RESULTS AND DISCUSSION

Network Performance

A maximum of 52 samples (some years 53) can be collected at each MDN site during the course of a complete year of operation. An annual summary of weekly sampling periods indicating the number of samples collected at each site and a distinction between sampling periods without precipitation, trace precipitation samples, and a valid versus invalid sample designation at each MDN site in Pennsylvania from 1997 through 2006 is shown in Table 2. In 2005 and 2006 the maximum number of possible weekly samples was 423 and 416, respectively. Forty-six of the sampling periods in 2005 and 35 in 2006 contained no precipitation, while 22 sampling periods in 2005 and 38 in 2006 were identified as invalid samples because of low sample volume relative to precipitation volume measurements or due to contamination, primarily to sample exposure to atmospheric inputs during rain-free periods. The vast majority of low volume samples occurred during the winter months when precipitation occurs as snow, which is much more difficult to capture than liquid precipitation. The latter category of invalid samples represents samples that were exposed to dry deposition and thus potentially not representative solely of wet deposition inputs. This group of samples would also include those contaminated with bird droppings, leaves, insects, etc. The number of valid sampling periods, which includes sampling periods without precipitation, in 2005 and 2006 were 401 and 378, respectively. These samples provided mercury concentration estimates for 98.7% and 98.0% of the total precipitation collected at the eight MDN sites in 2005 and 2006, respectively (Table 2). Since network operations began in 1997, approximately 91.2% of all sampling periods provided valid samples for mercury analysis and that these samples on averages captured more than 98% of the total precipitation across the network. These results indicate that the network is well run and that the vast majority of samples are collected correctly with minimal loss of results from missed precipitation or possible contamination of samples.

Results of all weekly analyses (valid and invalid samples) from all MDN sites in Pennsylvania for calendar years 2005 and 2006 are summarized in Appendix I, along with the first four-and-a half months of 2007. Although the results of some 2007 samples are included in Appendix I, these results are not included in the analysis section of this report. The 2005 and 2006 mean annual volume-weighted total mercury concentrations and annual wet deposition estimates for all MDN sites located in the United States and southern Canada are presented in the text of the report. These summary maps are based on only valid samples and include only those MDN sites that met MDN data completeness criteria. Weekly concentrations and wet deposition estimates for all MDN sites are available over the internet at http://nadp.sws.uiuc.edu/nadpdata/mdn.

The following discussion of total mercury concentrations and wet deposition estimates in Pennsylvania will, for the most part, be limited to data collected in 2005 and 2006, a period when eight sites were in operation across Pennsylvania for the entire year thus providing the greatest opportunity to assess spatial patterns across the Commonwealth. Seasonal variations at these sites will also be discussed. Discussion of long-term temporal patterns will be limited to the Hills Creek (PA90) and Allegheny-Portage NHS (PA13) sites both of which have been in

Table 2. Annual summary of weekly sampling periods indicating the number and distribution of sample types and quality at each Mercury Deposition Network site in Pennsylvania from 1997 through 2006.

			# Samples	# Trace Samples					Total Amount of Precipitation	
Site	Year	# Samples Collected	Without Precipitation	Analyzed	Not Analyzed	# Invalid Samples	# Samples Missing	# Valid Samples	Collected	With Analysis
									---------- Inches	-----------
PA00	2000*	5	2	0	0	2	0	3	3.62	3.62
	2001	51	7	0	1	7	0	44	23.56	23.45
	2002	52	3	0	1	7	0	45	40.13	39.38
	2003	52	4	0	1	5	2	47	46.46	45.94
	2004	52	6	0	0	7	0	45	50.30	49.40
	2005	53	6	0	1	4	0	49	41.40	41.35
	2006	52	8	0	0	2	0	50	42.58	42.05
PA13	1997	51	4	0	0	1	0	50	46.56	46.56
	1998	52	1	0	0	0	0	52	37.79	37.79
	1999	52	5	0	0	2	0	50	38.23	38.06
	2000	53	3	0	0	9	0	44	36.40	34.37
	2001	52	3	0	0	8	0	44	31.71	30.36
	2002	52	1	0	0	4	0	48	40.83	40.79
	2003	52	0	0	2	5	0	47	53.78	52.58
	2004	53	2	0	1	4	0	49	55.00	54.50
	2005	53	4	0	0	1	0	52	39.72	39.72
	2006	52	3	0	0	3	0	49	44.34	44.33
PA30	2000*	28	2	0	0	2	0	26	22.71	22.04
	2001	52	5	0	0	10	0	42	32.93	30.55
	2002	52	7	0	0	2	0	50	38.96	38.96
	2003	52	3	0	1	1	0	51	40.32	39.87
	2004	52	3	0	0	5	0	47	48.39	47.47
	2005	52	6	0	0	2	0	50	39.46	37.73
	2006	52	2	0	0	4	0	48	43.47	42.42
PA37	1999*	31	5	0	0	0	0	31	19.48	19.48
	2000	53	4	0	0	4	0	49	33.37	32.97
	2001	51	0	0	0	3	0	48	37.50	37.33
	2002	52	3	0	0	3	0	49	43.38	43.27
	2003	53	2	0	2	2	0	51	49.19	49.16
	2004	52	4	0	0	2	0	50	52.66	52.63
	2005	53	10	0	0	5	0	48	39.83	39.30
	2006	52	6	0	0	3	0	49	40.00	39.34

Table 2 (continued).

Site	Year	# Samples Collected	# Samples Without Precipitation	# Trace Samples		# Invalid Samples	# Samples Missing	# Valid Samples	Total Amount of Precipitation	
				Analyzed	Not Analyzed				Collected	With Analysis
									Inches	
PA47	2002*	5	0	0	0	1	0	4	4.95	2.65
	2003	53	3	0	0	3	0	50	53.94	53.91
	2004	52	8	0	0	5	0	47	46.92	45.79
	2005	53	4	0	2	3	0	50	44.39	44.38
	2006	52	5	0	2	5	0	47	48.71	48.62
PA60	1999*	5	1	0	0	0	0	5	4.36	4.36
	2000	53	8	0	0	1	0	52	47.10	47.10
	2001	51	10	0	0	6	0	45	29.56	28.75
	2002	52	7	0	0	2	2	50	47.15	46.74
	2003	52	6	0	2	2	0	50	54.69	54.68
	2004	50	7	0	1	4	0	46	54.18	53.70
	2005	53	9	0	0	4	0	49	45.12	42.89
	2006	52	4	0	0	12	0	40	52.86	52.84
PA72	2000*	16	1	0	0	3	0	13	11.15	11.13
	2001	52	4	0	0	2	0	50	35.20	35.17
	2002	52	3	0	0	4	0	48	46.72	45.10
	2003	52	4	0	0	5	0	47	60.30	60.11
	2004	52	5	0	0	1	0	51	51.73	51.31
	2005	53	5	0	0	1	0	52	59.98	59.98
	2006	52	4	0	0	4	0	48	53.09	48.33
PA90	1997	51	2	0	0	3	0	48	29.91	29.08
	1998	52	1	0	0	1	0	51	32.91	32.91
	1999	52	6	0	0	4	0	48	31.52	31.08
	2000	53	3	0	0	14	0	39	30.35	26.42
	2001	52	1	0	0	17	0	35	29.47	26.96
	2002	52	4	0	0	1	0	51	33.24	33.19
	2003	52	2	0	0	4	0	48	43.28	43.22
	2004	52	2	0	2	0	0	52	48.34	48.33
	2005	53	2	0	0	2	0	51	41.69	41.62
	2006	52	3	0	0	5	0	47	40.16	40.11
Total	1997	102	6	0	0	4	0	98	76.48	75.65
	1998	104	2	0	0	1	0	103	70.71	70.71
	1999	140	17	0	0	6	0	134	93.58	92.97
	2000	261	23	0	0	35	0	226	184.70	177.66
	2001	361	30	0	1	53	0	308	219.93	212.56
	2002	369	28	0	1	24	2	345	295.35	290.07
	2003	418	24	0	8	27	2	391	401.95	399.45
	2004	415	37	0	4	28	0	387	407.51	403.13
	2005	423	46	0	3	22	0	401	351.59	346.97
	2006	416	35	0	2	38	0	378	365.21	358.04

* Sampling conducted at this site for less than one year.

operation since 1997 and the Holbrook (PA37) and Valley Forge (PA60) sites that have been in operation since 1999. A multi-quadric equation spatial interpolation algorithm (Hardy, 1976) of 2005 and 2006 annual and seasonal total mercury concentrations and wet depositions are also presented. Where available, these interpolations include MDN sites located around the periphery to Pennsylvania. The resulting concentration and wet deposition spatial interpolations for 2005 and 2006 are discussed relative to 2003 and 2004 concentrations and wet deposition maps as well as mercury emissions data from major point sources in Pennsylvania as provided by the Pennsylvania Department of Environmental Protection, Bureau of Air quality Control.

Total Mercury Concentrations

Weekly total mercury concentrations and wet depositions are highly variable (Table 3). Since mercury monitoring began in Pennsylvania in 1997, weekly concentrations have ranged from 0.08 ng/L in Greene County (PA37) to 671.49 ng/L in Tioga County (PA90). In 2005, weekly mercury concentrations ranged from 0.60 ng/L in Tioga County (PA90) to 77.60 ng/L at the Milford site in Pike County (PA72) and in 2006 from 0.16 ng/L in Greene County (PA37) to 562.37 ng/L at the Valley Forge site (PA60) in Montgomery County (Table 3). Minimum weekly total mercury concentrations across Pennsylvania are generally in the 1 ng/L to 3 ng/L range at all sites for most years of observations. Maximum weekly concentrations are generally in the 20 ng/L to 50 ng/L range with some samples exceeding 100 ng/L (Table 3).

The range between maximum and minimum weekly wet deposition estimates (the product of weekly mercury concentration multiplied by precipitation volume) is even greater (Table 3). The lowest weekly wet deposition estimate over the past 10 years was 0.22 ng/m^2 that occurred at PA37 in Greene County; the highest weekly wet deposition (3,183.38 ng/m^2) was recorded at Allegheny-Portage NHS (PA13) in Cambria County. In 2005 and 2006 the lowest weekly wet deposition estimates were 3.19 ng/m^2 (PA37) and 1.54 ng/m^2 (PA37), respectively; the highest weekly wet depositions were recorded at PA60 (1001.15 ng/m^2) and PA90 (2002.64 ng/m^2), respectively (Table 3).

Mean annual and seasonal volume-weighted total mercury concentrations based on weekly precipitation samples collected at MDN sites in Pennsylvania from 1997 through 2006 are shown in Table 4. Annual means are presented for each calendar year (January through December) as well as for the December through November climatic year. The calendar year volume-weighted mean annual mercury concentrations and wet depositions for all MDN sites in the United States and southern Canada that met the MDN data completeness criteria for 2005 are shown in Figure 5. The 2005 calendar year mean concentrations in Pennsylvania varied from 5.36 ng/L at Milford (PA72) in Pike County to 8.96 ng/L at the Erie County (PA30) site (Table 4). The mean annual concentration at the Pike County site was consistent with a cluster of MDN sites in New England and southeastern Canada where 2005 mean annual mercury concentrations ranged from 3.8 ng/L to 5.6 ng/L (Figure 5). In contrast, the volume-weighted mean annual concentration at PA30 was one of the highest annual means reported in the New England and Mid-Atlantic regions of the United States in 2005 (Figure 5). The highest mean annual concentration in the United States in 2005 was recorded at a site in New Mexico (NM10). This New Mexico site is a high elevation site in the arid southwest that receives considerably less precipitation when

Table 3. Maximum and minimum weekly total mercury concentrations (ng/L) and wet depositions (ng/m^2) at eight Mercury Deposition Network (MDN) sites in Pennsylvania from 1997 through 2006.

		Weekly Mercury Concentrations (ng/L)		Weekly Mercury Deposition (ng/m^2)	
Site	Year	Maximum	Minimum	Maximum	Minimum
PA00	2001	63.77	2.67	503.62	25.97
	2002	29.85	2.06	566.93	17.60
	2003	19.99	1.36	946.88	42.84
	2004	29.23	2.18	966.81	22.27
	2005	42.11	2.12	715.06	10.06
	2006	50.23	1.47	1018.37	18.27
PA13	1997	48.47	2.15	879.54	8.37
	1998	50.57	2.80	722.88	5.82
	1999	55.41	2.19	762.76	20.80
	2000	23.92	3.07	547.97	24.20
	2001	124.10	3.32	3183.38	17.38
	2002	108.93	1.69	727.85	9.54
	2003	35.20	2.43	851.76	12.38
	2004	45.60	1.65	955.81	13.62
	2005	28.88	1.55	635.81	7.10
	2006	32.75	1.93	1194.94	8.32
PA30	2000*	30.78	0.83	1251.90	2.07
	2001	158.97	1.38	759.84	7.54
	2002	29.18	1.39	1302.96	14.94
	2003	218.66	2.84	779.27	15.40
	2004	35.04	1.55	929.75	3.00
	2005	54.96	1.40	992.12	11.22
	2006	39.28	2.61	894.28	17.19
PA37	1999*	46.20	2.15	830.10	8.40
	2000	62.58	2.13	715.29	9.32
	2001	35.56	1.86	633.42	4.72
	2002	25.67	0.08	1040.37	0.22
	2003	36.45	1.61	1398.73	8.05
	2004	26.74	1.15	1031.98	6.03
	2005	35.82	1.32	761.82	3.19
	2006	39.34	0.16	1087.73	1.54
PA47	2002*	12.14	1.21	132.04	17.21
	2003	30.39	2.14	913.53	23.48
	2004	44.23	2.77	676.66	9.15
	2005	29.56	1.11	853.25	5.98
	2006	56.42	2.06	1165.71	22.40

Table 3 (continued).

		Weekly Mercury Concentrations (ng/L)		Weekly Mercury Deposition (ng/m^2)	
Site	Year	Maximum	Minimum	Maximum	Minimum
PA60	1999*	9.32	3.82	420.19	37.84
	2000	154.73	2.84	1234.26	16.42
	2001	44.24	2.08	875.20	10.59
	2002	54.32	1.97	919.70	20.65
	2003	71.87	1.48	922.52	22.72
	2004	34.56	1.96	1258.62	17.56
	2005	58.60	3.23	1001.15	5.32
	2006	562.37	2.01	1070.98	42.37
PA72	2000*	25.92	2.60	759.00	14.53
	2001	36.55	2.58	663.37	16.69
	2002	53.04	1.94	2393.82	4.28
	2003	27.93	1.10	1217.17	6.25
	2004	84.93	1.29	1520.26	4.22
	2005	77.60	0.86	586.70	7.16
	2006	36.64	1.57	1181.93	17.84
PA90	1997	68.60	1.65	629.53	14.69
	1998	40.36	0.78	654.59	1.88
	1999	671.49	1.70	1193.91	7.12
	2000	38.91	1.82	481.93	7.57
	2001	56.42	1.42	474.35	21.40
	2002	38.88	0.59	858.09	3.35
	2003	27.27	1.46	1131.66	11.16
	2004	22.37	1.75	723.03	14.17
	2005	39.04	0.60	866.57	14.58
	2006	25.74	2.06	2002.64	7.89

* Sampling conducted at this site for less than one year.

Table 4. Mean annual and seasonal volume-weighted total mercury concentrations (ng/L) in precipitation at eight Pennsylvania Mercury Deposition Network sites from 1997 through 2006. Seasonal means are based on weekly samples collected from December-February (winter) March-May (spring), June-August (summer), and September-November (fall). Annual values are presented for both climatic (December-November) and calendar years (January-December).

		Volume-Weighted Mean Concentration (ng/L)									
Site	Season	1997	1998	1999	2000	2001	2002	2003	2004	2005	2006
PA00	Winter	-----	-----	-----	-----	10.47	10.19	5.24	4.88	6.12	6.60
	Spring	-----	-----	-----	-----	10.67	11.77	10.36	7.80	7.29	8.44
	Summer	-----	-----	-----	-----	16.00	9.20	10.99	10.43	12.44	9.85
	Fall	-----	-----	-----	-----	6.76	4.91	8.03	6.95	4.49	5.56
	Ann[1]	-----	-----	-----	-----	10.74	8.31	8.76	7.68	7.67	7.55
	Ann[2]	-----	-----	-----	-----	11.59	7.73	8.78	7.77	7.72	7.52
PA13	Winter	10.55	7.62	6.19	7.26	7.25	6.95	5.40	6.25	4.51	4.58
	Spring	12.64	8.65	9.85	8.76	27.95	10.65	9.62	9.05	7.63	9.77
	Summer	11.41	14.09	14.47	12.03	12.31	12.70	10.54	10.64	13.22	10.66
	Fall	4.21	13.04	6.80	9.37	7.74	5.68	6.57	5.60	5.00	6.04
	Ann[1]	9.01	10.32	9.44	9.37	15.26	9.29	8.53	8.12	7.06	7.87
	Ann[2]	9.18	10.17	9.25	9.58	14.82	9.25	8.51	8.02	6.68	8.36
PA30	Winter	-----	-----	-----	-----	12.00	8.77	6.18	7.01	6.29	7.66
	Spring	-----	-----	-----	-----	8.63	10.98	10.89	11.07	8.55	10.21
	Summer	-----	-----	-----	12.82	11.66	13.30	14.81	11.24	14.06	8.56
	Fall	-----	-----	-----	11.95	8.61	5.87	7.69	4.87	7.20	7.81
	Ann[1]	-----	-----	-----	12.44	10.10	9.02	10.27	8.75	8.73	8.47
	Ann[2]	-----	-----	-----	12.55	9.12	9.22	10.30	8.62	8.96	8.13
PA37	Winter	-----	-----	-----	6.04	7.50	7.34	4.85	4.79	5.25	5.69
	Spring	-----	-----	46.20	13.39	9.07	9.27	12.20	9.37	7.44	9.93
	Summer	-----	-----	15.51	12.63	11.29	11.26	12.18	11.59	11.16	12.98
	Fall	-----	-----	7.94	10.91	9.89	5.29	7.58	6.39	6.76	6.83
	Ann[1]	-----	-----	11.18	10.99	9.88	8.68	10.02	8.27	7.42	8.98
	Ann[2]	-----	-----	10.48	11.28	9.95	8.30	10.09	8.17	7.80	8.69

[1] Annual Period (December-November).

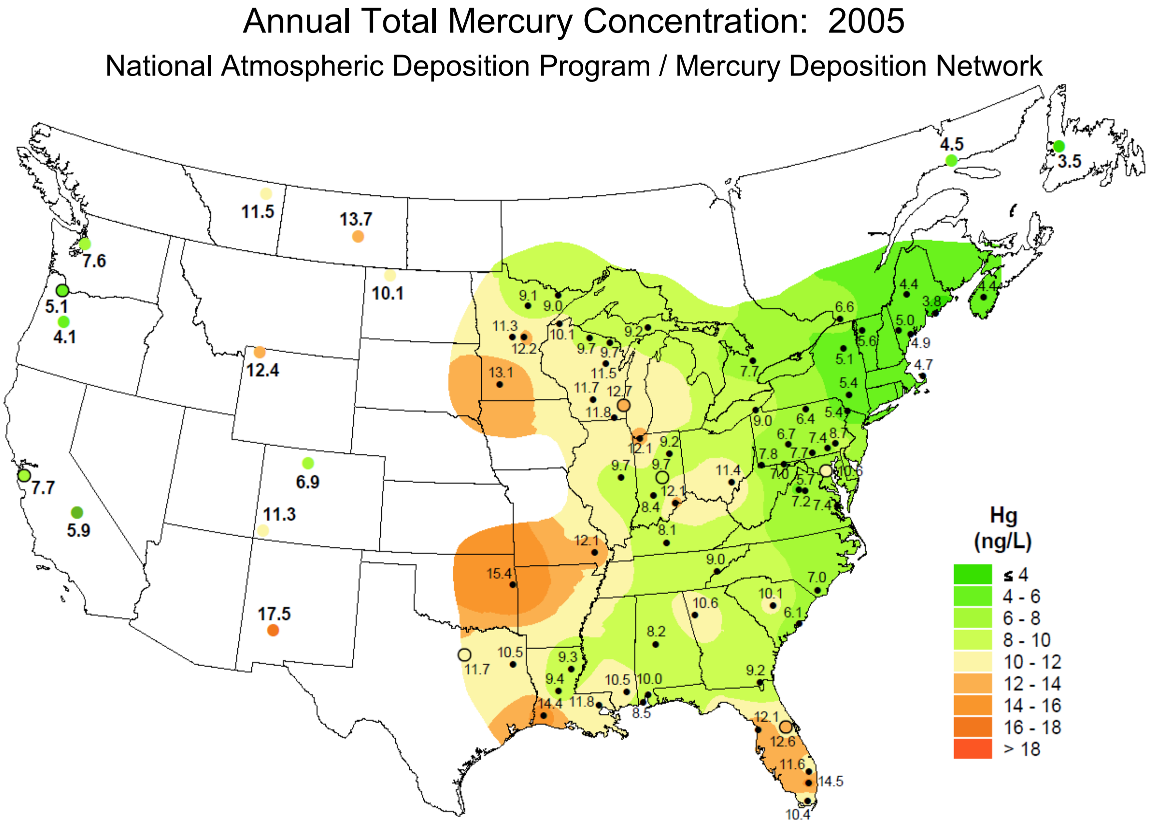
[2] Annual Period (January-December).

