MODULE 2 LAKES

2.1

MERCURY DEPOSITION NETWORK

MERCURY DEPOSITION NETWORK

Atmospheric deposition is thought to be a significant source of mercury to Maine surface waters. In order to determine the relative significance of sources throughout Maine and the Northeast region, Maine has joined the Mercury Deposition Network (MDN). The MDN was created as an adjunct to the National Atmospheric Deposition Program (NADP), that has been monitoring the effects of atmospheric deposition of other contaminants, including acid rain, across the US for over 10 years. Maine has 4 NADP stations, one each at Bridgton, Acadia National Park (ANP), Greenville, and Caribou.

The MDN measures mercury in wet deposition on a weekly basis and provides a measurement of annual deposition at each station. All stations use similar equipment, the same protocol, and all samples will be analyzed by the same lab. There is also a Northeast regional network of MDN and other types of stations that measures wet deposition, as well as dry and gaseous mercury in some locations, in the New England states and the Canadian Maritime provinces.

One goal of MDN is to continue monitoring for at least 5 years. In Maine there are currently MDN stations at Acadia National Park (ANP, since fall 1995), Bridgton (since July 1997), Greenville (since September 1996), and Freeport (since 1998). The ANP station was supported equally by the National Park Service (NPS) and DEP through SWAT ($6000). The Greenville station was funded entirely by SWAT ($16500). The Bridgton station was funded primarily by an EPA REMAP grant, with DEP providing the station operator and mailing of the samples ($3150 SWAT). The Freeport station was supported entirely by a grant from EPA.

Annual deposition is greatest for the coastal stations, Freeport and Acacia National Park, followed by Bridgton and Greenville. Mean volume weighted concentration generally follows the same pattern. Ratios of annual deposition to mean concentration show that higher deposition along the coast is not entirely due to higher concentrations, but also due to increased precipitation.

TABLE 2.1 MERCURY IN WET DEPOSITION AT MAINE MDN STATIONS

ANNUAL DEPOSITION (ug/m2)

STATION	ID	1995	1996	1997	1998	1999	2000
Bridgton	ME02			5.7e	6.9	6.9	6.9
Greenville	ME09		5.5e	5.4	6.7	6.9	5.2
Freeport	ME96				12.0e	8.4	7.9
ANP	ME98	5.2e	7.8	7.7	9.0	8.0	8.7

e= estimated, site started during year

MEAN CONCENTRATION (ng/l)

STATION	ID		1996	1997	1998	1999	2000
Bridgton	ME02			8.4e	6.6	6.3	6.4
Greenville	ME09		4.0e	5.9	5.9	5.5	5.1
Freeport	ME96				7.8	7.3	6.6
ANP	ME98	5.2e	6.0	6.8	6.1	6.1	7.0

e=estimated since station began during the year

Mercury Deposition Network: a NADP Network

MDN Objectives

The objective of the MDN is to develop a national database of weekly concentrations of total mercury in precipitation and the seasonal and annual flux of total mercury in wet deposition. The data will be used to develop information on spatial and seasonal trends in mercury deposited to surface waters, forested watersheds, and other sensitive receptors.
Analysis of precipitation samples for total- and methylmercury is performed by Frontier Geosciences, Inc., Seattle WA, USA. Frontier Geosciences provides the environmental sciences community with uncompromisingly high-quality contract research, project design and management, and analytical chemistry services concerned with the sources, fate and effects of trace metals.
The MDN began a transition network of 13 sites in 1995. Beginning in 1996, MDN became an official network in NADP with 26 sites in operation. Over 50 sites were in operation during 2000 (see site map). The MDN is anticipated to operate for a minimum of five years and will be managed at the NADP Coordination Office. The network uses standardized methods for collection and analyses. **Weekly** precipitation samples are collected in a modified Aerochem Metrics model 301 collector. The "wet-side" sampling glassware is removed from the collector every Tuesday and mailed to the **Hg Analytical Laboratory (HAL)** at Frontier Geosciences in Seattle, WA for analysis by cold vapor atomic fluorescence. The MDN provides data for total mercury, but also includes methylmercury if desired by a site sponsor. Data are available via this Web page for the transition network (1995) and for 1996 through the second quarter of 2000.
The following journal articles and presentations describe the network design, including the sampling and analytical protocols, used in the MDN:

Lindberg, S. and Vermette, S. 1995. Workshop on Sampling Mercury in Precipitation for the National Atmospheric Deposition Program. Atmospheric Environment. 29, 1219-1220.
Vermette, S., Lindberg, S., and Bloom, N. 1995. Field Tests for a Regional Mercury Deposition Network - Sampling Design and Preliminary Test Results. Atmospheric Environment. 29, 1247-1251.

