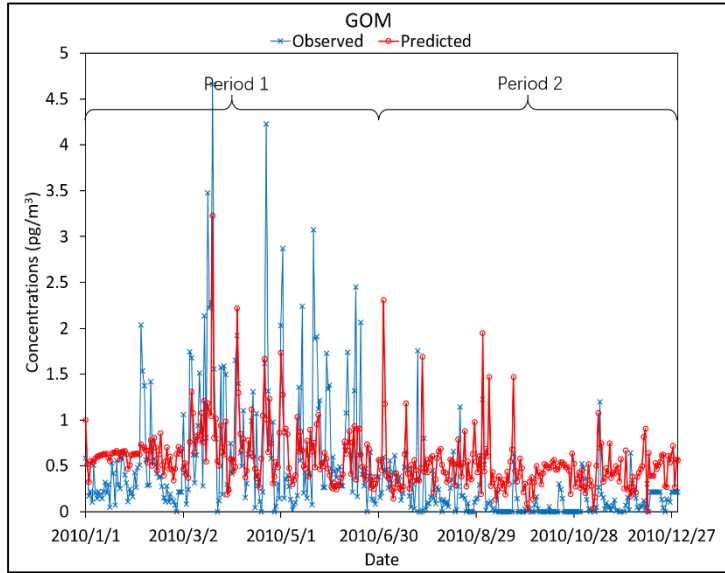
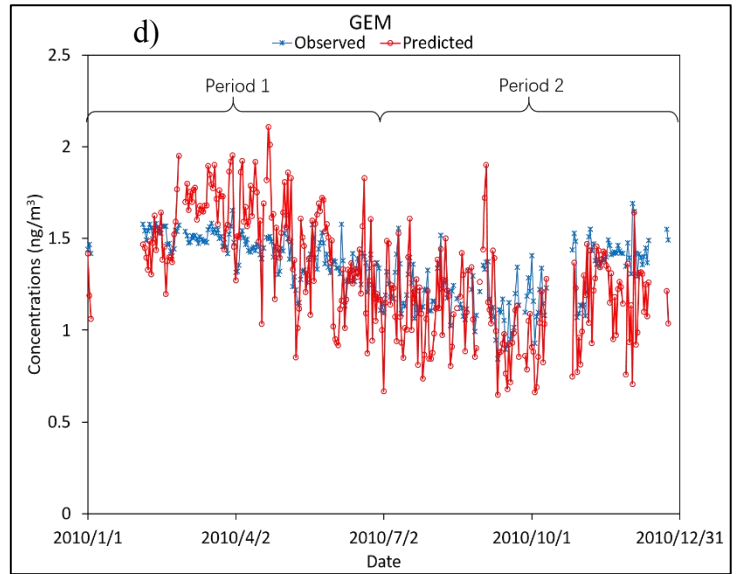
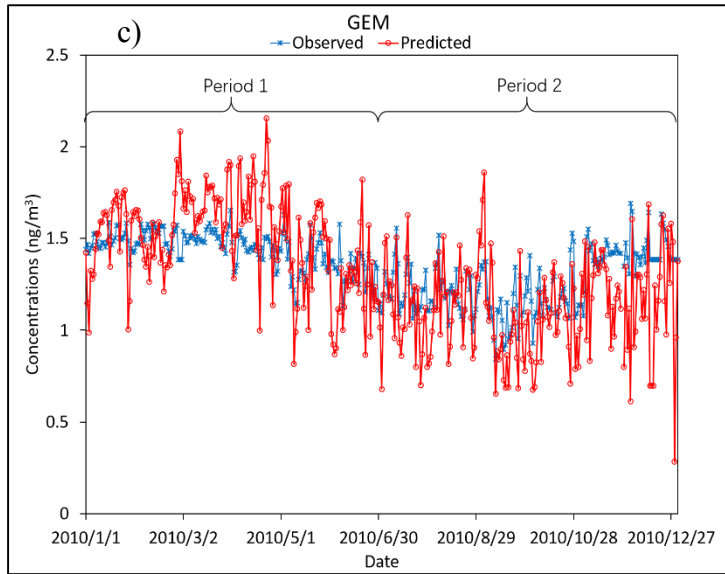


Figure S3. Obs/Pred time series in 2009. a) Case 2009, b) Case 09+mean, c) Case 09+median, d) Case 09+RM, e) Case 09-RM, and f) Case 09ScaleRM, observed GOM and PBM have been scaled.