Table 4 (continued).

		Volume-Weighted Mean Concentration (ng/L)									
Site	Season	1997	1998	1999	2000	2001	2002	2003	2004	2005	2006
PA47	Winter	-----	-----	-----	-----	-----	-----	4.62	5.47	5.69	4.47
	Spring	-----	-----	-----	-----	-----	-----	8.13	11.44	8.08	8.68
	Summer	-----	-----	-----	-----	-----	-----	9.67	10.46	11.45	11.33
	Fall	-----	-----	-----	-----	-----	9.27	6.74	6.28	4.87	5.75
	Ann[1]	-----	-----	-----	-----	-----	9.27	7.59	8.68	7.85	7.72
	Ann[2]	-----	-----	-----	-----	-----	4.59	7.61	8.84	7.40	7.99
PA60	Winter	-----	-----	-----	7.86	11.36	7.06	4.86	6.25	5.50	8.20
	Spring	-----	-----	-----	11.92	10.99	13.64	11.56	16.09	6.34	16.41
	Summer	-----	-----	-----	9.53	12.13	11.15	9.97	9.77	13.79	8.99
	Fall	-----	-----	8.17	10.50	8.04	4.61	7.57	5.43	10.79	8.06
	Ann[1]	-----	-----	8.17	10.14	11.27	9.34	8.31	8.81	8.22	9.75
	Ann[2]	-----	-----	7.95	10.45	10.93	8.36	8.72	8.71	8.65	10.01
PA72	Winter	-----	-----	-----	-----	7.07	8.85	4.77	3.75	4.51	3.64
	Spring	-----	-----	-----	-----	8.17	15.53	7.37	9.85	6.17	10.13
	Summer	-----	-----	-----	-----	13.92	10.46	11.13	12.90	10.92	7.85
	Fall	-----	-----	-----	8.75	7.10	3.00	6.08	8.64	3.84	7.75
	Ann[1]	-----	-----	-----	8.75	9.11	8.80	7.89	9.29	5.71	6.91
	Ann[2]	-----	-----	-----	7.81	9.39	8.34	7.63	9.64	5.36	7.50
PA90	Winter	8.71	5.34	12.45	5.14	7.04	7.73	3.67	3.36	3.77	3.55
	Spring	10.64	9.14	7.64	7.01	6.80	7.69	8.52	8.66	4.93	8.36
	Summer	13.36	14.14	10.70	14.99	7.99	10.22	9.53	8.59	12.68	12.42
	Fall	5.12	7.94	5.39	10.17	5.54	4.18	5.60	5.17	4.62	4.83
	Ann[1]	9.57	9.07	8.62	9.41	6.79	7.52	7.33	6.88	6.71	7.77
	Ann[2]	9.50	8.97	8.56	9.69	6.58	7.36	7.13	7.06	6.37	8.40

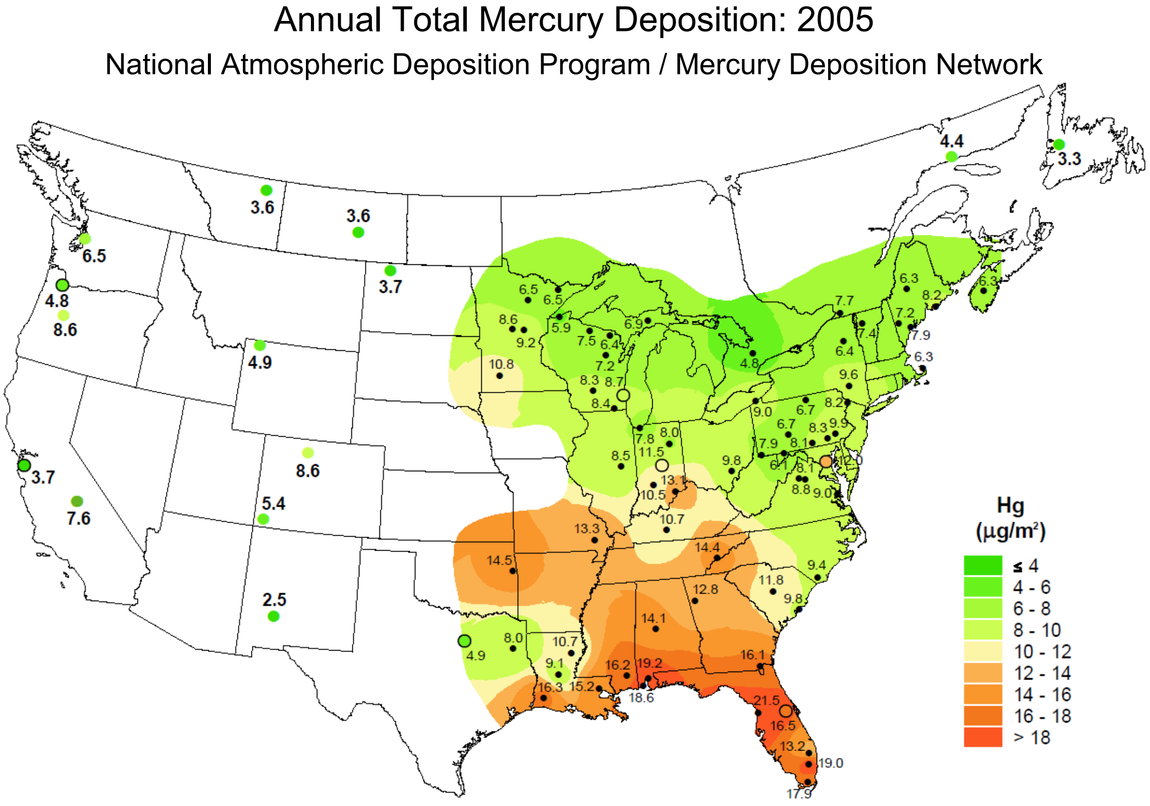
[1] Annual Period (December-November).

[2] Annual Period (January-December).



Annual Total Mercury Concentration: 2005

National Atmospheric Deposition Program / Mercury Deposition Network



Annual Total Mercury Deposition: 2005

National Atmospheric Deposition Program / Mercury Deposition Network

Figure 5. Volume-weighted mean annual total mercury concentrations (ng/L) and estimated wet depositions (μg/m^2) at all Mercury Deposition Network sites in the United States and southern Canada that met NADP/MDN's 75% data completeness criteria in 2005.

compared to sites located in Pennsylvania and the rest of the Eastern United States. The lowest mean annual concentration in the United States (3.8 ng/L) was recorded at Acadia National Park in Maine (ME98).

The overall spatial pattern of total mercury concentrations across the Commonwealth in 2005 (Figure 6) was one of relatively high concentrations in the northwest (PA30) and southeast (PA60) regions of the state, with much lower concentrations across the north central (PA90), central (PA13) and southwest (PA37) regions of Pennsylvania. Mercury concentrations in south eastern Pennsylvania were higher than in central Pennsylvania, but lower than those measured in the northwestern portion of the Commonwealth. Overall, total mercury concentrations across Pennsylvania in 2005 were generally lower than at sites located around the Great Lakes states, portions the Ohio River Valley and the Southeast (Figure 5). The highest concentrations in non-arid climates were found in Florida and at some sites around the Gulf and Upper Mississippi River states. New England had the overall lowest total mercury concentrations in the United States in 2005 (Figure 5).

Mean annual and seasonal volume-weighted total mercury concentrations in Pennsylvania for 2006 are shown in Table 4. Annual means are presented on a calendar year basis as well as a climatic year (December through November). Calendar year volume-weighted mean annual concentrations for all MDN sites in the United States and southern Canada that met MDN data completeness criteria for 2006 are shown in Figure 7. The 2006 calendar year mean concentrations in Pennsylvania varied from 7.50 ng/L at the Milford site (PA72) and 7.52 ng/L at the Arendtsville site (PA00) in Adams County to 10.01 ng/L at the Valley Forge site (PA60) in Montgomery County (Table 4). The 2006 mean annual concentrations across Pennsylvania were generally higher than those observed in 2005, especially throughout the central portion of the Commonwealth. At the Hills Creek site in Tioga County, the 2006 mean annual concentration was the highest reported since 2000. Higher mercury concentrations appear to be the case throughout most of the eastern half of the United States in 2006 as well (Figures 5 and 7). In contrast, the volume-weighted mean annual concentration at the Arendtsville site in Adams County (PA00) and the Erie site (PA30) were lower in 2006 than 2005 (Table 4). The overall spatial pattern of mean annual total mercury concentrations across the state in 2006 (Figure 8) is one of higher concentrations in southwestern (PA37) and southeastern Pennsylvania (PA60) and lower concentrations in the northeast (PA72).

Overall, total mercury concentrations across Pennsylvania in 2006 were generally lower than at sites located around The Great Lakes, portions of the Ohio River Valley and most of the states located around the Gulf of Mexico (Figure 7). The highest concentrations in non-arid climates were found in Florida, at selected sites in the Upper Mississippi Valley, and around the Great Lakes. New England had the overall lowest total mercury concentrations in the United States in 2006. The highest (20.7 ng/L) and lowest (3.6 ng/L) mean annual total mercury concentrations in the United States in 2006 were recorded at NM10 (New Mexico) and OR10 (Oregon), respectively.

It should be noted that the Valley Forge site (PA60) did not meet the MDN data completeness criteria in 2006 because of an unusual number (12) of invalid samples because of potential contamination. As a result, data from PA60 was not included in Figure 7, but was included in

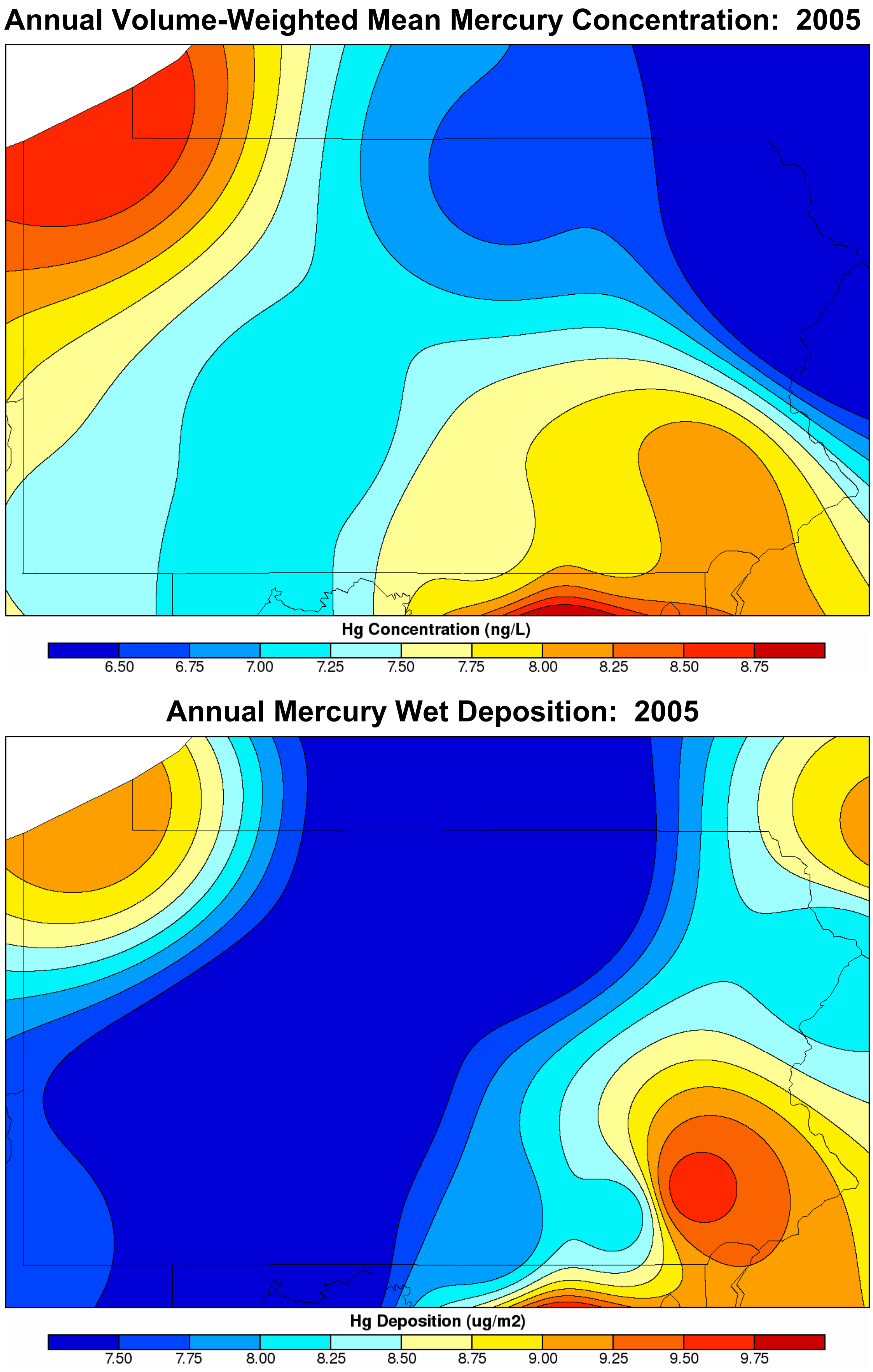


Figure 6. Spatial interpolation of 2005 volume-weighted mean annual total mercury concentrations (ng/L) and estimated wet depositions (µg/m^2) based on data from Mercury Deposition Network sites in Pennsylvania and peripheral states.

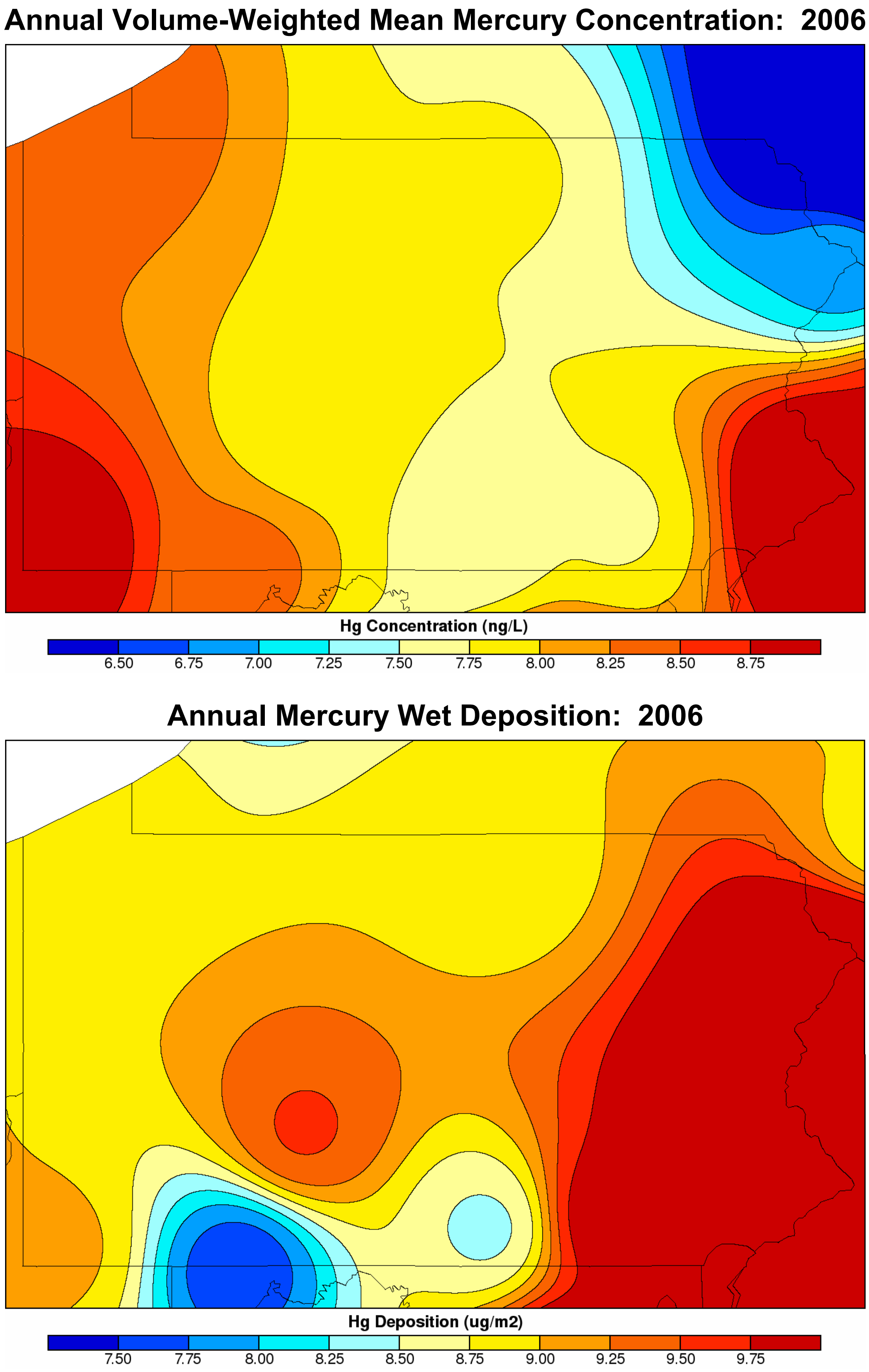
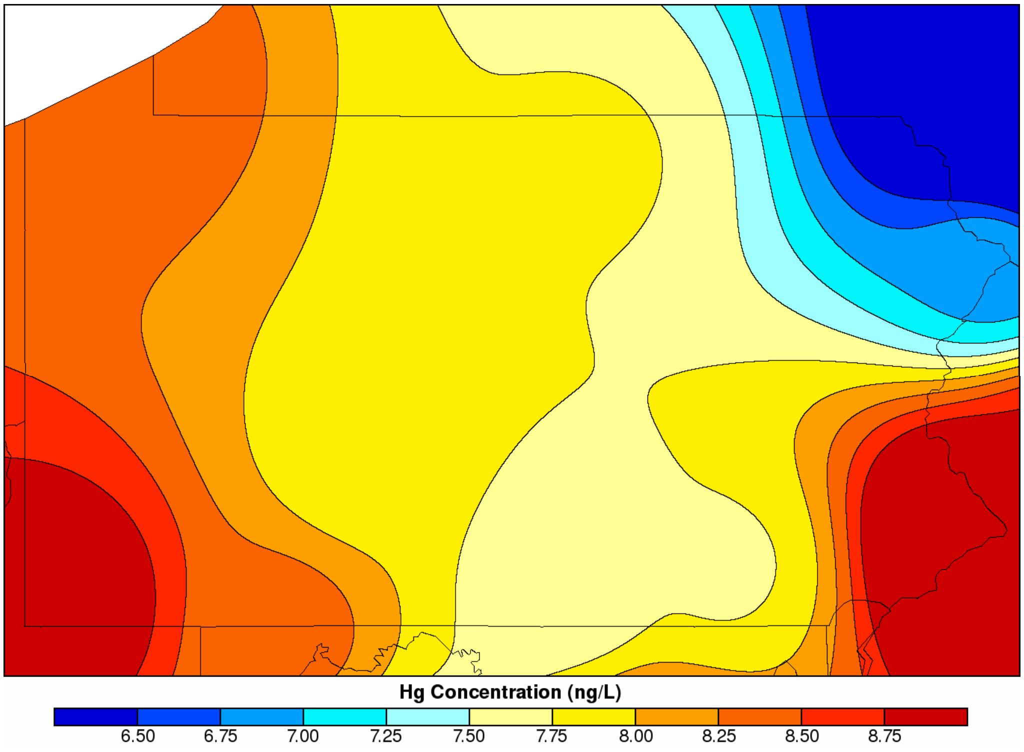


Figure 7. Volume-weighted mean annual total mercury concentrations (ng/L) and estimated wet depositions (µg/m2) at all Mercury Deposition Network sites in the United States and southern Canada that met NADP/MDN's 75% data completeness criteria in 2006.