Welker, M. and Vermette, S.J., 1996. Mercury Deposition Network: QA/QC Protocols. Paper 96-RP129.01, Proceedings of the 89th Annual Meeting of the Air and Waste Management Association, A&WMA, Pittsburgh, PA.
Sweet, C.W. and Prestbo, E. 1999. Wet Deposition of Mercury in the U.S. and Canada. Presented at "Mercury in the Environment Specialty Conference", September 15-17, 1999, Minneapolis, MN. Proceedings published by Air and Waste Management Association, Pittsburgh, PA.
(Available from NADP Program Office)

Image credit: Mackerel On Mercury by Scot F. Hacker , 1995.

MDN DATA FIELDS
SITE CODE: 2-letter state or province designator plus SAROAD county code (US) or sequential number (Canada).
START DATE: (mm/dd/yyyy)
END DATE: (mm/dd/yyyy)
SUBPPT: Rain Gauge (RG) precipitation amount in mm if available, otherwise precipitation amount in mm is calculated from the net rain volume caught in the sample bottle.
PPT: Precipitation amount in mm from the rain gauge (RG), if blank, no RG data.
HG CONC: total mercury concentration reported by the lab in ng/L.
DEPOSITION: product of SUBPPT and HG CONC, units are ng/m2.
Quality rating (QR) CODE: A = fully qualified with no problems
B = valid data with minor problems, used for summary statistics
C = invalid data, not used for summary statistics
BLANK= no sample submitted for this time period
SAMPLE TYPE:
W = wet sample, measurable precipitation (> or = 0.03 in.) on the rain gauge (RG) or net bottle catch (BC) = or > 10.0 mL if RG data are missing. Concentration and deposition data are reported unless the QR Code = C.
D = dry sample, no indication of sampler openings on the RG or net BC < 1.5 mL if RG event recorder data are missing. No concentration data are reported. ppt, subppt, and deposition are set to zero.
T = trace sample, RG shows openings or a trace precipitation amount (<0.03 inches). If the RG data are missing, a net BC between 1.5 and 10.0 mL (inclusive) will be coded as a T sample type. Concentration data may or may not be reported depending whether the BC is 1.5 mL or higher. If BC = 1.5 mL or higher, then ppt is blank , Subppt = BC, and deposition is based on the BC. If BC < 1.5 mL, then ppt subppt and deposition are all set to zero.
Q = sampler was used for a Quality assurance (QA) sample, no ambient sample submitted. No concentration values are reported (QA values will be published in the QA report). Deposition is only reported where the value is zero (D or T samples with no measurable precipitation).

NOTES:	QR CODE	Valid for Summaries (Y/N)
s = short sample time (< 6days)	B	Y
e = extended sample time (> 8days)	B	Y
d = debris present (previously x)	B	Y

m = missing information (previously, r, no event recorder, and p, missing RG precipitation record)	B	Y
z = site operations problems	B	Y
h = sample handling problems (z and h include equipment and handling problems that don’t seriously compromise the sample)	B	Y
i = low volume sample (1.49mL < net BC < 10.00mL) (Hg conc. data are reported but they are less certain than those for samples with a net BC of at least 10 mL)	B	Y
b = bulk sample (wet side open the whole time)	C	N
v = RG indicates precipitation occurred but BC < 1 mL or < 10% of indicated RG precipitation amount.	C	N
u = undefined sample (wet side open during dry periods)	C	N
f = serious problems in field operations that compromise sample integrity.	C	N
l = laboratory error	C	N
c = sample compromised due to contamination	C	N
p = no ppt data from either RG or BC	C	N
n = no sample submitted	--	N