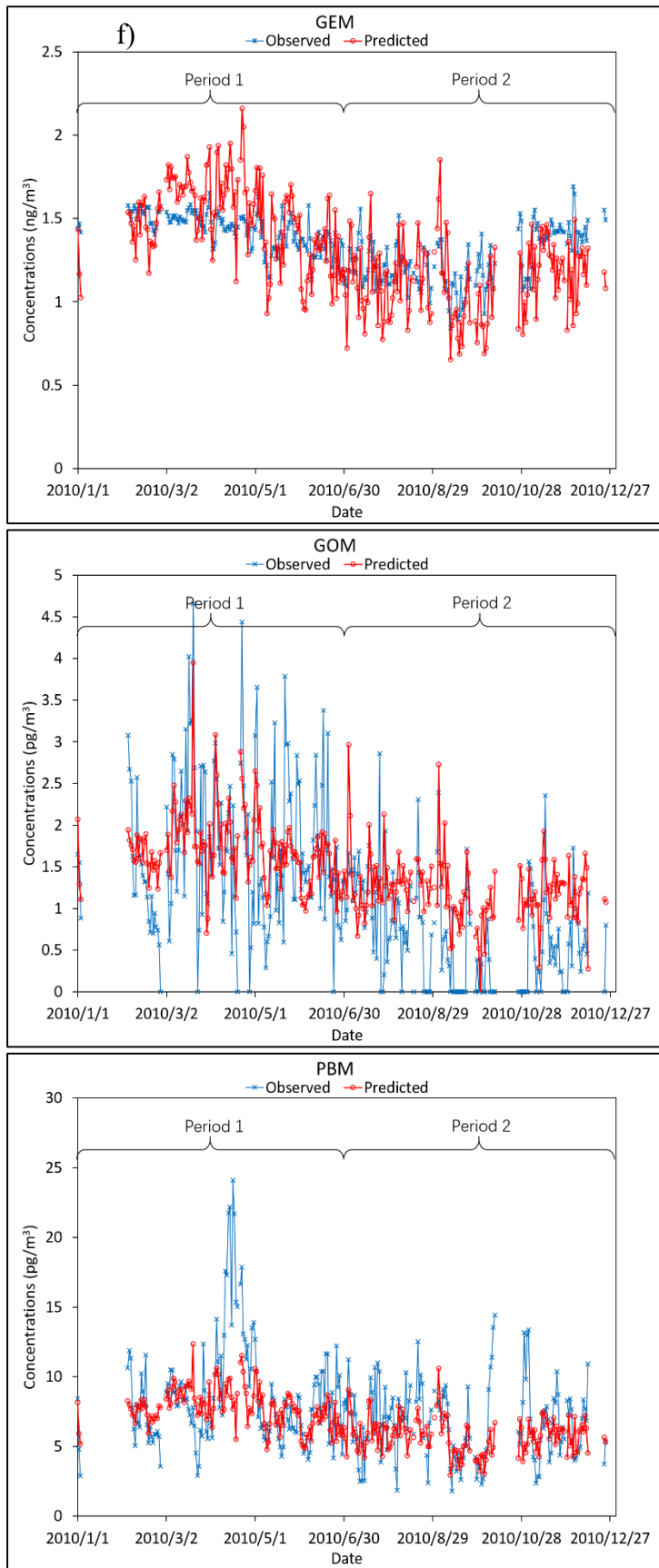


Figure S4. Obs/Pred time series in 2010. a) Case 2010, b) Case 10+mean, c) Case 10+median, d) Case 10+RM, e) Case 10-RM, and f) Case 10ScaleRM, observed GOM and PBM have been scaled.