Annual Volume-Weighted Mean Mercury Concentration: 2006



Annual Mercury Wet Deposition: 2006

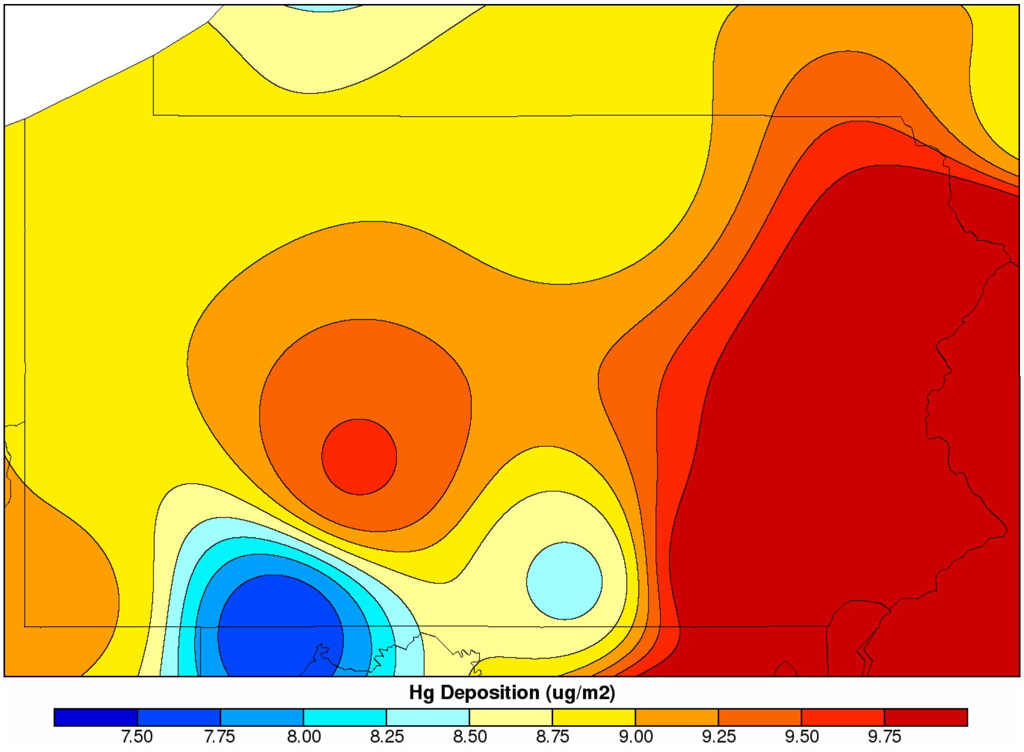


Figure 8. Spatial interpolation of 2006 volume-weighted mean annual total mercury concentrations (ng/L) and estimated wet depositions (µg/m²) based on data from Mercury Deposition Network sites in Pennsylvania and peripheral states.

the development of the spatial maps for Pennsylvania. If the invalid samples were included in the data set, the mean annual volume-weighted concentration at PA60 would be 8.84 ng/L, which is still the highest annual average in Pennsylvania in 2006, but significantly lower than the 10.01 ng/L average based solely on valid samples. Including valid PA60 data in the spatial analysis across Pennsylvania does not appreciably alter the distribution across the state given the fact that the mean concentration of all samples (valid and invalid) results in a mean concentration that is still higher than any other MDN site in Pennsylvania. However, if the PA60 mean based on only valid samples was included in the national map (Figure 7), the 10.1 ng/L concentration would have been the highest concentration in the northeast U.S. and the southeast as far south as Florida. Obviously, data completeness criteria on a national scale are very important in preserving site comparability and in describing spatial patterns across the country.

Estimated Wet Total Mercury Depositions

Individual weekly total mercury wet depositions in Pennsylvania are highly variable (Table 3). Differences in weekly depositions have varied by more than a factor of 1000 (PA37), although at most sites the differences range from a factor of 100 to 600. This variability is a product of both highly variable mercury concentrations (Table 3) and weekly precipitation volumes. In 2005, the highest weekly mercury deposition (1001.2 ng/m^2) was measured at the Valley Forge site (PA60) in Montgomery County; the lowest weekly deposition (3.19 ng/m^2) was recorded at the Holbrook site (PA37) in southwestern Pennsylvania. The minimum weekly wet deposition in 2006 (1.54 ng/m^2) was also recorded at the Holbrook site; the maximum weekly deposition (2002.6 ng/m^2) was recorded at the Hills Creek site in Tioga County (PA90). These values are consistent and comparable to maximum and minimum wet deposition levels recorded in previous years of network operation. The maximum weekly wet deposition recorded to date occurred at the Cresson Mountain site (PA13) in Cambria County in 2001 and measured 3183.4 ng/m^2. Maximum weekly deposition measurements at this site have been remarkably consistent for all years except 2001. At the rest of the sites, the maximum weekly deposition estimates vary from year to year by a factor of 2 to 4. With the exception of the Cresson Mountain site, the greatest variability in weekly mercury depositions occurred at the Millersville site (PA47) in Lancaster County and the least variability occurred at the Erie site (PA30) in northwestern Pennsylvania (Table 3).

Annual and seasonal wet deposition estimates for each of the Pennsylvania MDN sites for all years of operation are listed in Table 5. Over this ten-year period, annual total mercury wet depositions ranged from 4.93 μg/m^2 at the Tioga County site (PA90) in 2001 to 12.78 μg/m^2 at the Valley Forge site (PA60) in 2003. Annual deposition at the Valley Forge site was actually greater (13.44 μg/m^2) in 2006. However, as previously discussed, this site had a larger than usual number of invalid samples and did not satisfy data completeness criteria of the MDN and thus may not be comparable to previous years of operation. The Tioga County site has consistently received the smallest amount of wet mercury deposition of any region of Pennsylvania (Table 5). However, in 2006 the Arendtsville site (PA00) in Adams County recorded the lowest deposition in the state. In contrast, maximum annual wet depositions have varied from year to year between western (PA13 and PA30) and eastern (PA60 and PA72)

Table 5. Annual and seasonal total mercury wet depositions (µg/m2) at eight Pennsylvania Mercury Deposition Network sites from 1997 through 2006. Seasonal depositions are based on weekly samples collected from December- February (winter) March-May (spring), June-August (summer), and September-November (fall). Annual values are presented for both climatic (December-November) and calendar years (January-December).

		Wet Deposition (µg/m^2)									
Site	Season	1997	1998	1999	2000	2001	2002	2003	2004	2005	2006
PA00	Winter	-----	-----	-----	-----	1.808	1.139	1.341	1.195	1.352	1.684
	Spring	-----	-----	-----	-----	2.506	3.317	3.473	2.562	1.950	1.604
	Summer	-----	-----	-----	-----	1.842	1.881	3.432	3.918	3.293	3.306
	Fall	-----	-----	-----	-----	0.962	1.855	2.434	2.463	1.161	1.867
	Ann[1]	-----	-----	-----	-----	7.118	8.192	10.681	10.138	7.756	8.461
	Ann[2]	-----	-----	-----	-----	7.250	7.932	10.617	9.951	8.116	8.135
PA13	Winter	0.931	1.923	1.204	1.149	0.936	1.175	1.197	1.714	1.337	1.291
	Spring	4.511	3.072	2.443	2.865	6.429	3.785	3.007	3.572	1.541	1.831
	Summer	2.956	3.395	3.553	2.666	3.196	3.244	4.867	3.911	2.469	4.429
	Fall	1.786	1.755	1.845	1.958	1.256	1.527	2.245	2.190	1.282	1.969
	Ann[1]	10.183	10.145	9.044	8.639	11.818	9.732	11.315	11.387	6.629	9.520
	Ann[2]	10.863	9.767	8.985	8.854	11.933	9.596	11.628	11.233	6.744	9.414
PA30	Winter	-----	-----	-----	-----	1.600	2.588	0.985	1.066	1.813	1.283
	Spring	-----	-----	-----	-----	2.041	3.588	2.872	3.580	1.603	2.023
	Summer	-----	-----	-----	3.895	1.988	1.879	4.180	3.858	3.024	2.590
	Fall	-----	-----	-----	2.756	2.050	2.167	2.337	1.693	2.747	2.998
	Ann[1]	-----	-----	-----	6.650	7.678	10.222	10.374	10.197	9.188	8.894
	Ann[2]	-----	-----	-----	7.236	7.939	9.778	10.390	10.727	8.980	8.977
PA37	Winter	-----	-----	-----	1.271	1.005	1.129	0.779	0.979	1.548	0.931
	Spring	-----	-----	0.016	3.505	2.464	3.818	3.716	3.193	1.883	2.208
	Summer	-----	-----	2.817	3.354	4.179	3.258	6.283	3.792	2.628	3.340
	Fall	-----	-----	1.961	1.405	1.576	1.353	2.116	2.399	1.630	2.566
	Ann[1]	-----	-----	4.795	9.536	9.224	9.558	12.895	10.363	7.689	9.045
	Ann[2]	-----	-----	5.185	9.563	9.523	9.151	12.609	10.374	7.890	8.828

[1] Annual Period (December-November).

[2] Annual Period (January-December).

Table 5 (continued).

		Wet Deposition (μg/m^2)									
Site	Season	1997	1998	1999	2000	2001	2002	2003	2004	2005	2006
PA47	Winter	-----	-----	-----	-----	-----	-----	1.294	1.330	1.307	1.137
	Spring	-----	-----	-----	-----	-----	-----	2.154	3.411	1.866	1.724
	Summer	-----	-----	-----	-----	-----	-----	4.044	4.226	3.444	4.628
	Fall	-----	-----	-----	-----	-----	0.052	2.780	1.758	1.525	2.463
	Ann[1]	-----	-----	-----	-----	-----	0.052	10.273	10.725	8.142	9.951
	Ann[2]	-----	-----	-----	-----	-----	0.577	10.422	10.502	8.348	9.886
PA60	Winter	-----	-----	-----	1.691	2.951	1.147	1.786	1.446	2.055	2.269
	Spring	-----	-----	-----	4.023	2.898	3.704	3.402	4.772	1.767	3.193
	Summer	-----	-----	-----	3.263	2.352	2.562	4.453	4.467	2.456	4.274
	Fall	-----	-----	0.315	2.865	0.851	1.647	3.519	2.277	3.308	3.691
	Ann[1]	-----	-----	0.315	11.842	9.051	9.060	13.160	12.962	9.586	13.426
	Ann[2]	-----	-----	0.879	12.613	8.447	9.843	12.779	12.521	9.910	13.439
PA72	Winter	-----	-----	-----	-----	1.867	1.213	1.292	0.989	1.630	1.383
	Spring	-----	-----	-----	-----	2.181	5.184	2.214	2.658	1.826	2.384
	Summer	-----	-----	-----	-----	3.309	2.667	4.928	5.611	2.606	3.438
	Fall	-----	-----	-----	1.172	1.422	1.242	2.987	3.416	2.046	3.164
	Ann[1]	-----	-----	-----	1.172	8.778	10.306	11.420	12.674	8.108	10.370
	Ann[2]	-----	-----	-----	2.213	8.400	9.892	11.725	12.673	8.172	10.109
PA90	Winter	0.505	1.179	2.051	0.798	0.560	0.950	0.651	0.625	0.927	0.810
	Spring	1.985	2.754	1.401	2.023	1.469	1.836	1.808	2.354	0.965	1.451
	Summer	3.182	2.668	2.120	3.024	1.782	2.599	4.196	3.642	3.530	5.241
	Fall	1.230	1.085	1.303	1.394	1.153	0.939	1.378	1.666	1.300	1.351
	Ann[1]	6.902	7.685	6.875	7.238	4.964	6.323	8.033	8.287	6.721	8.853
	Ann[2]	7.217	7.500	6.853	7.599	4.929	6.214	7.838	8.343	6.743	8.570

[1] Annual Period (December-November).

[2] Annual Period (January-December).

Pennsylvania sites, although for the majority of years the highest deposition has been in eastern Pennsylvania (Table 5).

A comparison of mean annual volume-weighted total mercury concentrations (Table 4) with wet deposition estimates (Table 5) illustrates nicely that precipitation amounts and likely other meteorological determinants exert substantial influence on year-to-year wet deposition patterns across the state. A comparison of mean annual concentrations and wet deposition measurements in 2005 (Figure 6) and 2006 (Figure 8) when eight sites were in operation across the state helps illustrate this complex inter-action between mercury concentrations and precipitation volumes.

Annual total mercury wet depositions across Pennsylvania in 2005 ranged from 6.74 $\mu g/m^2$ at PA90 (Hills Creek, Tioga County) and PA13 (Allegheny-Portage NHS, Cambria County) to 9.91 $\mu g/m^2$ at the Valley Forge site (PA60) in Montgomery County (Table 5). Mercury deposition at the Hills Creek and Allegheny-Portage sites were within the 10 lowest amounts measured in the entire eastern United States in 2005 (Figure 5). Sites in the western United States generally record lower mercury depositions than eastern sites and were not included in this comparison. The highest mercury depositions in 2005 in the Unites States were reported at selected sites in Florida and some of the Gulf States (Figure 5). The highest wet deposition (21.5 $\mu g/m^2$) was reported at a site in central Florida.

Wet mercury deposition across portions of Pennsylvania in 2005 were well within the range of values reported across North America but generally higher than amounts measured around the Great Lakes and in New England, except for central Pennsylvania where deposition amounts were some of the lowest in the Eastern United States (Figure 5). Precipitation volumes across Pennsylvania in 2005 were, in general, near their long-term averages for each region of the state, although precipitation volumes were much higher in eastern Pennsylvania that either the central or western portions of the state. As a result, 2005 wet mercury depositions were highest in the extreme southeast (Valley Forge) and northwest (Erie) corners of the state and relatively low across the rest of the state (Figure 6). The importance of precipitation volume is very evident when comparing differences in the spatial patterns of mercury concentrations and wet depositions (Figure 6) in 2005. In 2005, mercury concentrations were highest at the Erie site (PA30) even though the highest deposition occurred in the southeast (PA60). Above average precipitation at the Valley Forge site contributed to the relatively high deposition at this site.

Wet mercury deposition in Pennsylvania in 2006 ranged from 8.14 $\mu g/m^2$ at the Arendtsville site (PA00) in Adams County to 13.44 $\mu g/m^2$ at the Valley Forge site (PA60) located approximately 75 miles to the east of Arendtsville (Table 5). If the Valley Forge site is excluded from this comparison because of the high number of invalid samples, the highest annual wet deposition in 2006 (10.11$\mu g/m^2$) occurred at the Milford site (PA72) in Pike County. Unlike previous years, the lowest annual wet mercury deposition in Pennsylvania in 2000 was measured at Arendtsville (PA00) and not at the Hills Creek site (PA90). Total mercury wet deposition in Pennsylvania was higher than observed at most sites in New England, the Mid-Atlantic, and the Great Lakes regions, but lower than generally reported in the southeastern half of the country (Figure 7). Sites in the western United States generally record lower mercury depositions than eastern sites. The highest mercury depositions in the Unites States in 2006 were reported at selected sites in the Mid-west and at most sites around the Gulf States. The highest wet deposition (18.8 $\mu g/m^2$) was reported at a site in the extreme southern portion of Florida (Figure 7).

The 2005 and 2006 spatial patterns in total mercury wet deposition across the state are quite different (Figures 6 and 8). The most striking features are the relatively low and uniform wet deposition rates across the central and southwestern regions of the state in 2005 and the very intense gradient across the southeastern corner of the state between the Millersville (PA47) and Valley Forge (PA60) sites and the Valley Forge and Milford (PA72) sites. In contrast, wet deposition in 2006 was moderately high across most of the western and central portions of the state to high across the eastern region. An intense eastern gradient was evident between the Arendtsville (PA00) and Millersville (PA47) sites and a southern gradient between the Allegheny-Portage site (PA13) and a site located near Frostburg Maryland (MD08). The volume-weighted mean annual total mercury concentration at MD08 in 2006 was 7.5 ng/L, the second lowest annual mean reported in the Mid-Atlantic region in this year (Figure 7).

Seasonal Total Mercury Deposition Patterns

Total Mercury Concentrations - At most MDN sites in North America and Canada, concentrations of total mercury in precipitation exhibit definite seasonal patterns (NADP, 2005). Average summer (June-August) total mercury concentrations for the entire network are generally higher than any other seasonal period and about 1.5 to 2.0 times the concentrations observed during the fall (September-November) and winter (December-February) periods. Although seasonal mean concentration patterns in Pennsylvania are similar to the national network, they are highly variable between sites as well as within sites and between years (Table 4). Mean statewide seasonal total mercury concentrations based on all years of record ranged from 6.37 ng/L (range 5.06 ng/L at PA47 to 7.98 ng/L at PA30) during the winter months to 11.48 ng/L (range 10.73 ng/L at PA47 to 12.30 ng/L at PA13) during the summer. Spring and fall mean concentrations averaged 9.95 ng/L (range 7.64 ng/L at PA90 to 12.42 ng/L at PA60) and 6.74 ng/L (range 5.91 ng/L at PA47 to 7.86 ng/L at PA60), respectively. Between year variability in seasonal mean concentrations of total mercury is illustrated nicely in Figures 9 and 10. Volume-weighted mean concentrations during the spring 2005 were relatively low and quite uniform across the entire state in contrast to spring 2006 concentrations which were much higher and more variable. The reverse was true during the summer months when mean concentrations were much higher and less variable in 2005 than they were in 2006 (Figures 9 and 10).

Volume-weighted seasonal mean total mercury concentrations over the past 10 years ranged from 3.0 ng/L at Milford (PA72) in 2002 to 27.95 ng/L at Allegheny-Portage NHS (PA13) in Cambria County in 2001 (Table 4). Summer mean concentrations exhibited the least variability (based on a ratio of 1.6 between maximum and minimum mean concentrations), while the greatest variability occurred during the fall (a ratio of 2.3) months. At individual sites, the range in seasonal means is about the same, with ratios ranging from 2.5 (PA90 and PA13) to 1.3 at the Millersville site (PA47) which exhibited the least variability regardless of season. Except for Allegheny-Portage NHS (PA13), sites in the eastern half of the state appear to exhibit more seasonal variability that sites in western Pennsylvania (Table 4).

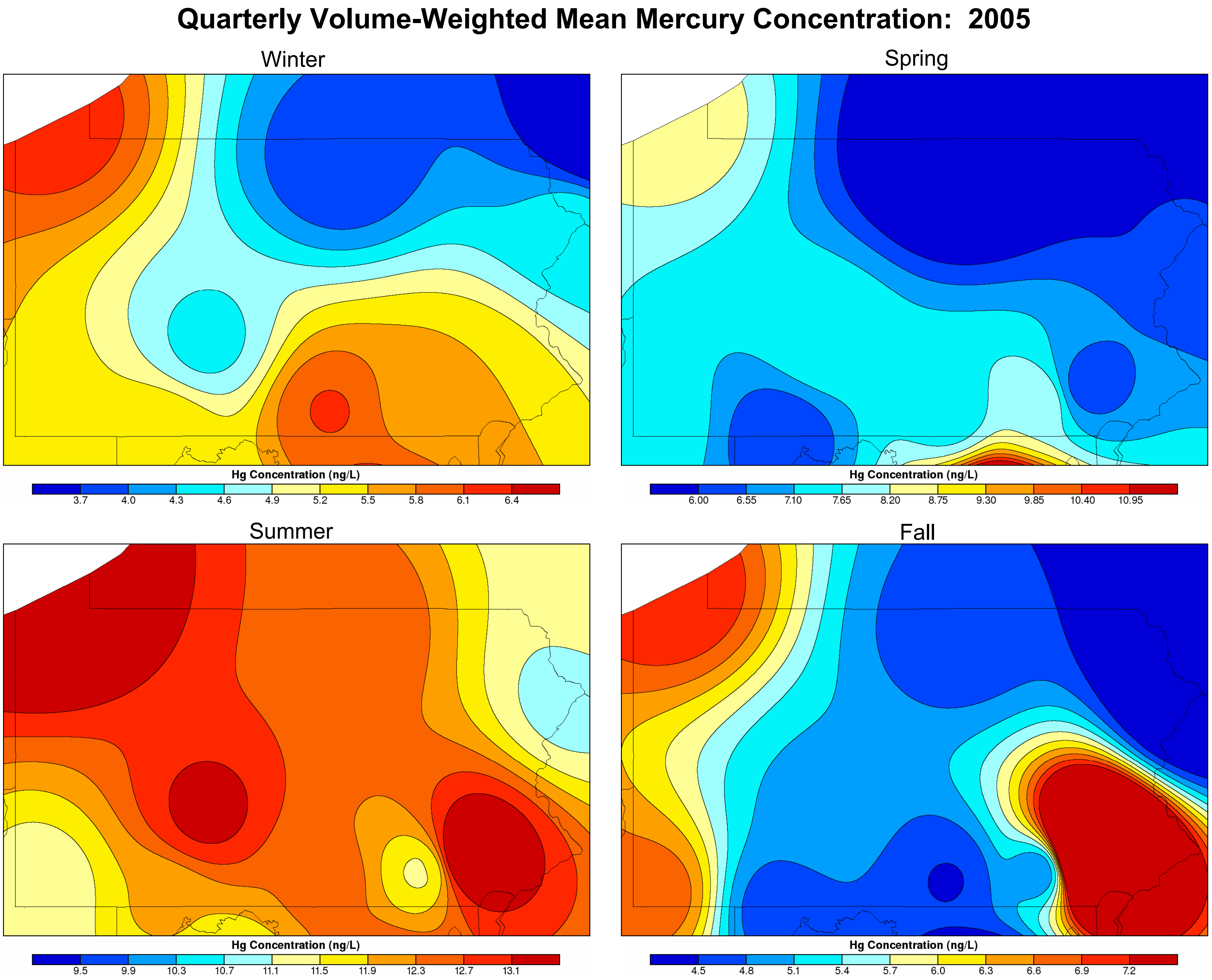


Figure 9. Seasonal variations in volume-weighted mean total mercury concentrations (ng/L) across Pennsylvania in 2005.