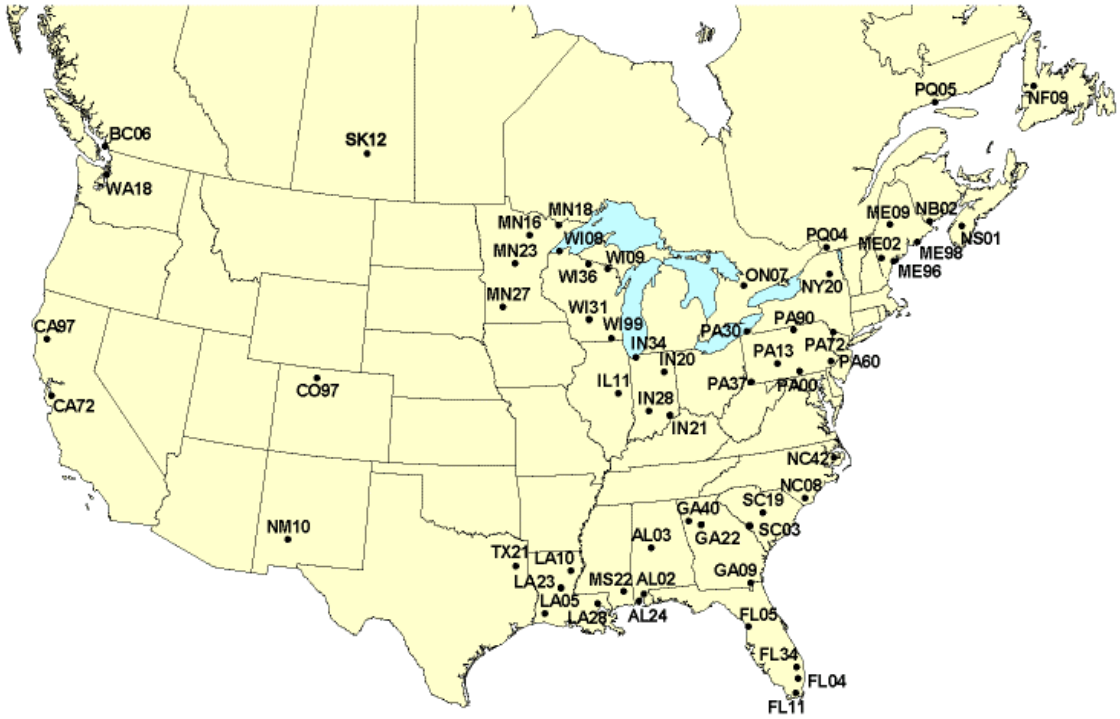
Calculation of Deposition:

1. If a valid precipitation amount can be read from the rain gauge chart (RG >= 0.03 inches), the sample type is set to “W” (wet); and the value from the RG chart is used to calculate deposition (RG amount in mm times Hg concentration in ng/mL). If the RG chart event recorder shows no sampler openings, sample type is set to “D” (dry) and precipitation amount and deposition are set to 0.
2. If the precipitation amount from the RG chart is not available, the net bottle catch (BC) will be used to calculate deposition as long as BC > 1.49mL. If the BC < 1.5 mL, the precipitation amount will be set to 0 and the sample type set to “D” (dry). If the BC is between 1.5 and 10.0 mL, the sample type will be set to “T” (trace) and the BC used to calculate deposition. These samples are also coded with an “i” in the Notes field and downgraded to a “B” Quality Rating to indicate uncertainty due to low volume. If the BC is > 10 mL, the sample type will be set

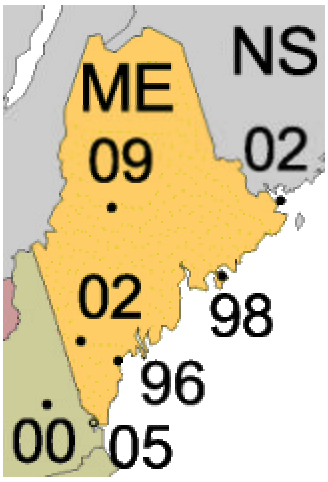
to “W” (wet) and the BC will be used to calculate deposition.
3. If the RG indicates sampler openings, but the precipitation amount can’t be determined accurately from the RG chart (RG < 0.03 inches) the sample type will be coded “T” (trace) and the BC will be used to calculate deposition as long as the BC is >= 1.5mL. If the BC is < 10mL, samples will be coded for low volume as in 2. If the BC is < 1.5mL, no concentration will be reported and the ppt, subppt, and deposition will be set to 0.
4. In cases where there is a valid precipitation amount from either RG or BC but invalid or missing concentration data, seasonal or annual summary deposition values will be calculated using the site-specific, seasonal, volume-weighted average concentration. This deposition value will not be displayed for individual weeks in the WEB database, but it will be used only for the calculation seasonal and annual average concentrations and deposition amounts on maps and other summary products.