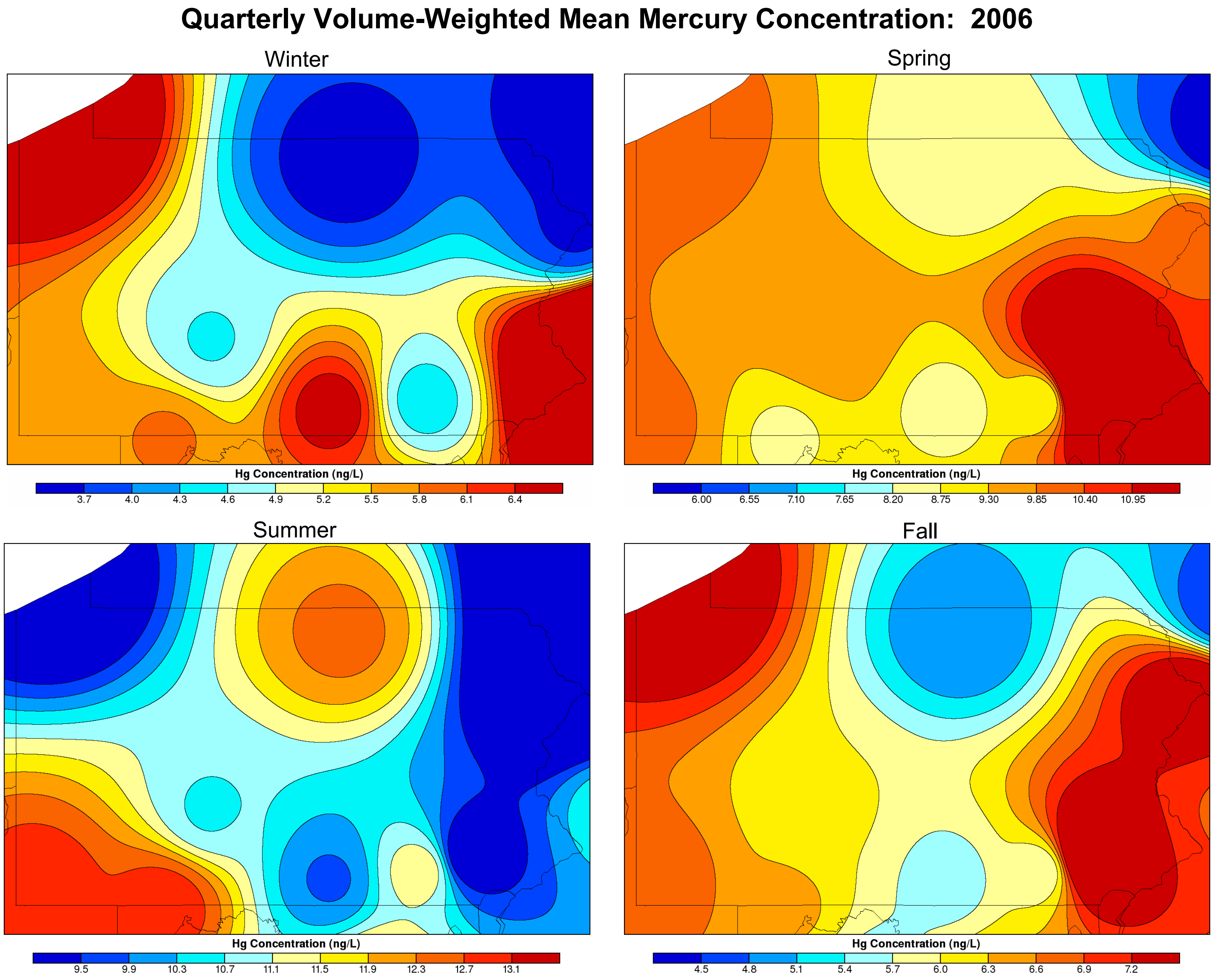


Figure 10. Seasonal variations in volume-weighted mean total mercury concentrations (ng/L) across Pennsylvania in 2006.

Seasonal variability is largely controlled by climatic parameters, such as the form and amount of precipitation, air temperature, atmospheric stability, storm direction, among others and likely seasonal variations in mercury emissions. As noted above, the highest mean seasonal total mercury concentrations generally occurred during the summer months (June-August) and less frequently during the spring months (March-May) and this is exactly the case in Pennsylvania. Given the number of complete years of observations and the variable number of sites in operation from year to year, there are a total of 54 maximum seasonal mean observations across the state since mercury monitoring began in Pennsylvania. Of the 54 possible maximum seasonal means, 38 (70.4%) of them occurred during the summer while 14 (25.9%) occurred during the spring (Table 4). The remaining two maximum seasonal means occurred during the winter, one each at PA30 (Erie) and PA90 (Hills Creek). Only the Valley Forge site (PA60) had a greater number of high seasonal means during the spring (5) than the summer (2) (Table 4). In contrast, the lowest seasonal mean concentrations occurred during the winter (December - February) or fall (September - November) months. Although no consistent pattern is evident for all sites, 63% (34 of the 54 possible minimum seasonal mean concentrations) occurred during the winter months with the remaining minimums being observed during the fall (Table 4). Although the lowest seasonal mean concentrations occurred during the winter at most sites, PA00, PA30 and PA60 reported a greater frequency of minimum seasonal means during the fall (Table 4). Despite the obvious influence that season has on total mercury concentrations, there is no consistent year to year pattern across the state or at any individual site in Pennsylvania with four or more years of observations.

At the two Pennsylvania MDN sites with nine complete years of observations (PA13 and PA90) and thus the best sites to characterize long-term seasonal patterns, the highest seasonal mean concentrations occurred during the summer at both sites, while the lowest mean concentrations occurred most frequently during the winter at PA13 (78%) and less frequently (67%) at PA90. However, during 1999, the winter season mean concentration at PA90 was the highest seasonal mean recorded for that site that year. At PA13, the second highest seasonal mean was observed during the fall months for some years (1998 and 2000) and the lowest in 1997, 2002 and 2004. A review of weekly total mercury concentrations collected during the winter months revealed that some of these “unusually high seasonal means” were a result of one or more unusually high weekly samples. For example, at PA90 (Hills Creek) in 1999 the highest seasonal mean concentration occurred during the winter, not the summer. This three-month period had one weekly sample with a measured total mercury concentration of 671.5 ng/L (See Table 3 for a listing of the maximum weekly concentration measured at each site for each year of operation), which had a profound influence on the magnitude of the 1999 winter mean concentration. Although such spikes in mercury concentrations in Pennsylvania are not common (see Table 3), they do occur periodically but not frequently enough to explain all of the unusual departures in the seasonal distribution of mean total mercury concentrations. Although mercury concentrations in weekly samples are inversely correlated with precipitation volume (Table 6), fluctuations in seasonal precipitation at these long-term sites do not appear to be a major factor. Regression analysis of seasonal mean mercury concentrations and seasonal precipitation volumes showed extremely low correlation for both the Hills Creek and Allegheny-Portage NHS sites.

Total Mercury Wet Deposition – Total mercury wet deposition is also highly variable across the state reflecting not only the previously discussed differences in total mercury concentrations but also temporal and spatial differences in the volume of precipitation. Individual weekly estimates of wet mercury deposition (Table 3) in the state since monitoring began ranged from 0.22 ng/m^2 (PA37, 2002) to 3183.4 ng/m^2 (PA13, 2001). On average, the minimum weekly wet deposition across the network is 13.18 ng/m^2, while the maximum weekly deposition averages 957.41 ng/m^2. Given such a wide range in weekly wet depositions, it is not surprising that seasonal wet deposition estimates shown in Table 5 exhibit considerable variability as well. On a statewide basis, mean seasonal total mercury wet depositions ranged from 1.364 µg/m^2 (range 0.950 µg/m^2 at PA90 to 1.906 µg/m^2 at PA60) during the winter months to 3.459 µg/m^2 (range 2.920 µg/m^2 at PA30 to 4.086 µg/m^2 at PA37) during the summer. Spring wet depositions accounted for about 28% (2.692 µg/m^2, range 1.785 µg/m^2 at PA90 to 3.394 µg/m^2 at PA60) of the average annual deposition. Fall deposition on average accounted for 21% (2.020 µg/m^2, range 1.286 µg/m^2 at PA90 to 2.594 µg/m^2 at PA60) of the annual total mercury deposition. On a percentage basis, summer wet deposition accounted for 36.6% of the annual load while only 14.3% of the annual deposition fell during the winter months. The lowest seasonal mercury deposition (0.560 µg/m^2) measured over the past 10 years was recorded at Hills Creek (PA90) in 2001; the largest amount of mercury deposition for any season (6.429 µg/m^2) was measured at Allegheny-Portage (PA13) in 2001 (Table 5). Although these maximum and minimum values represent extreme observations, every site in the network exhibited fluctuations in weekly and seasonal wet mercury deposition of one or more orders of magnitude each year. The sites that exhibited the greatest amount of seasonal variability are PA90, PA60, and PA13; the sites with the least amount of variability between seasons are PA47, PA00, and PA37.

The largest seasonal fluctuations in wet depositions occurred during the spring and fall months; the smallest fluctuations occurred during the spring and winter months. Some of this variability is likely the result of differences in climatic patterns. Winter and spring months tend to be dominated by frontal storm systems that have more uniform precipitation patterns covering large areas of the state. In contrast, summer climatic patterns are dominated by highly variable convectional storms that can result in non-uniform precipitation patterns and very large volumes of rainfall. The fall represents the transition from convectional to frontal storm patterns which can also contribute to observed spatial differences. Fall months are traditionally the driest months of the year in Pennsylvania, but are periodically influenced by tropical storms, particularly in eastern Pennsylvania.

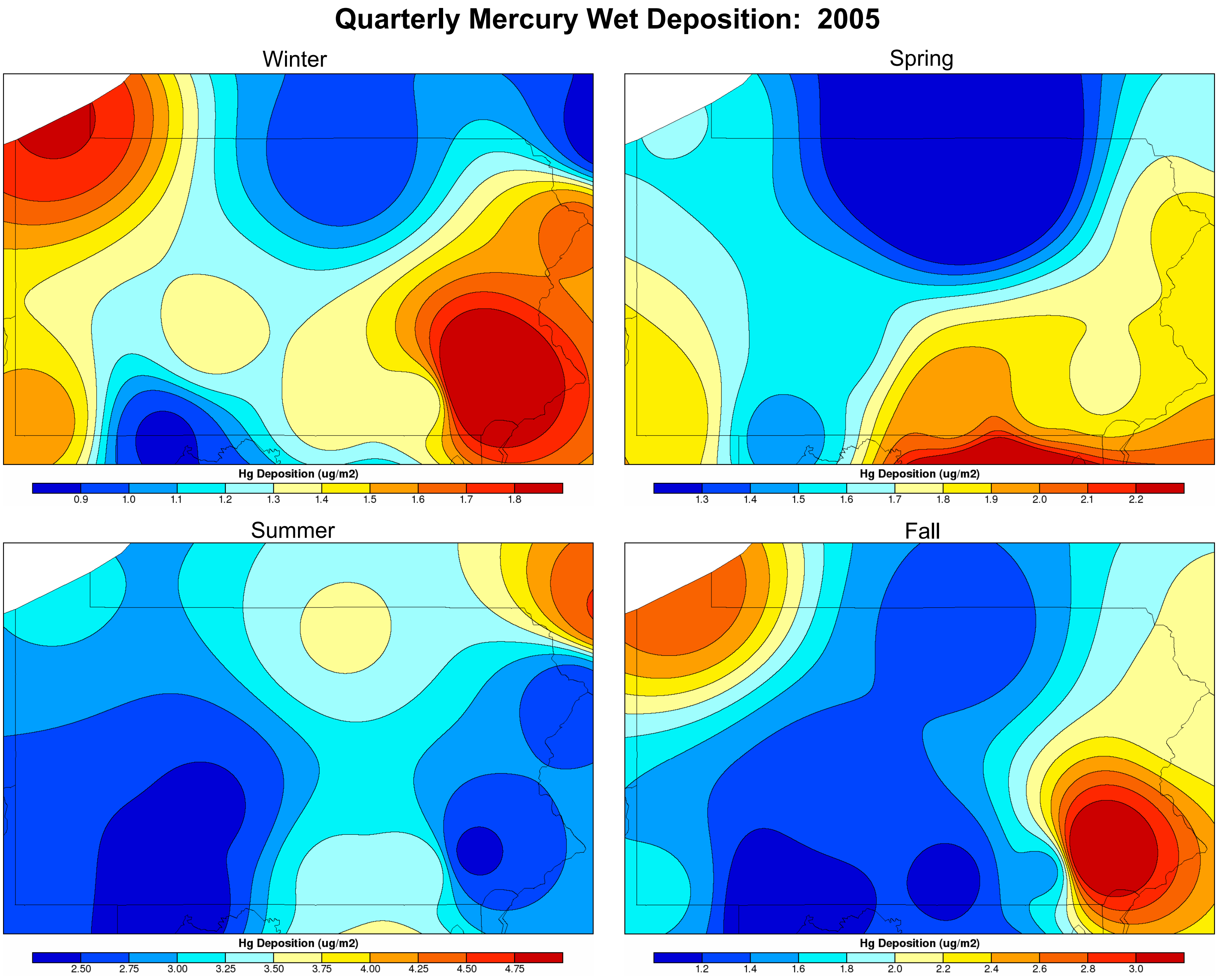
The highest summer wet deposition occurred at the Hills Creek site (PA90); the lowest amount occurred at the Erie site (PA30). The highest winter deposition occurred at Valley Forge (PA60); the lowest winter deposition occurred at the Holbrook site (PA37). Higher mercury depositions during the spring and summer months are a function of both higher mercury concentrations in precipitation and higher rainfall volumes at most sites. Although on average wet mercury deposition is greatest during the summer months, only two-thirds (36) of the 54 maximum seasonal depositions fell during the summer; 25.9% (14 of 54) occurred during the spring. Two sites (PA60 and PA90) recorded their highest seasonal deposition during the winter and two sites (PA30 and PA60) recorded the highest seasonal deposition during the fall. The Valley Forge site exhibited the greatest seasonal variability. However, only PA47 (Millersville) consistently (year to year) recorded the highest deposition doing the summer months. In contrast, the lowest average seasonal deposition occurred during the winter (77.8%). With a couple of exceptions, most of which occurred during the fall, this has been the case at nearly all sites. One of the largest exceptions was recorded at PA30 in 2002 when the lowest seasonal mercury deposition occurred during the summer.

Very high winter mercury deposition was also measured at Hills Creek (PA90) in 1999 and at Erie (PA30) in 2002. Each of these exceptions is associated with one or more weekly samples of very high mercury deposition. For example, during the winter of 2001, the Valley Forge (PA60) site received two storms of both high concentrations and high volumes that together deposited 2.10 $\mu g/m^2$ of mercury, or nearly 70% of the total winter deposition, at this site. At PA90 in 1999, one sample accounted for nearly 60% (1.19 $\mu g/m^2$) of the winter deposition. This particular event was caused primarily by very high total mercury concentrations. Similar conditions existed at the PA30 site in 2002. Such spikes in deposition are unusual, but do occur frequently enough to distort seasonal and/or spatial wet deposition patterns and may have significant impact on the environment.

Seasonal wet deposition patterns for 2005 and 2006 are shown in Figures 11 and 12, respectively. Accept for the summer of 2006, the only consistent seasonal pattern that is evident for both years is the relatively low mercury deposition across the north central portion of the Commonwealth as represented by deposition at the Hills Creek site in Tioga County (PA90). These maps also illustrate nicely that wet mercury deposition in southeastern Pennsylvania during the summer of 2005 (Figure 11) was much lower than for the same period in 2006 (Figure 12). Deposition in this region of Pennsylvania is dominated by measurements at the Valley Forge site (PA60) in Montgomery County. Likewise, spring mercury deposition in 2005 (Figure 11) in the northwest was much lower than it was during the same period in 2006 (Figure 12). Unfortunately, the interpretation and significance of these patterns are limited by the fact that only four years of observations exist in the state with a sufficient number of sites to permit such spatial analyses.

Mercury Emissions versus Total Mercury Depositions

The distribution of major point sources of mercury emissions in Pennsylvania are shown in Figure 13. By far the largest mercury emission sources are located in the western portion of the State with few sources located in the central region. Although no large sources are in eastern Pennsylvania, there are nearly twice as many mercury emissions sources in eastern than in western Pennsylvania. Available annual mercury emissions from each of the sources shown in Figure 13 were summarized by county for 2003 through 2006. These annual countywide mercury emissions are illustrated in Figure 14. Clearly the largest emissions are located in the west central region of Pennsylvania and although the level of emissions fluctuate from year to year the location of the major emissions remain relatively consistent from year to year. These county summaries also illustrate that mercury emissions are very low in the central region and more widespread in the eastern portion of the state (Figure 14). This general distribution of mercury emissions in Pennsylvania likely accounts for some of the spatial pattern across the state. Mercury emissions in adjacent states are likely influencing the spatial pattern across the



Quarterly Mercury Wet Deposition: 2005

Winter

Spring

Summer

Fall

Figure 11. Seasonal variations in total mercury wet depositions (µg/m^2) across Pennsylvania in 2005.

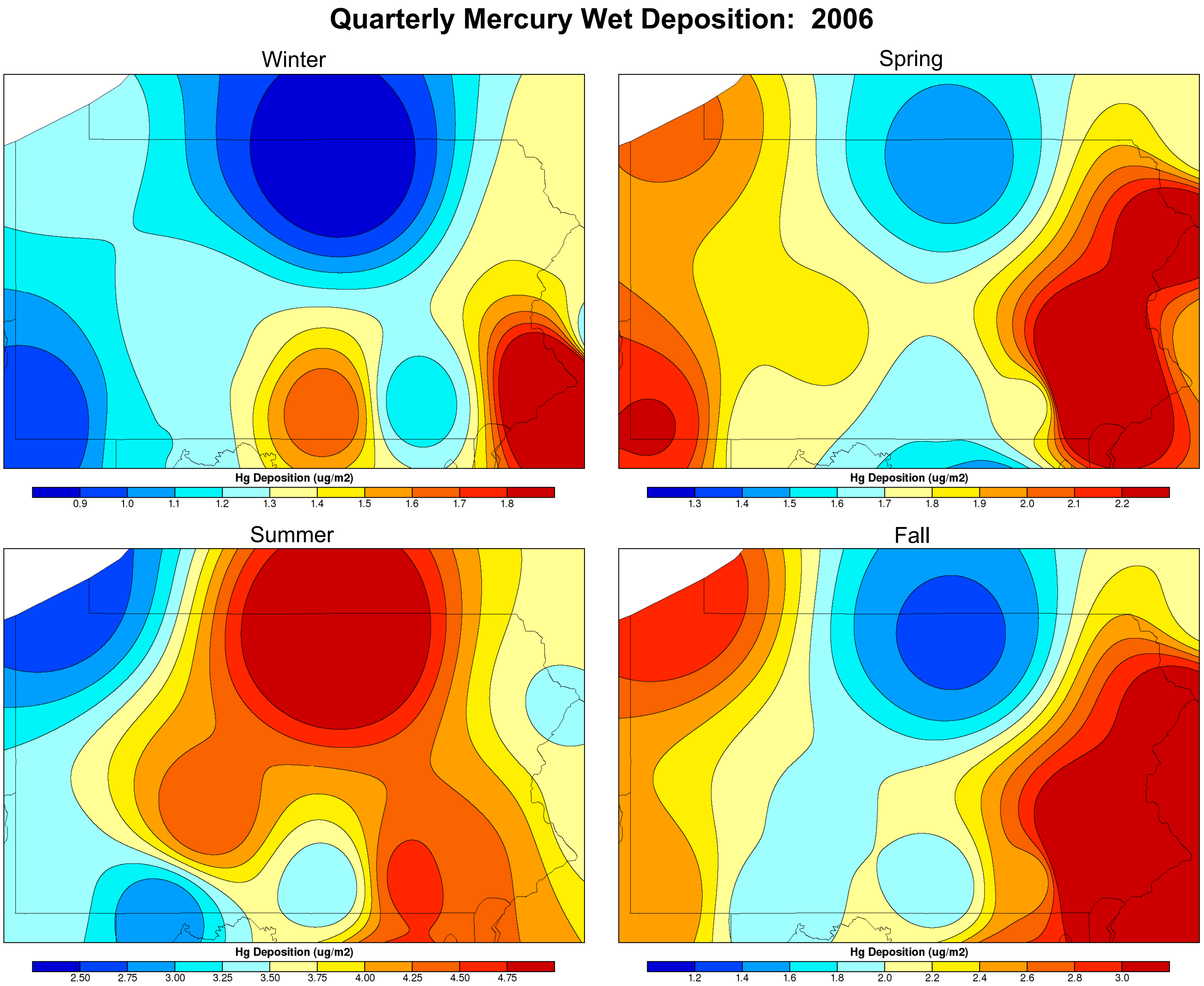


Figure 12. Seasonal variations in total mercury wet depositions (μg/m^2) across Pennsylvania in 2006.

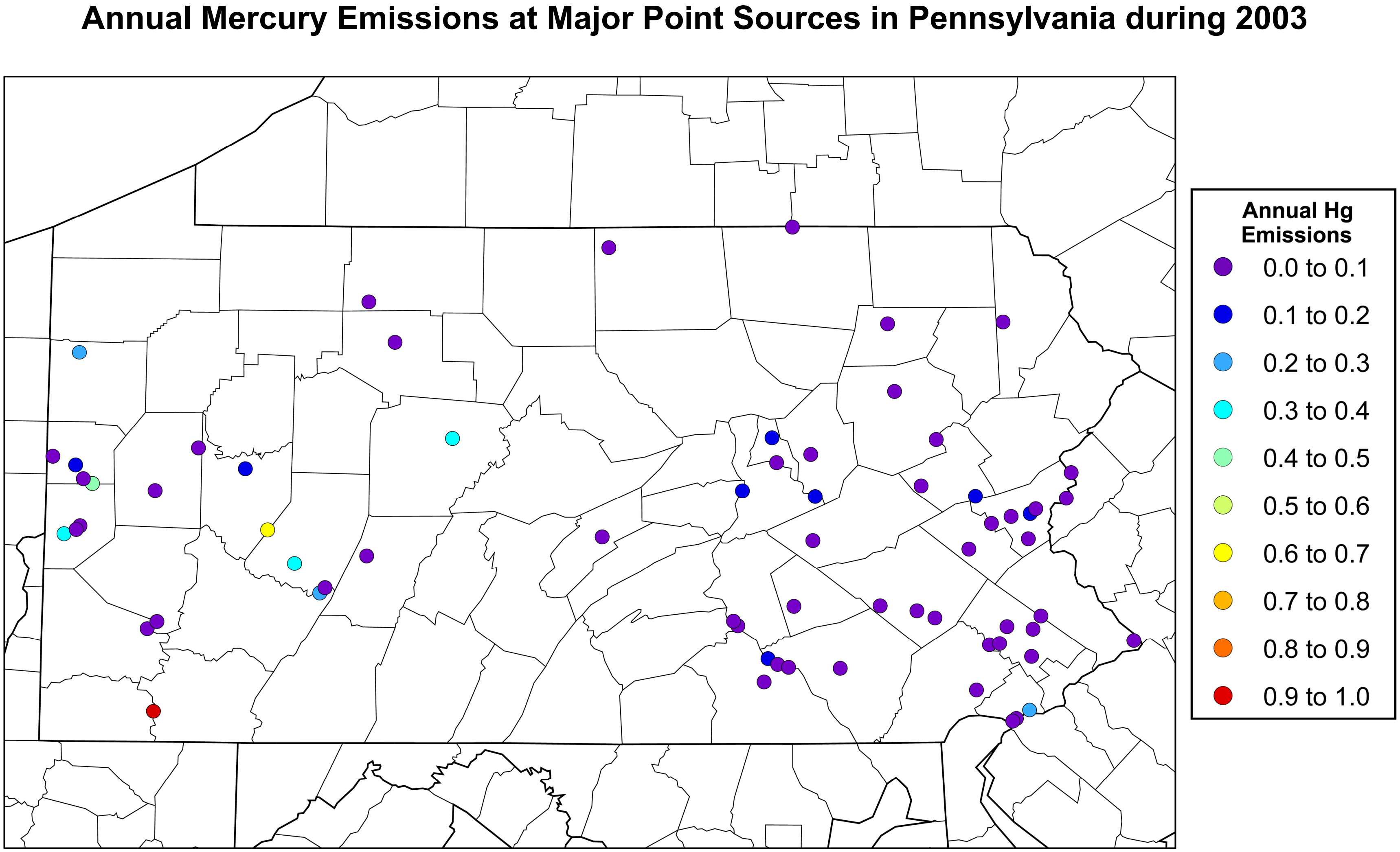


Figure 13. Location of annual mercury emissions (tons/year) at major point sources in Pennsylvania in 2003 through 2006.

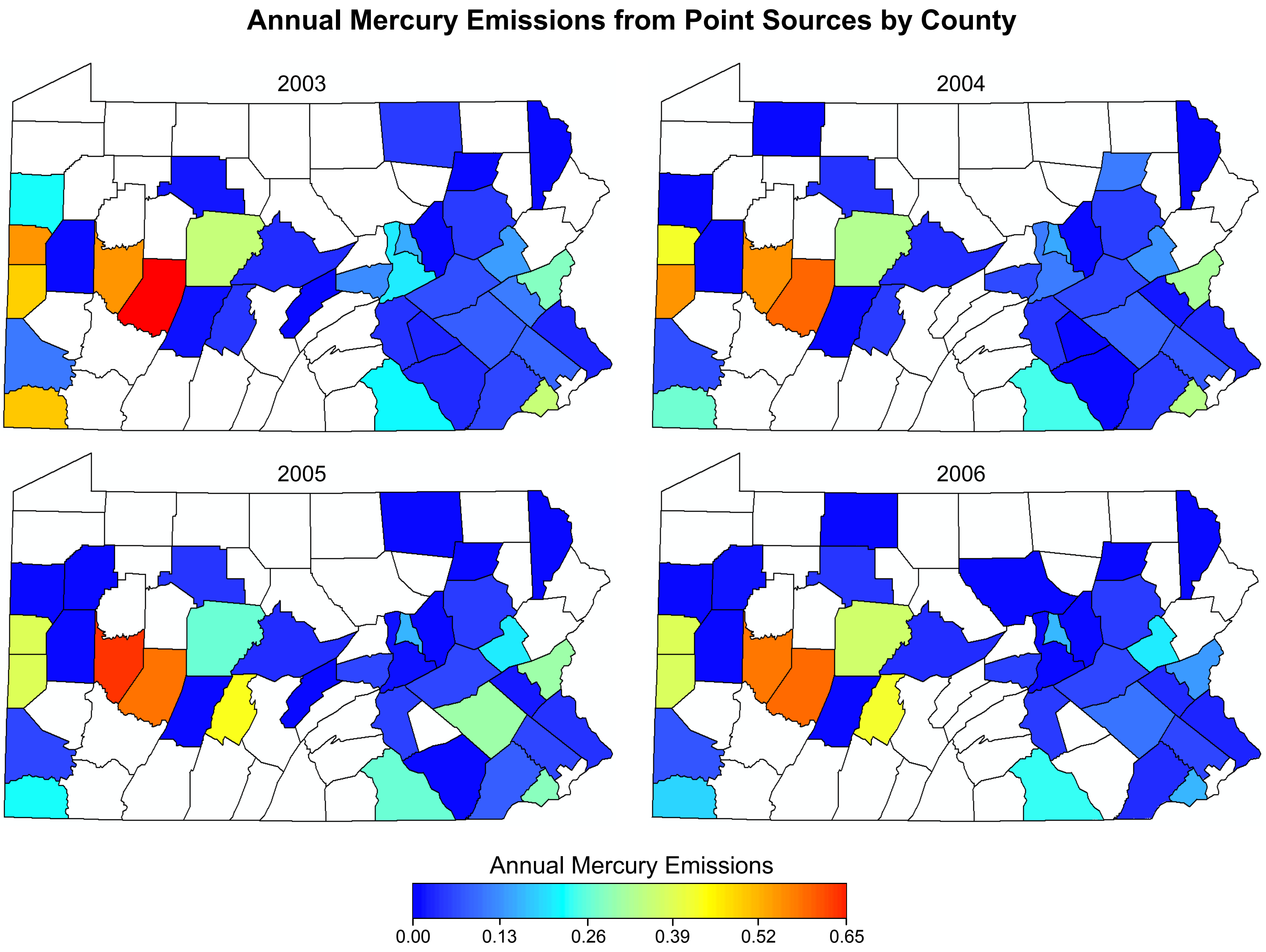


Figure 14. County summary of annual mercury emissions (tons/year) at major point sources in Pennsylvania in 2003 through 2006.

state as well, especially in the eastern and northwestern regions of Pennsylvania. It should also be noted that these observations are based on a visual correlation of annual total mercury concentrations (Figure 15) and wet depositions (Figure 16) and the county summaries of annual mercury emissions from major point sources in the state in 2003 through 2006, and not on a statistical analysis. Patterns of wet deposition of emitted materials are strongly determined by the interactions of lower- and upper-level winds, atmospheric lifting, storm path, and the timing of precipitation events. These meteorological determinants of deposition can exhibit dramatic year-to-year, season-to-season, and even event-to-event variability. Therefore, the only satisfactory way to identify and assess source-receptor relationships is to utilize a dynamic atmospheric transport model that incorporates meteorological parameters to define the pathways between emission sources and deposition sites on a per-event basis. Nevertheless, data presented in Figures 14 and 15 do appear to be consistent, at least from the standpoint that where you have large or numerous emissions sources you are likely to find your highest concentrations of mercury in precipitation. However, one must also bear-in-mind that mercury emissions to the atmosphere in the United States, including Pennsylvania, are broadly classified as re-emitted mercury, natural mercury emissions, and anthropogenic mercury emissions. Re-emitted mercury is mercury that was previously deposited on the Earth’s surface following either anthropogenic or natural releases and is re-emitted to the atmosphere by natural, biologic or geologic processes. Natural mercury emissions occur when geologically-bound mercury is released during natural processes, such as volcanic eruptions, geothermal releases, and from naturally enriched substances. The role that natural and re-emitted mercury emissions play relative to total mercury concentrations and wet deposition in the Commonwealth is unknown. Only anthropogenic mercury emissions in Pennsylvania are depicted in Figure 14. However, mercury concentrations and wet depositions shown in Figures 15 and 16 represent a composite of all emission sources including contributions from the global cycle.

Correlation and Stepwise Regression Analyses

Correlation and stepwise regression analyses between mercury and cation and anion concentrations in precipitation provide potential insight into where some of the mercury might be coming from at a given site. Each MDN site in Pennsylvania is collocated with a precipitation sampler that provides weekly samples for analysis of the concentrations of major cation and anion in precipitation. Pearson correlation coefficients between weekly mercury concentrations and the concentrations of the major cations (calcium, magnesium, potassium, ammonium and sodium), anions (sulfate, nitrate, and chloride), and precipitation volumes are shown in Table 6 for all MDN sites in the state. The results illustrate that sulfate and nitrate are positively correlated with total mercury concentrations at each site and all of the coefficients are statistically significant (Table 6). The same is also true for calcium, magnesium and potassium concentrations. The correlation coefficients between weekly mercury concentrations and sodium and chloride are much weaker, although still statically significant at some sites, namely PA37, PA60, PA72 and PA90. Ammonium concentrations are positively correlated with mercury concentrations at all sites, although the coefficients are not statistically significant at PA30 and PA47. As might be expected, precipitation volumes are inversely correlated and statistically significant with mercury concentrations at all sites (Table 6).

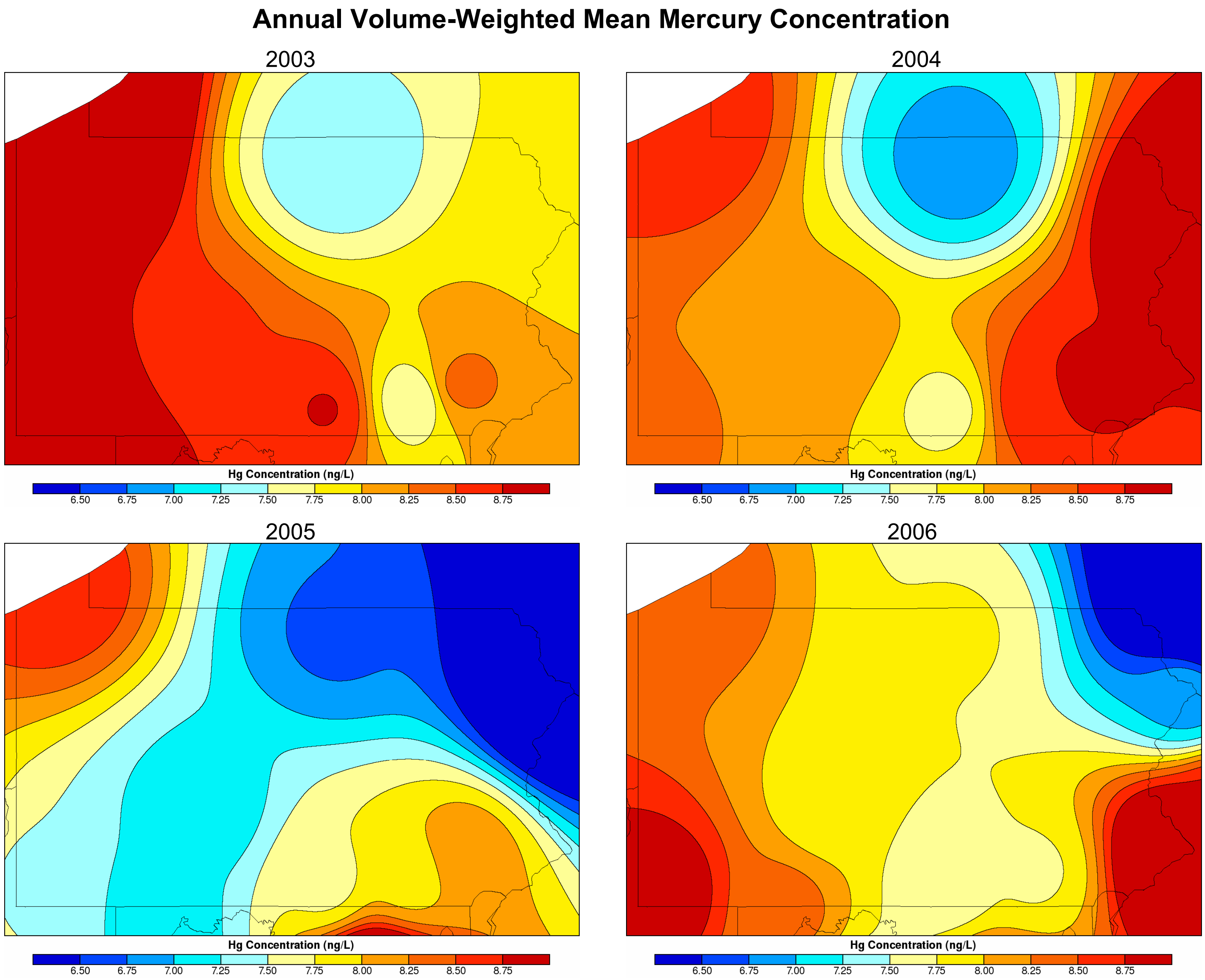


Figure 15. Spatial interpolations of volume-weighted mean annual total mercury concentrations (ng/L) across Pennsylvania in 2003 through 2006.

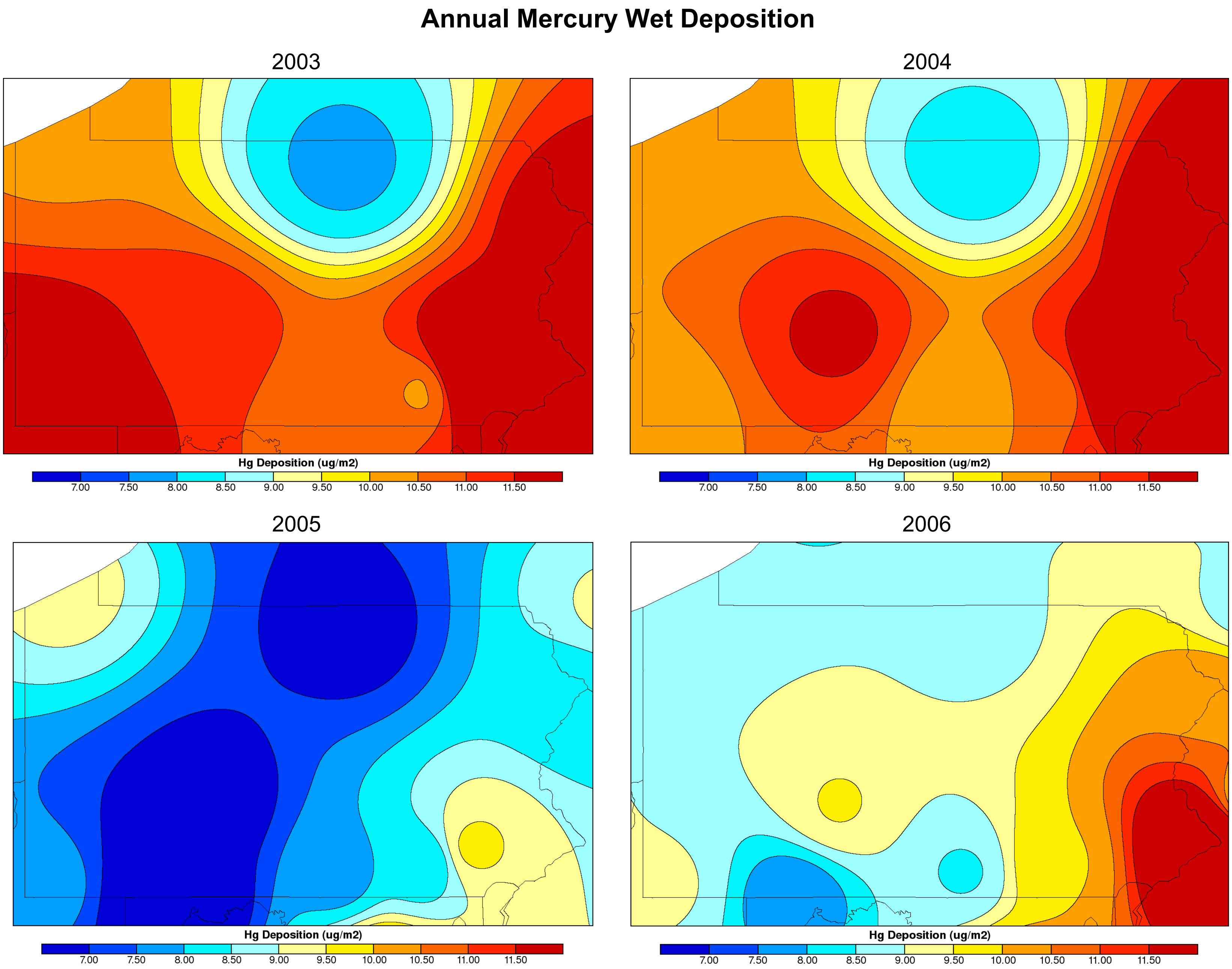


Figure 16. Spatial interpolations of annual total mercury wet deposition across Pennsylvania in 2003 through 2006.

Table 6. Pearson correlation coefficients (*r*) between total mercury and the major cation and anion concentrations in precipitation as measured at eight Pennsylvania collocated mercury (MDN) and acid rain monitoring sites. All concentrations were log-transformed. Precipitation was transformed by taking its square root.

Site	sulfate	nitrate	chloride	calcium	magnesium	potassium	sodium	ammonium	precipitation
PA00									
r	0.607	0.566	0.165	0.610	0.526	0.481	0.110	0.434	−0.427
Prob>|*r*|	0.0001	0.0001	0.0412	0.0001	0.0001	0.0001	0.1839	0.0001	0.0001
No. Obs.	148	148	154	148	148	148	148	154	169
PA13									
r	0.617	0.480	−0.016	0.432	0.374	0.333	0.092	0.150	−0.360
Prob>|*r*|	0.0001	0.0001	0.8290	0.0001	0.0001	0.0001	0.2341	0.0432	0.0001
No. Obs	171	171	183	168	168	168	168	183	185
PA30									
r	0.593	0.477	−0.057	0.468	0.450	0.258	0.273	0.040	−0.438
Prob>|*r*|	0.0001	0.0001	0.4474	0.0001	0.0001	0.0007	0.0003	0.5902	0.0001
No. Obs	168	168	180	168	168	168	168	180	181
PA37									
r	0.427	0.215	0.158	0.320	0.405	0.281	0.161	0.302	−0.143
Prob>|*r*|	0.0001	0.0053	0.0369	0.0001	0.0001	0.0002	0.0377	0.0001	0.0597
No. Obs	167	167	174	167	167	167	167	174	174
PA47									
r	0.648	0.642	0.149	0.717	0.563	0.552	0.022	0.138	−0.043
Prob>|*r*|	0.0001	0.0001	0.5368	0.0001	0.0001	0.0001	0.7932	0.0819	0.001
No. Obs	140	140	161	140	140	140	140	161	170
PA60									
r	0.615	0.540	0.207	0.508	0.453	0.462	0.257	0.345	−0.508
Prob>|*r*|	0.0001	0.0001	0.0094	0.0001	0.0001	0.0001	0.0019	0.0001	0.0001
No. Obs	145	145	156	142	142	142	142	156	157
PA72									
r	0.700	0.631	0.148	0.688	0.536	0.591	0.069	0.509	−0.422
Prob>|*r*|	0.0001	0.0001	0.0499	0.0001	0.0001	0.0001	0.3830	0.0001	0.0001
No. Obs	161	161	176	161	161	161	161	176	180
PA90									
r	0.689	0.528	0.182	0.608	0.667	0.388	0.394	0.441	−0.285
Prob>|*r*|	0.0001	0.0001	0.0129	0.0001	0.0001	0.0001	0.0001	0.0001	0.0001
No. Obs	179	179	186	178	178	178	178	186	187

Log_{10} transformed weekly cation and anion concentrations were included in a stepwise regression analyses with dependent variable log_{10} mercury concentrations. The statistically significant ($p<0.05$) variables and their order of selection are shown in Table 7. The first and most statistically significant variable in predicting mercury concentrations at PA13, PA30, PA37, PA60, PA72, and PA90 was sulfate concentrations. At PA00 and PA47, calcium concentrations were selected first followed by sulfate. At these sites where sulfate was the primary variable, the second most important variable in each model was either calcium (PA30, PA72), magnesium (PA37 and PA90), or potassium (PA60). Only two sites (PA60 and PA90) had more than two variables in their predictive model. At both sites calcium was selected as the third most significant variable. At PA90, ammonium was also included in the model. The model r-squared values, a measure of the amount of explained variation between dependent and independent variables, ranged from 0.23 at PA37 to 0.62 at PA90 with most model r-squared values exceeding 0.40 (Table 7). When included in model development, precipitation volume was the second most significant predictive variable at PA30 and PA60, replacing one of the cations (Table 7). Precipitation was not a significant predictive variable of mercury concentrations in precipitation at any of the other MDN sites in Pennsylvania.