MDN STATIONS

National Atmospheric Deposition Program
Mercury Deposition Network



Mercury Deposition Network Maine stations



Site ID	Site Name	Start Date	End Date	Elevation (meters)
Active Sites				
ME02	Bridgton	06/04/1997		222
ME09	Greenville Station	09/03/1996		322
ME96	Freeport	01/01/1998		15
ME98	Acadia National Park - McFarland Hill	09/26/1995		129
Inactive Sites				

National Atmospheric Deposition Program/MDN

Weekly Mercury Concentrations and Depositions

BRIDGTON ME02

Site	Date On	Date Off	Sub ppt	Pptrec	HgConc	HgDep	QR	Sample Type	Notes
			mm	mm	ng/L	ng/m²			
ME02	12/28/1999	01/04/2000	12.7	12.7	12.1	153.7	B	W	d
ME02	01/04/2000	01/11/2000	40.5	40.5	4.6	188.2	B	W	d
ME02	01/11/2000	01/18/2000	10.2	10.2	6.2	62.9	B	W	d
ME02	01/18/2000	01/25/2000	0.0	0.0	--	0.0	A	D	
ME02	01/25/2000	02/01/2000	22.9	22.9	2.8	63.2	B	W	dh
ME02	02/01/2000	02/08/2000	0.0	0.0	--	0.0	B	D	d
ME02	02/08/2000	02/15/2000	34.7	34.7	4.1	143.0	B	W	d
ME02	02/15/2000	02/22/2000	12.1	12.1	3.2	38.3	B	W	d
ME02	02/22/2000	02/29/2000	14.4	14.4	--	--	C	W	fv
ME02	02/29/2000	03/07/2000	3.0	3.0	12.8	39.1	B	W	d
ME02	03/07/2000	03/14/2000	29.5	29.5	5.2	154.5	B	W	d
ME02	03/14/2000	03/21/2000	11.4	11.4	4.4	50.1	B	W	d
ME02	03/21/2000	03/28/2000	45.7	45.7	4.7	216.6	B	W	dh
ME02	03/28/2000	04/04/2000	45.4	45.4	7.7	347.8	B	W	d
ME02	04/04/2000	04/11/2000	31.2	31.2	5.8	181.2	B	W	d
ME02	04/11/2000	04/18/2000	6.2	6.2	5.1	31.7	B	W	d
ME02	04/18/2000	04/25/2000	79.1	79.1	3.1	248.2	B	W	d
ME02	04/25/2000	05/02/2000	1.3	1.3	16.1	20.4	B	W	di
ME02	05/02/2000	05/09/2000	12.2	12.2	15.7	191.6	B	W	dh
ME02	05/09/2000	05/16/2000	30.4	30.4	9.6	292.4	B	W	d
ME02	05/16/2000	05/23/2000	10.2	10.2	7.5	76.9	A	W	
ME02	05/23/2000	05/30/2000	31.0	31.0	5.2	159.9	B	W	h
ME02	05/30/2000	06/06/2000	15.4	15.4	--	--	C	W	uz
ME02	06/06/2000	06/13/2000	22.2	22.2	10.0	222.0	B	W	d
ME02	06/13/2000	06/20/2000	8.0	8.0	13.2	105.4	B	W	d
ME02	06/20/2000	06/27/2000	18.8	18.8	5.5	103.1	B	W	h
ME02	06/27/2000	07/04/2000	44.5	44.5	--	--	C	W	ufd
ME02	07/04/2000	07/11/2000	26.4	26.4	--	--	C	W	bd
ME02	07/11/2000	07/18/2000	39.0	39.0	--	--	C	W	udf
ME02	07/18/2000	07/25/2000	2.0	2.0	26.0	52.8	A	W	
ME02	07/25/2000	08/01/2000	37.5	37.5	11.3	421.9	B	W	d
ME02	08/01/2000	08/08/2000	4.6	4.6	11.1	50.9	B	W	m
ME02	08/08/2000	08/15/2000	34.7	34.7	16.9	585.9	B	W	dm
ME02	08/15/2000	08/22/2000	3.9	3.9	12.4	48.8	B	W	dm