Sulfate concentrations in precipitation result from the oxidation of sulfur dioxide released to the atmosphere during combustion. The primary source of sulfur dioxide in Pennsylvania is from coal-fired power plants. We also know that sulfate concentrations in precipitation are highest during the summer and lowest during the winter months, the same temporal pattern that is frequently observed for mercury concentrations at all MDN sites in Pennsylvania. We also know that coal combustion is one of the primary sources of mercury emissions to the atmosphere in the eastern United States (EPA, 2002). The close association of sulfate concentrations with mercury concentrations as illustrated by the correlation coefficients (Table 6) and stepwise regression models (Table 7) appears to indicate that at least some of the mercury in precipitation at all Pennsylvania MDN sites is from combustion sources, most likely coal-fired power plants. At PA13 (Allegheny-Portage) sulfate is the only variable that is significantly associated with mercury concentrations at that site. This site is located on top of Cresson Mountain and is downwind of several large coal-fired utilities located in western Pennsylvania and the largest mercury emission sources in the state (Figure 14).

Wind blown soil particles are an important source of cations in precipitation, particularly calcium and magnesium. The close association of these cations with mercury concentrations would seem to indicate that potential terrestrial/agricultural sources are important at all sites. The fact that calcium is the most significant variable in predicting mercury concentrations at the Arendtsville (PA00) and Millersville (PA47) sites would tend to support this position. Both of these sites are located in areas that are dominated by agricultural activities, particularly at PA47 were calcium concentrations explain more than 50% of the variance in weekly mercury concentrations. Lindberg et al. (1998) reported that mercury is constantly recycled between terrestrial ecosystems and the atmosphere. How agricultural activities would affect the release of mercury cannot be determined by this analysis. However, mercury has been used extensively in agriculture for many years and may be a contributing factor. In addition, soil disturbance in agricultural areas and the subsequent increase in wind erosion may also be factor.

Table 7. Summary of stepwise regression analyses with dependent variable $\log_{10}$Mercury concentrations and $\log_{10}$ transformed major cation and anion concentrations in precipitation collected at 8 Pennsylvania collocated mercury (MDN) and acid rain monitoring sites. All variables in each model are significant at the 0.050 level or higher.

Site	Variables In Model	Partial R-square	Model R-square	C(p) Value	F-value	Pr > F
PA00	Ca^{2+}	0.372		20.699	86.45	< 0.0001
	SO_4^{2-}	0.080	0.45	1.864	21.00	< 0.0001
PA13	SO_4^{2-}	0.393	0.39	0.396	107.57	< 0.0001
PA30	SO_4^{2-}	0.352		17.960	90.29	< 0.0001
	Ca^{2+}	0.052	0.40	5.433	14.32	0.0002
PA37	SO_4^{2-}	0.182		12.063	36.82	< 0.0001
	Mg^{2+}	0.046	0.23	4.200	9.79	0.0021
PA47	Ca^{2+}	0.514		15.875	145.94	< 0.0001
	SO_4^{2-}	0.034	0.55	7.399	10.15	0.0018
PA60	SO_4^{2-}	0.364		17.971	80.06	< 0.0001
	K^+	0.054		6.867	12.75	0.0005
	Ca^{2+}	0.019	0.44	4.283	4.57	0.0342
PA72	SO_4^{2-}	0.490		37.696	152.47	< 0.0001
	Ca^{2+}	0.089	0.58	5.696	33.43	< 0.0001
PA90	SO_4^{2-}	0.476		61.253	159.67	< 0.0001
	Mg^{2+}	0.119		9.635	51.66	< 0.0001
	Ca^{2+}	0.015		4.857	6.74	0.0102
	NH_4^+	0.009	0.62	2.783	4.13	0.0438
-----------If the square root of precipitation is included as an independent variable----------						
PA30	SO_4^{2-}	0.352		24.702	90.29	< 0.0001
	Ppt	0.056		10.384	15.62	0.0001
	Mg^{2+}	0.025	0.43	5.079	7.26	0.0078
PA60	SO_4^{2-}	0.364		22.341	80.06	< 0.0001
	Ppt	0.060		9.222	14.47	0.0002
	K^+	0.041	0.46	1.027	10.42	0.0016

Correlation and stepwise regression analyses were also conducted on weekly wet mercury depositions and the depositions of major cations and anions measured at each MDN site. The correlation coefficients are presented in Table 8. Results of the stepwise regression analyses are presented in Table 9. Results from the wet deposition analysis are similar to the concentration analysis in that sulfate concentrations are the primary and most statistically significant predictive variable in the regression models at all MDN sites. With the exception of the Holbrook site (PA37), wet sulfate deposition accounted for nearly 70% of the variability in wet mercury deposition at each MDN site (Table 9). The much higher r-squared values for the wet deposition analyses versus the concentration analyses results from the fact that precipitation volumes used to calculate wet deposition is the same for both sulfate and mercury wet deposition calculations. It should also be noted that although cation wet depositions were important in each site's model, they were not the primary variable at any site as they were for the concentration analysis. In addition, other ions such as sodium, chloride, and nitrate were significant variables at some sites (e.g., PA30, PA72 and PA90). The significance of these variables in explaining wet mercury deposition is not clear, although in all cases, their contribution (based on partial r-squared values) to each model is relatively small and may be indicative of broad regional land use activities or a function of long-range transport of pollutants including mercury to individual sites.

Trends in Total Mercury Deposition

Ten years of observations exist at the Hills Creek (PA90) and Allegheny-Portage NHS (PA13) sites; thus, these sites represent the best opportunity to assess trends in mercury concentrations and wet depositions. Trend analyses were also conducted at the Valley Forge site (PA60) and the Holbrook site (PA37), both of which have seven years of weekly observations. At the Hills Creek site, total mercury concentrations have decreased from 1997 through 2006 (Figure 17). The decreasing trend (4.37% per year) is statistically significant ($p<0.0016$). The decline is not driven by trends in precipitation volume, although precipitation volumes are likely a contributing factor. Seasonal mercury concentrations and precipitation volumes at this site are not statistically related, although weekly volumes are inversely correlated with weekly mercury concentrations (Table 6). Despite the decline in mercury concentrations at this site, wet mercury deposition has actually increased slightly (Figure 18), although the trend is not statistically significant ($p<0.01$). The lack of a significant wet mercury deposition trend at the Hills Creek site is strongly influenced by very high precipitation volumes and wet depositions in 2003, 2004 and 2006. Similar mercury concentration (Figure 19) and wet deposition (Figure 20) trends are evident at the Allegheny Portage NHS site in Cambria County and like Hills Creek, the concentration trend is statistically significant ($p=0.0001$) while the deposition trend is not. The rate of decline in mercury concentrations at PA13 is 4.56% per year. Like the Hills Creek site, there is no statistical evidence to indicate that precipitation volumes at the Allegheny-Portage site have influenced the mercury concentration trend. The decreasing seasonal total mercury concentration trend (Figure 21) of 4.31% per year at the Holbrook (PA37) site is also significant ($p=0.0051$) while the wet deposition trend (Figure 22) is not. At Valley Forge (PA60), neither the seasonal mercury concentration (Figure 23) nor wet deposition (Figure 24) trends are significant. In fact, the concentration trend at Valley Forge is level while the wet deposition pattern appears to be increasing slightly as a result of a general increasing pattern in precipitation volume since 2000 (Figure 24).

Table 8. Pearson correlation coefficients (*r*) between wet total mercury deposition and cation and anion wet depositions as measured at eight Pennsylvania collocated mercury (MDN) and acid rain monitoring sites. All depositions were log-transformed. Precipitation was transformed by taking its square root.

Site	sulfate	nitrate	chloride	calcium	magnesium	potassium	sodium	ammonium	precipitation
PA00									
r	0.819	0.779	0.635	0.739	0.741	0.752	0.538	0.690	0.781
Prob>|*r*|	0.0001	0.0001	0.0001	0.0001	0.0001	0.0001	0.0001	0.0001	0.0001
No. Obs.	148	148	154	148	148	148	148	154	169
PA13									
r	0.823	0.708	0.484	0.541	0.517	0.464	0.422	0.656	0.778
Prob>|*r*|	0.0001	0.0001	0.0001	0.0001	0.0001	0.0001	0.0001	0.0001	0.0001
No. Obs	171	171	183	168	168	168	168	183	185
PA30									
r	0.825	0.689	0.441	0.593	0.553	0.528	0.565	0.552	0.751
Prob>|*r*|	0.0001	0.0001	0.0001	0.0001	0.0001	0.0007	0.0001	0.0001	0.0001
No. Obs	168	168	180	168	168	168	168	180	181
PA37									
r	0.612	0.398	0.330	0.366	0.472	0.370	0.353	0.406	0.788
Prob>|*r*|	0.0001	0.0001	0.0001	0.0001	0.0001	0.0001	0.0001	0.0001	0.0001
No. Obs	167	167	174	167	167	167	167	174	174
PA47									
r	0.849	0.803	0.530	0.787	0.743	0.719	0.421	0.510	0.753
Prob>|*r*|	0.0001	0.0001	0.0001	0.0001	0.0001	0.0001	0.0001	0.0001	0.001
No. Obs	140	140	161	140	140	140	140	161	170
PA60									
r	0.788	0.674	0.503	0.565	0.623	0.550	0.540	0.609	0.789
Prob>|*r*|	0.0001	0.0001	0.0001	0.0001	0.0001	0.0001	0.0001	0.0001	0.0001
No. Obs	145	145	156	142	142	142	142	156	157
PA72									
r	0.819	0.763	0.584	0.738	0.685	0.716	0.421	0.686	0.727
Prob>|*r*|	0.0001	0.0001	0.0001	0.0001	0.0001	0.0001	0.0001	0.0001	0.0001
No. Obs	161	161	176	161	161	161	161	176	180
PA90									
r	0.849	0.745	0.565	0.683	0.746	0.562	0.658	0.691	0.750
Prob>|*r*|	0.0001	0.0001	0.0001	0.0001	0.0001	0.0001	0.0001	0.0001	0.0001
No. Obs	179	179	186	178	178	178	178	186	187

Table 9. Summary of stepwise regression analyses of $\log_{10}$-transformed total mercury wet deposition against $\log_{10}$-transformed major cation and anion wet depositions measured at eight Pennsylvania collocated mercury (MDN) and acid rain monitoring sites. All predictors the models are significant at $p < 0.05$.

Site	Variables In Model	Partial R-square	Model R-square	C(p) Value	F-values	P > F
PA00	SO_4^{2-}	0.670		17.550	296.42	< 0.0001
	Ca^{2+}	0.029		5.256	14.07	0.0003
	NO_3^{-}	0.009	0.71	2.632	4.67	0.0324
PA13	SO_4^{2-}	0.680		11.928	353.38	< 0.0001
	Na^{+}	0.019	0.70	3.230	10.68	0.0013
PA30	SO_4^{2-}	0.680		17.228	353.08	< 0.0001
	Na^{+}	0.024		5.509	13.51	0.0003
	Cl^{-}	0.011	0.71	1.557	6.04	0.0150
PA37	SO_4^{2-}	0.375		10.856	98.82	< 0.0001
	NH_4^{+}	0.022		6.651	6.07	0.0148
	K^{+}	0.016	0.41	4.156	4.49	0.0356
PA47	SO_4^{2-}	0.721		16.254	357.16	< 0.0001
	Ca^{2+}	0.027	0.75	3.535	33.43	0.0002
PA60	SO_4^{2-}	0.634		15.543	242.33	< 0.0001
	K^{+}	0.030		4.941	12.43	0.0006
	Na^{+}	0.012	0.68	1.802	5.22	0.0238
PA72	SO_4^{2-}	0.671		43.051	324.51	< 0.0001
	Ca^{2+}	0.043		18.972	23.69	< 0.0001
	Cl^{-}	0.012		13.764	6.79	0.0101
	K^{+}	0.009		10.417	5.17	0.0244
	NO_3^{-}	0.010	0.75	6.085	6.33	0.0129
PA90	SO_4^{2-}	0.714		55.380	439.32	< 0.0001
	Mg^{2+}	0.050		17.568	36.75	< 0.0001
	Na^{+}	0.011		10.751	8.49	0.0040
	NH_4^{+}	0.007	0.78	7.379	5.30	0.0225

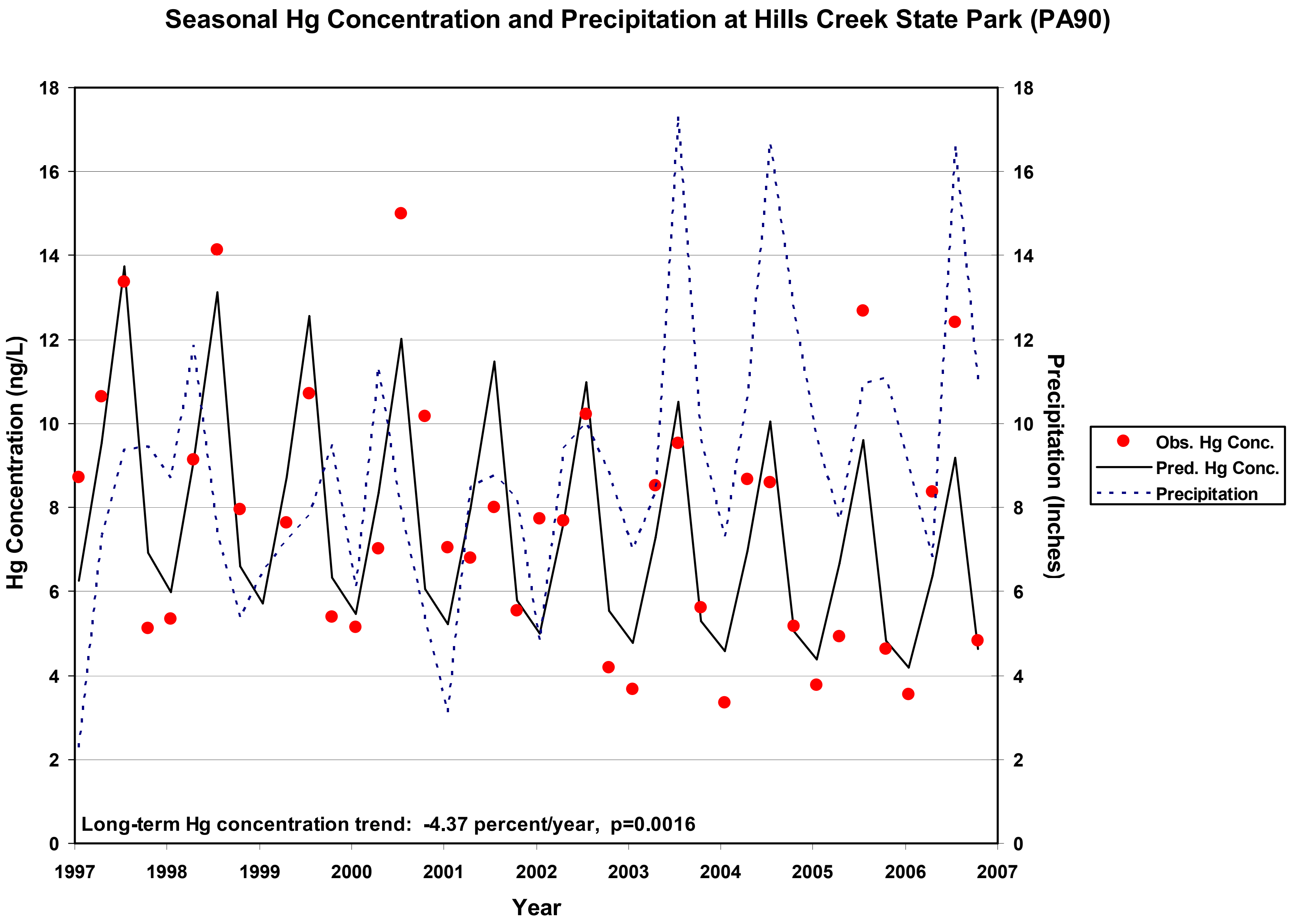


Figure 17. Seasonal trends in total mercury concentrations and precipitation at the Hills Creek State Park MDN site in Tioga County Pennsylvania from 1997 through 2006.

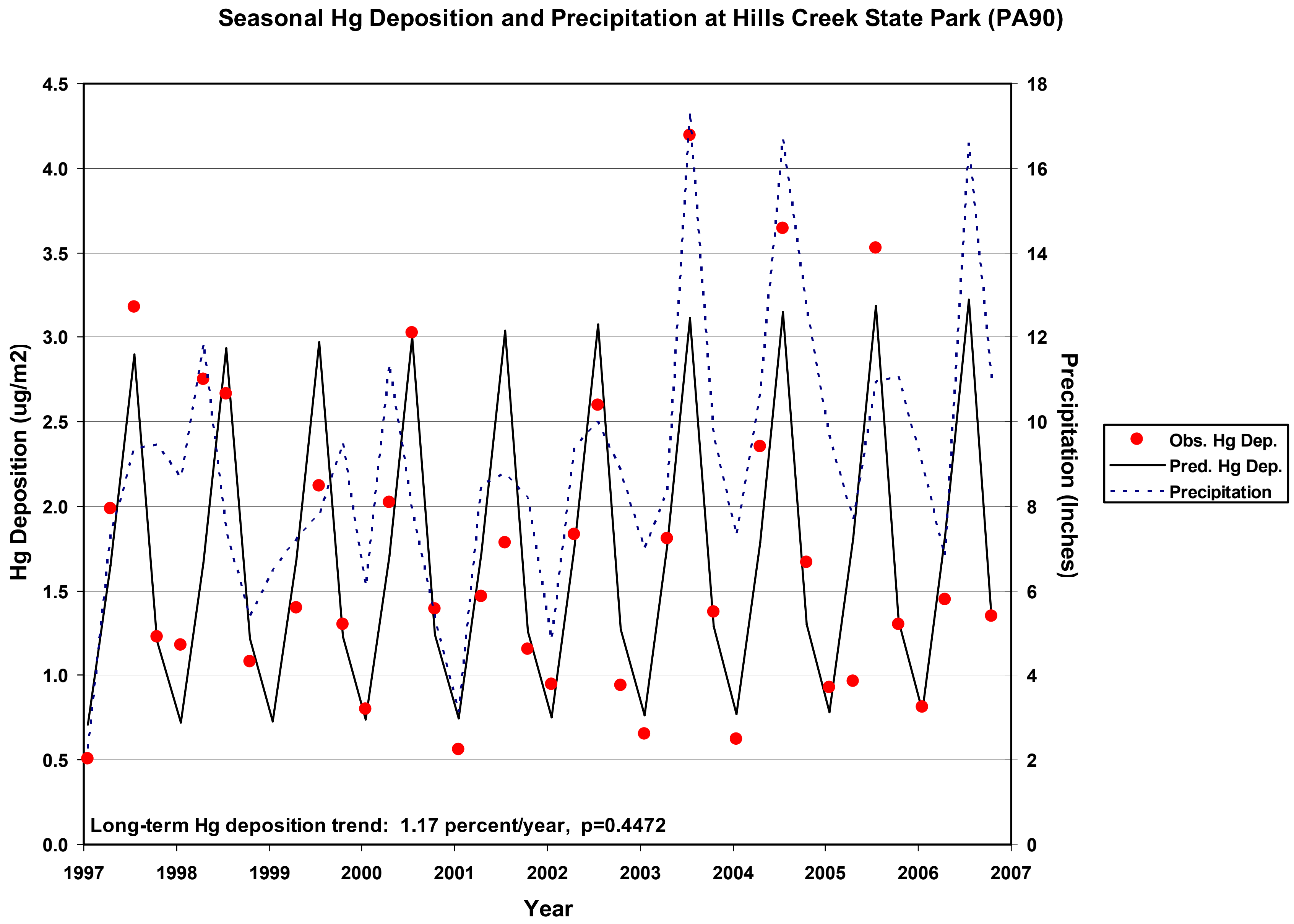


Figure 18. Seasonal trends in total mercury wet deposition and precipitation at the Hills Creek State Park MDN site in Tioga County Pennsylvania from 1997 through 2006.

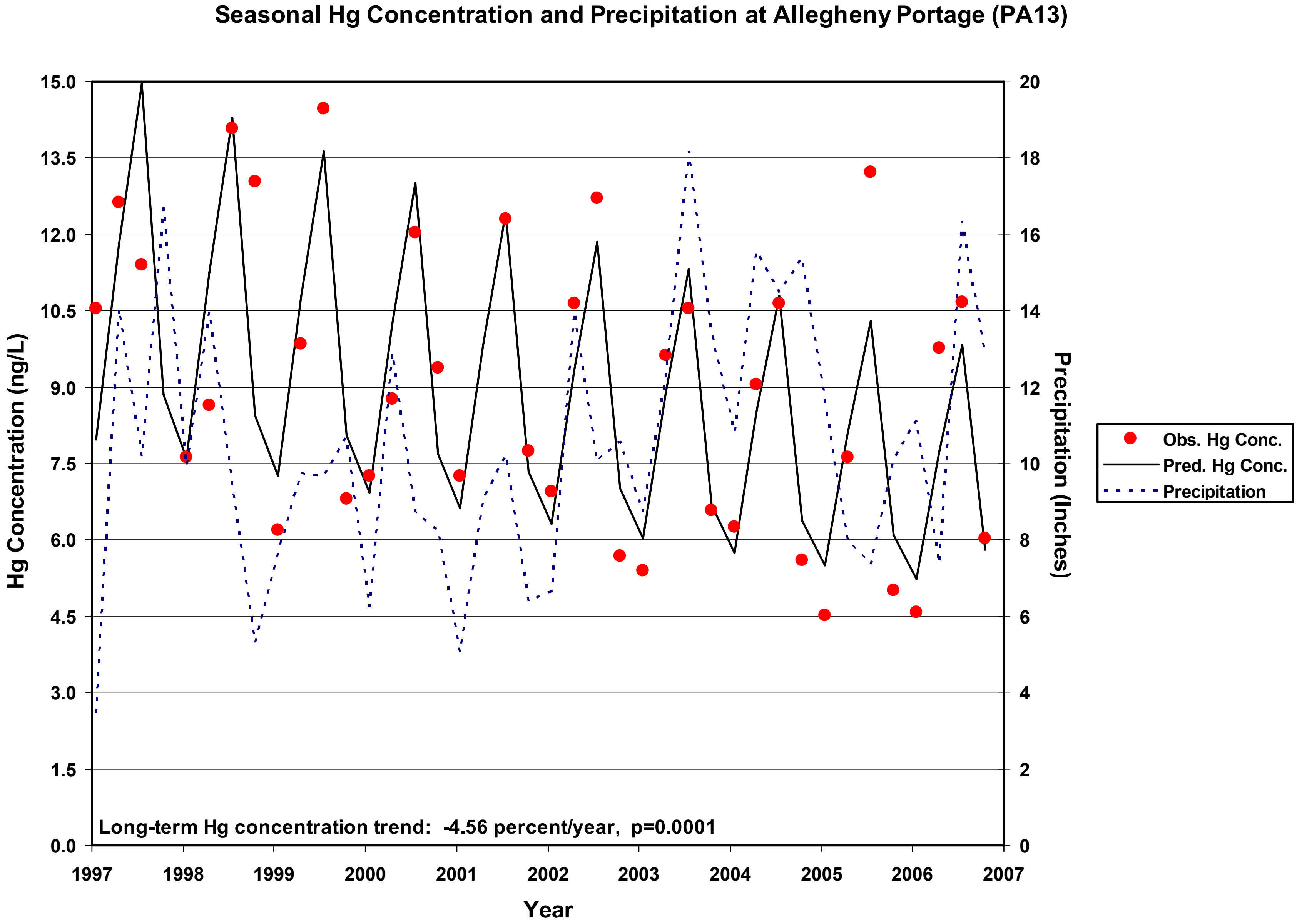


Figure 19. Seasonal trends in total mercury concentrations and precipitation at the Allegheny Portage MDN site in Cambria County Pennsylvania from 1997 through 2006.

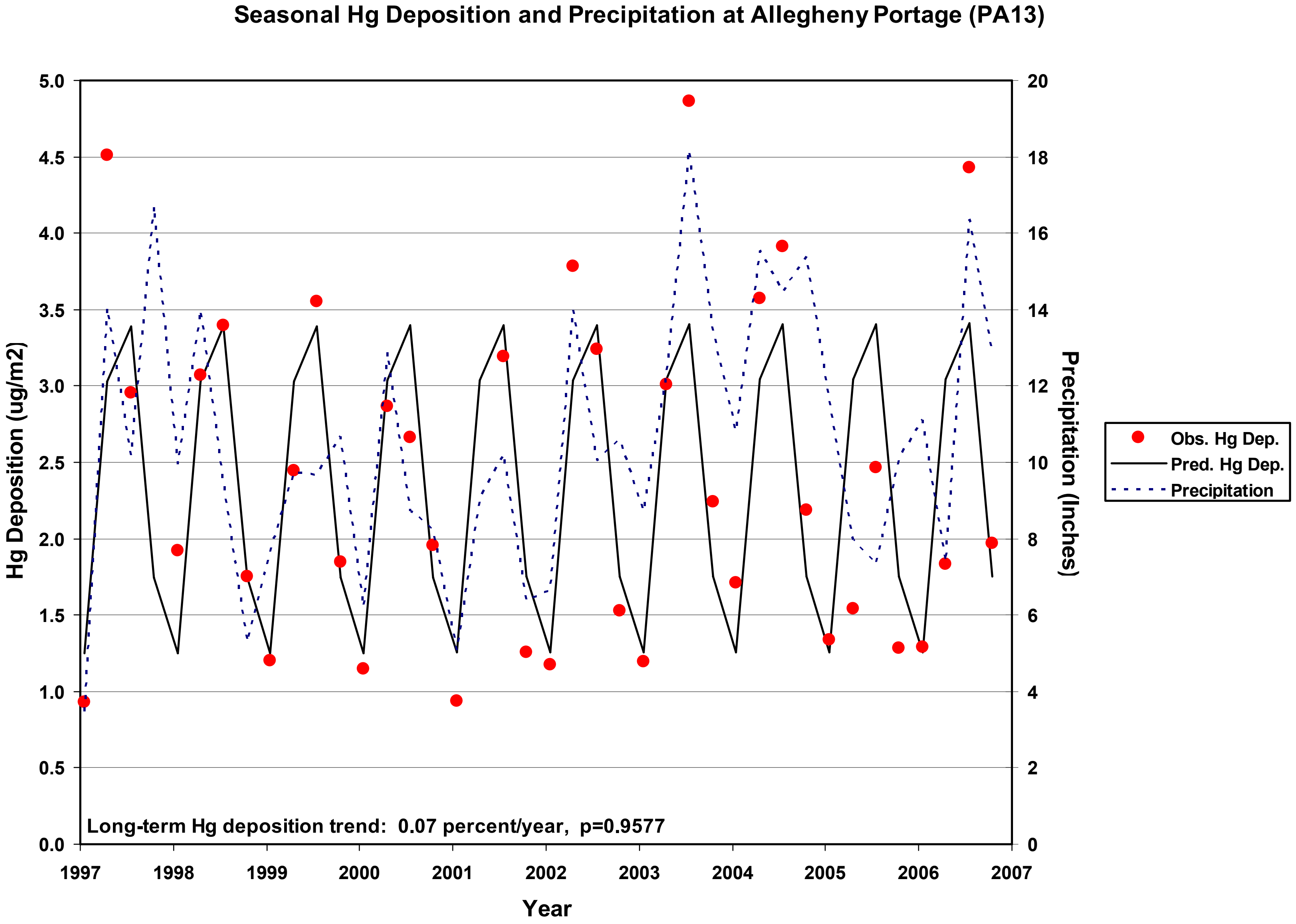


Figure 20. Seasonal trends in total mercury wet deposition and precipitation at the Allegheny Portage MDN site in Cambria County Pennsylvania from 1997 through 2006.

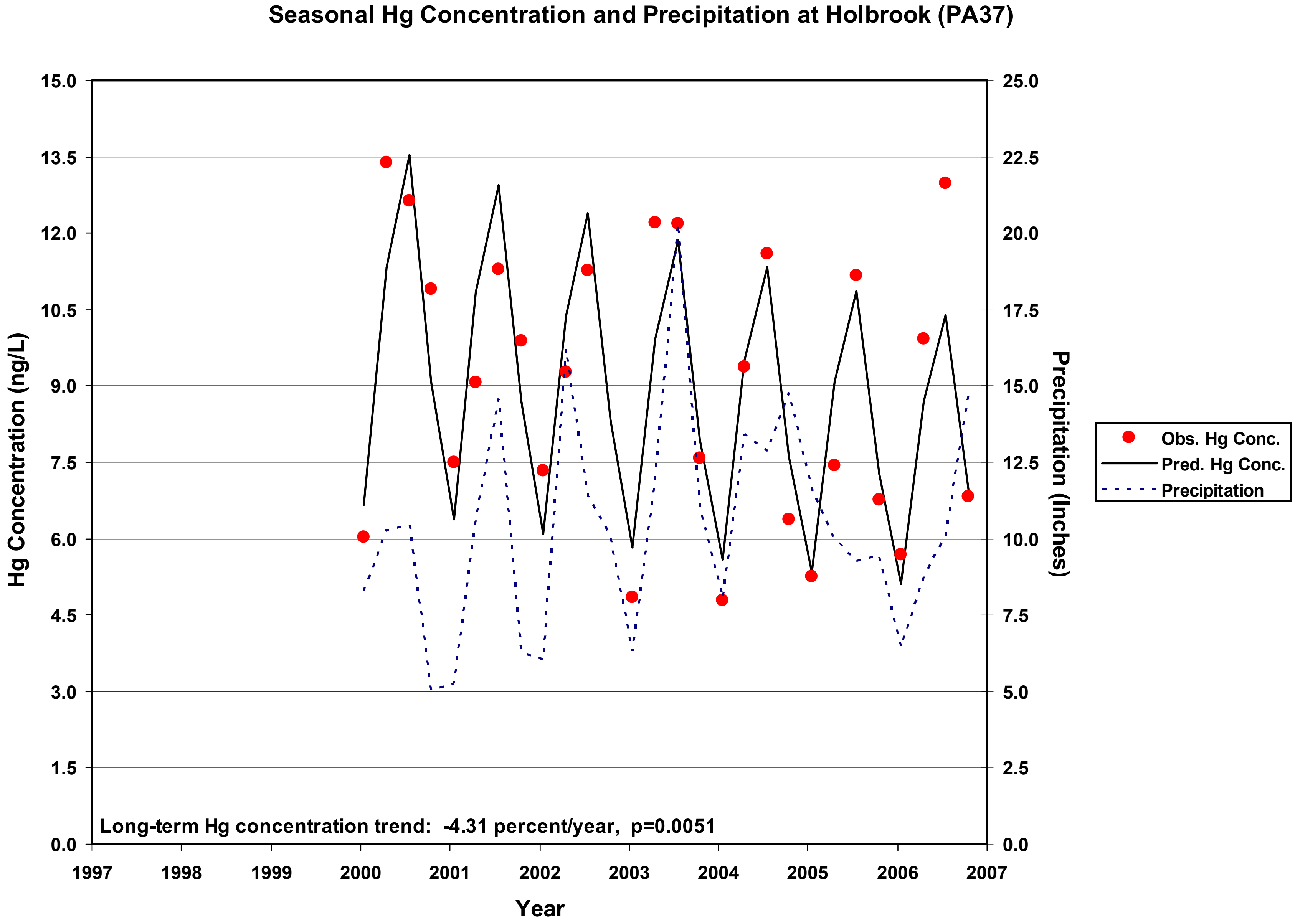


Figure 21. Seasonal trends in total mercury concentrations and precipitation at the Holbrook MDN site in Greene County Pennsylvania from 2000 through 2006.

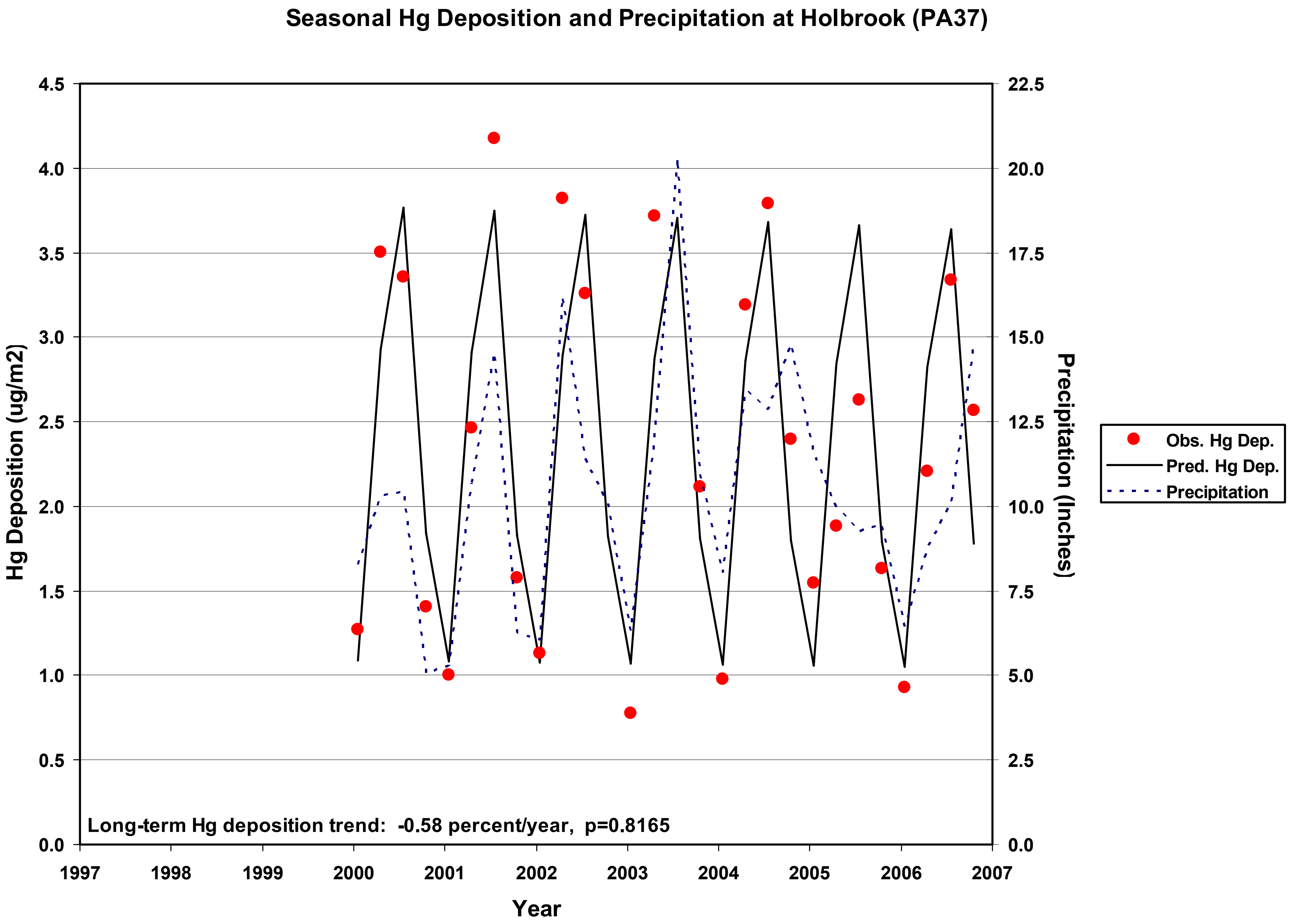


Figure 22. Seasonal trends in total mercury wet depositions and precipitation at the Holbrook MDN site in Greene County Pennsylvania from 2000 through 2006.

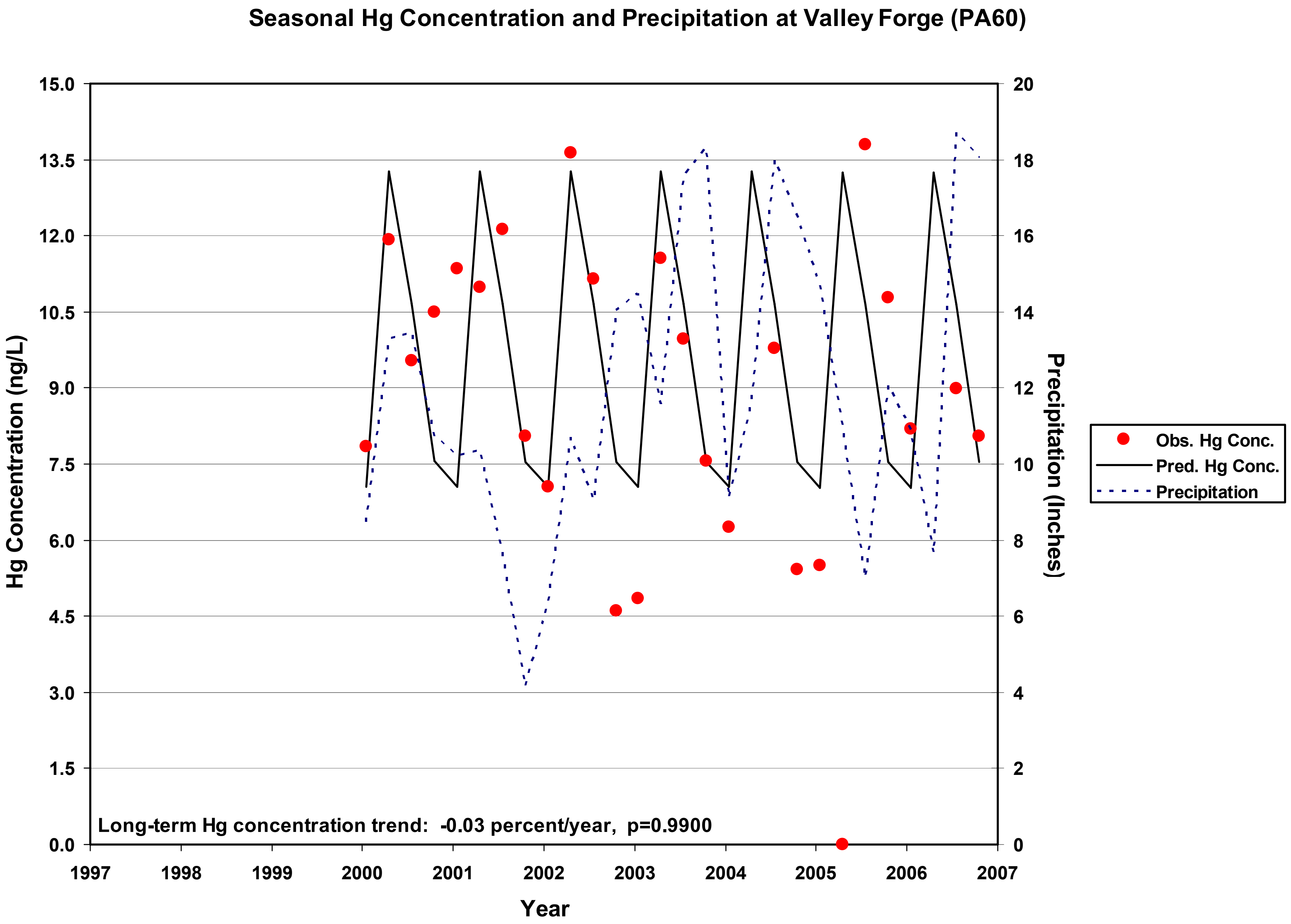


Figure 23. Seasonal trends in total mercury concentrations and precipitation at the Valley Forge MDN site in Montgomery County Pennsylvania from 2000 through 2006.

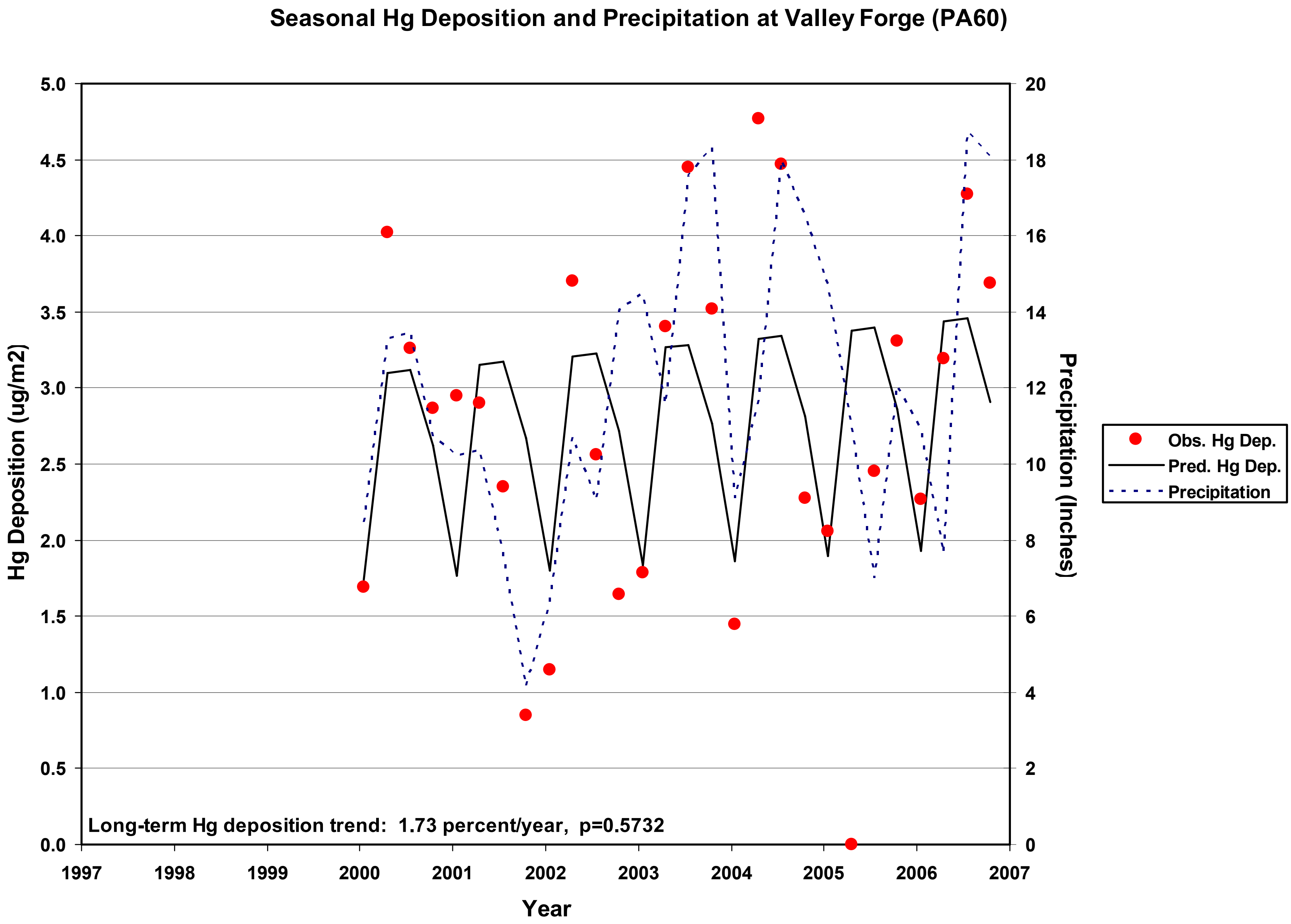


Figure 24. Seasonal trends in total mercury wet deposition and precipitation at the Valley Forge MDN site in Montgomery County Pennsylvania from 2000 through 2006.

The statistically significant decreasing total mercury concentration trends at Hills Creek (PA90), Allegheny-Portage NHS (PA13), and Holbrook (PA37) are similar to observed decreasing seasonal sulfate concentration trends at these sites (Figures 25-27), although the seasonal sulfate concentrations trends are not statistically significant ($p<0.05$). Given the close association between sulfate and mercury concentrations at these sites (Table 6 and 7), it is reasonable to assume that some of the decline total mercury concentrations would be related to reductions in sulfate concentrations that are in turn directly related to decreasing sulfur dioxide emissions in the eastern half of the United States following implementation in 1995 of Title IV of the Clean Air Act Amendments of 1990 (CAAA). Sulfur emissions reductions were achieved through a combination of scrubber technology, fuel switching (e.g., oil or natural gas for coal), the use of lower sulfur coal for Mid-western sources, and/or emissions trading. It is possible that this combination of approaches to reduced sulfur dioxide emissions has resulted in a concurrent reduction in mercury emissions as well. Given the location of the Hills Creek site and the lack of nearby mercury and sulfur emissions sources, it is reasonable to assume that long range transport of pollutants from upwind sources are the likely sources that were affected by the CAAA emissions reductions and are thus likely the sources also contributing to the reductions in mercury concentrations and deposition at his site. This would not necessarily be the case at either the Allegheny-Portage or Holbrook sites, because of their relative close proximity to major point sources. In addition, the stepwise regression analysis at each site with the possible exception of PA13 (Table 7) suggests multiple mercury emissions sources which may account for the lack of concurrent statistically significant trends in both mercury and sulfate concentrations at these sites. As stated earlier, sources of mercury emissions to the atmosphere in the United States can be broadly classified as natural mercury emissions, anthropogenic mercury emissions and re-emitted mercury. Re-emitted mercury is mercury that was previously deposited on the Earth's surface following either anthropogenic or natural releases and is re-emitted to the atmosphere by natural, biologic or geologic processes. Multiple sources of mercury at the Valley Forge site (Figure 28) where sulfate concentrations are decreasing ($p=0.023$) would also help to explain the lack of a concurrent decrease in mercury concentrations at this site.

Summary

Annual total mercury wet deposition at all MDN sites in the United States and southern Canada that met data completeness criteria for 2005 and 2006 ranged from 2.5 $\mu g/m^2$ in New Mexico to 21.5 $\mu g/m^2$ in west central Florida in 2005 (Figure 5) and from 2.1 $\mu g/m^2$ in Nevada to 18.8 $\mu g/m^2$ in southern Florida (Figure 7) in 2006. Wet mercury deposition in Pennsylvania in 2005 ranged from 6.7 $\mu g/m^2$ in Tioga and Cambria counties to 9.9 $\mu g/m^2$ in Montgomery County (Table 5). Wet mercury deposition in 2006 was lowest (8.1 $\mu g/m^2$) in Adams County and highest (10.1$\mu g/m^2$) in Pike County (Table 5). Because the Montgomery County site did not meet MDN data completeness criteria because of a large number of invalid samples it was not included in this comparison despite the fact that mercury deposition at the site measured 13.4 $\mu g/m^2$ in 2006. Volume-weighted mean annual concentrations of total mercury in precipitation in the United States ranged from 3.5 ng/L (southeast Canada) to 17.5 ng/L (New Mexico) in 2005 and from 3.6 ng/L (Oregon) to 20.7 ng/L (New Mexico) in 2006 (Figures 5 and 7). The volume-weighted

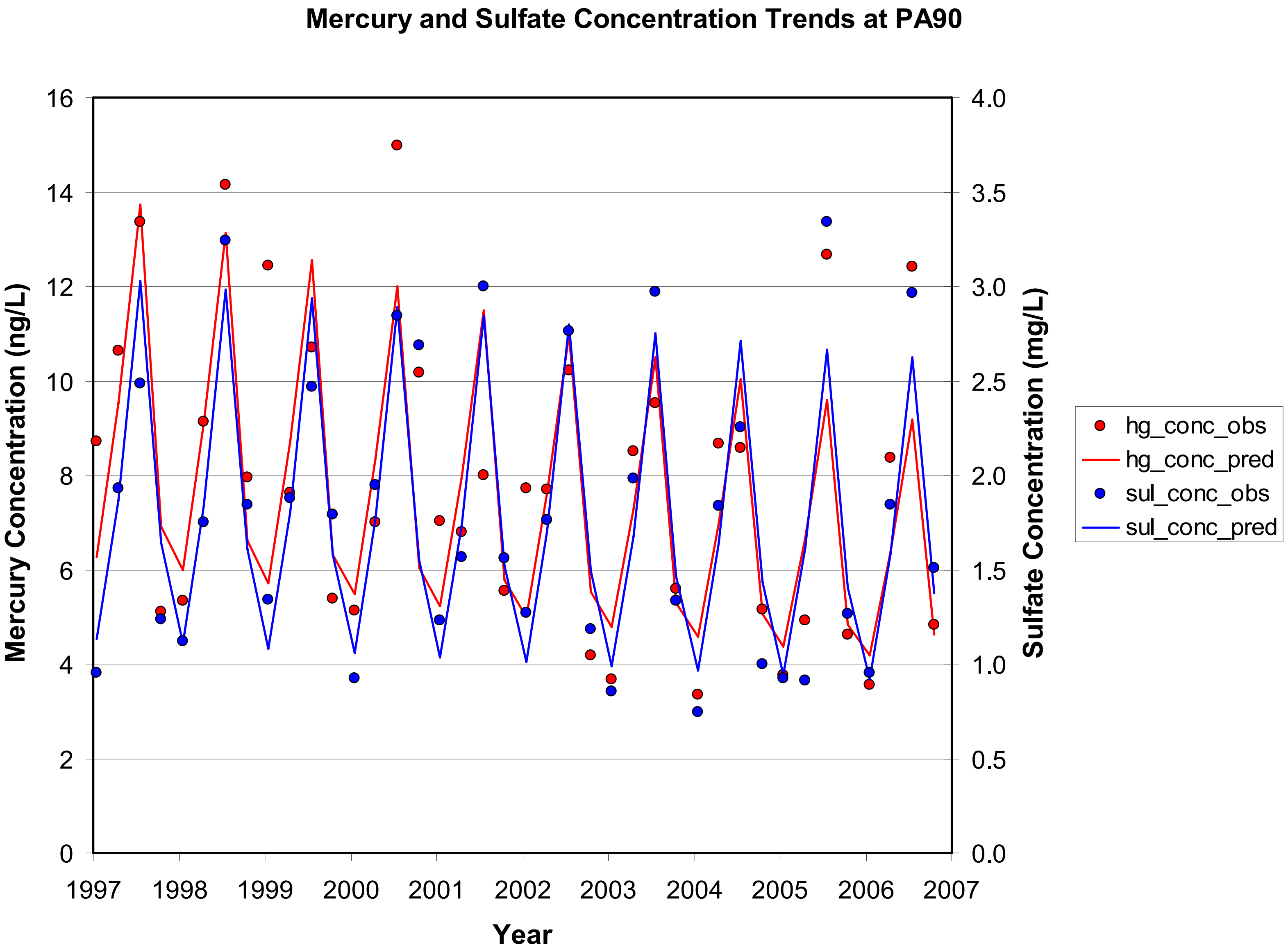


Figure 25. Trends in total mercury and sulfate concentrations at the Hills Creek MDN site (PA90) from 1997 through 2006

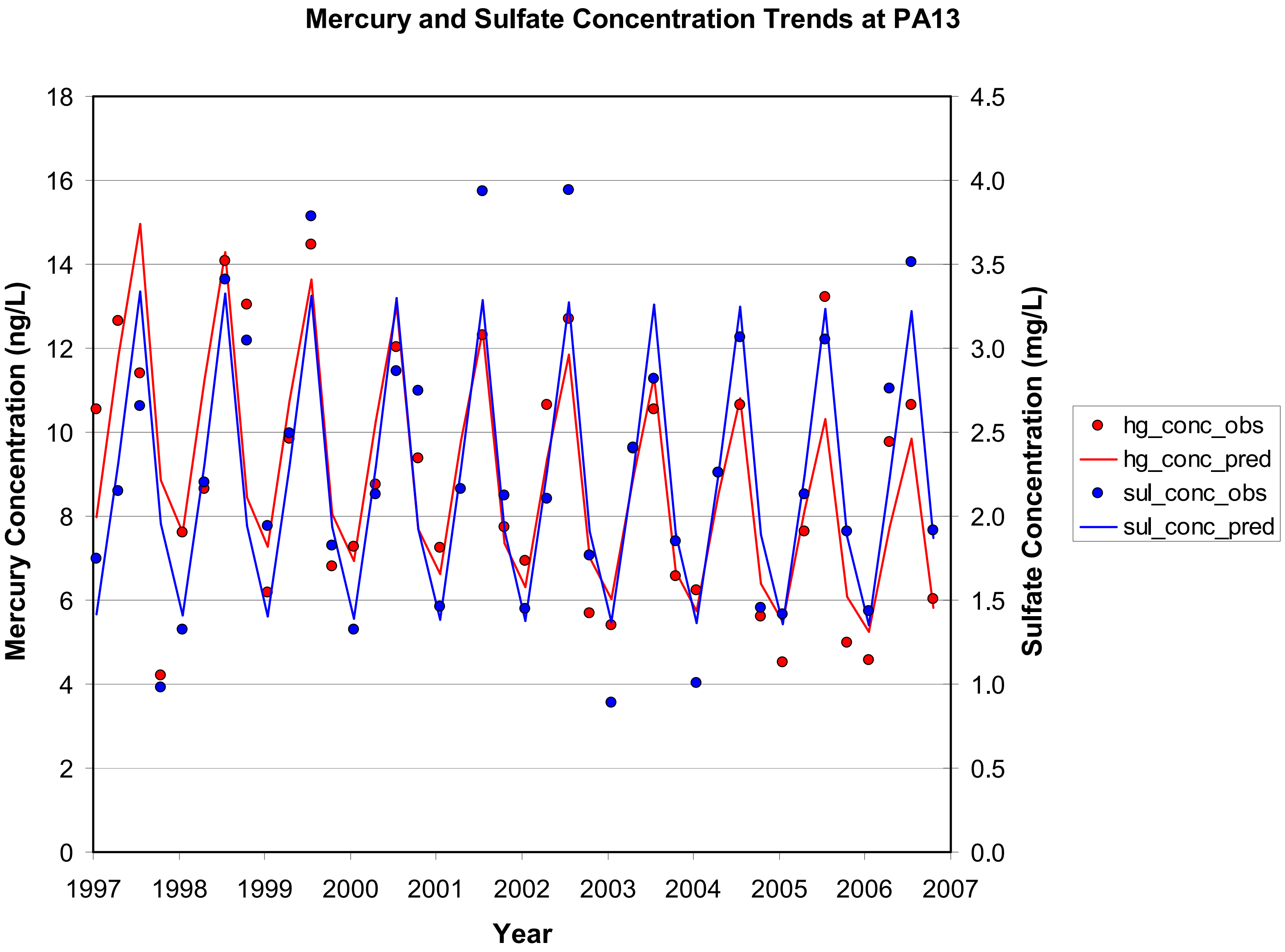


Figure 26. Trends in total mercury and sulfate concentrations at the Allegheny-Portange NHS MDN site (PA13) from 1997 through 2006.

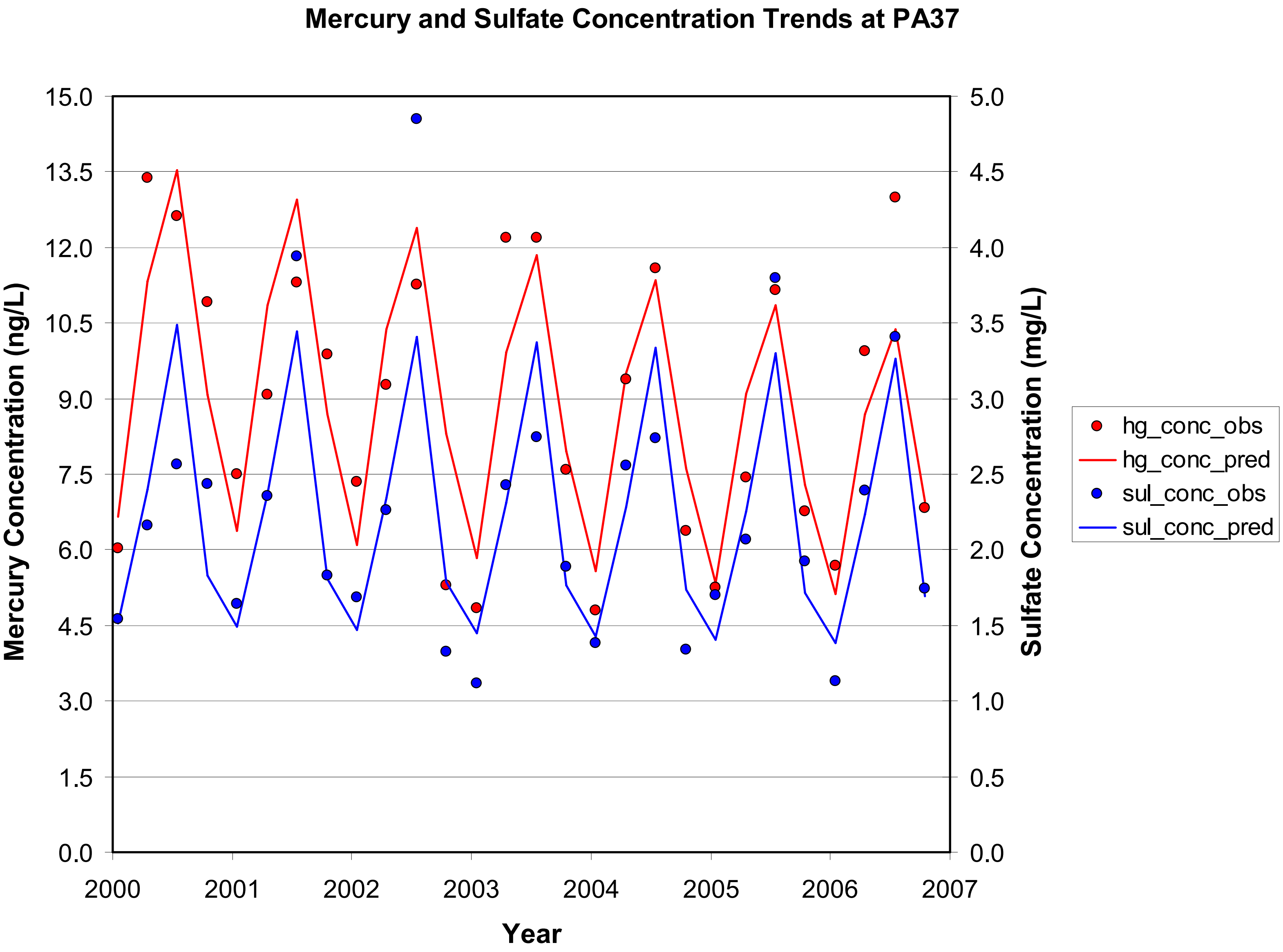


Figure 27. Trends in total mercury and sulfate concentrations at the Holbrook MDN site (PA37) from 2000 through 2006.

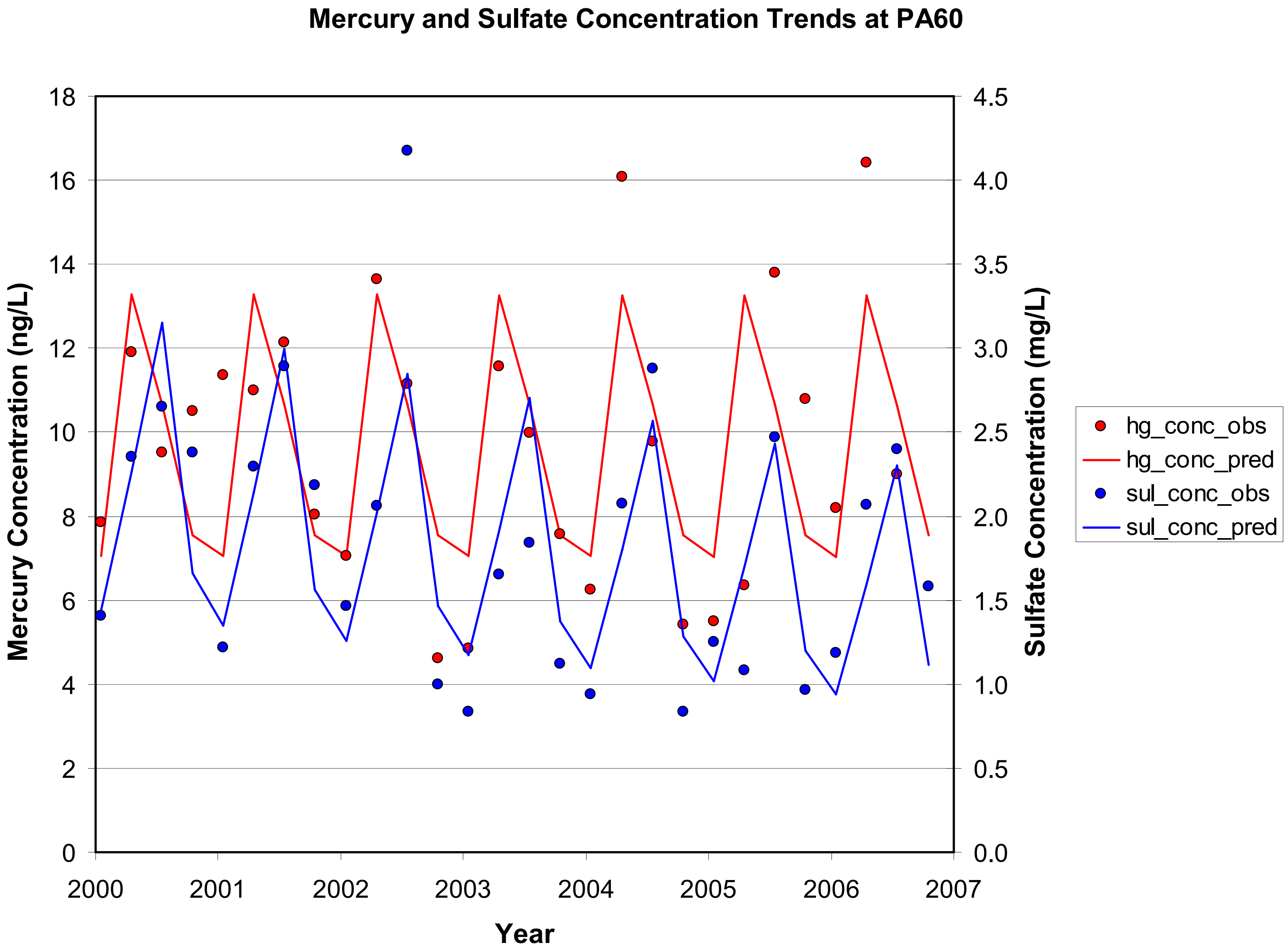


Figure 28. Trends in total mercury and sulfate concentrations at the Valley Forge MDN site (PA60) from 200 through 2006.