ME02	08/22/2000	08/29/2000	4.1	4.1	11.1	45.0	B	W	m
ME02	08/29/2000	09/05/2000	4.8	4.8	10.0	48.4	B	W	dm
ME02	09/05/2000	09/12/2000	0.0	0.0	--	0.0	B	D	m
ME02	09/12/2000	09/19/2000	21.6	21.6	--	--	C	W	fm
ME02	09/19/2000	09/26/2000	5.7	5.7	5.4	30.7	B	W	m
ME02	09/26/2000	10/03/2000	0.0	0.0	--	0.0	A	D	
ME02	10/03/2000	10/10/2000	32.7	32.7	4.9	161.8	B	W	dm
ME02	10/10/2000	10/17/2000	6.6	6.6	3.2	21.3	B	W	m
ME02	10/17/2000	10/24/2000	37.8	37.8	3.1	115.5	B	W	m
ME02	10/24/2000	10/31/2000	10.7	10.7	1.9	20.5	B	W	hm
ME02	10/31/2000	11/07/2000	16.1	16.1	7.9	127.8	B	W	dm
ME02	11/07/2000	11/14/2000	40.5	40.5	4.1	164.3	B	W	dm
ME02	11/14/2000	11/21/2000	27.6	27.6	2.0	55.4	B	W	dm
ME02	11/21/2000	11/28/2000	22.8	22.8	2.1	48.6	B	W	m
ME02	11/28/2000	12/05/2000	0.5	0.5	2.2	1.1	B	T	mi
ME02	12/05/2000	12/12/2000	15.0	15.0	0.0	0.0	B	W	dm
ME02	12/12/2000	12/19/2000	72.1	72.1	4.9	354.7	B	W	dm
ME02	12/19/2000	12/26/2000	4.2	--	3.2	13.3	B	W	m
ME02	12/26/2000	01/02/2001	0.0	0.0	4.5	0.0	B	W	m

National Atmospheric Deposition Program/MDN

Weekly Mercury Concentrations and Depositions

GREENVILLE MEO9

Site	Date On	Date Off	Sub ppt	Pptrec	HgConc	HgDep	QR	Sample Type	Notes
			mm	mm	ng/L	ng/m²			
ME09	12/28/1999	01/04/2000	6.6	6.6	--	--	C	W	fd
ME09	01/04/2000	01/11/2000	30.9	30.9	--	--	C	W	fd
ME09	01/11/2000	01/18/2000	14.6	14.6	--	--	C	W	fdv
ME09	01/18/2000	01/25/2000	--	--	--	--	C	W	fm
ME09	01/25/2000	02/01/2000	34.3	34.3	--	--	C	W	fdm
ME09	02/01/2000	02/08/2000	0.0	--	--	0.0	B	T	m
ME09	02/08/2000	02/15/2000	38.9	38.9	1.7	65.4	B	W	dh
ME09	02/15/2000	02/22/2000	5.1	5.1	2.7	13.6	B	W	d
ME09	02/22/2000	02/29/2000	19.8	19.8	2.8	55.5	B	W	dm
ME09	02/29/2000	03/07/2000	13.1	13.1	3.7	48.9	B	W	d
ME09	03/07/2000	03/14/2000	31.8	31.8	3.7	118.3	B	W	d
ME09	03/14/2000	03/21/2000	8.3	8.3	2.8	23.5	B	W	h
ME09	03/21/2000	03/28/2000	0.6	0.6	17.9	11.4	B	T	i
ME09	03/28/2000	04/04/2000	56.4	56.4	3.0	171.2	B	W	d
ME09	04/04/2000	04/11/2000	82.2	82.2	4.4	361.5	B	W	d
ME09	04/11/2000	04/18/2000	9.0	9.0	2.7	24.4	A	W	
ME09	04/18/2000	04/25/2000	78.0	78.0	2.1	166.8	B	W	d
ME09	04/25/2000	05/02/2000	2.4	2.4	14.5	35.1	B	W	dh
ME09	05/02/2000	05/09/2000	11.4	11.4	--	--	C	W	cm
ME09	05/09/2000	05/16/2000	57.2	57.2	8.6	491.7	B	W	d
ME09	05/16/2000	05/23/2000	13.0	13.0	7.0	90.9	B	W	d
ME09	05/23/2000	05/30/2000	6.7	6.7	15.3	103.2	B	W	d
ME09	05/30/2000	06/06/2000	4.1	4.1	9.4	38.1	B	W	d
ME09	06/06/2000	06/13/2000	24.5	24.5	9.6	234.4	B	W	dh
ME09	06/13/2000	06/20/2000	0.8	0.8	18.2	13.8	B	W	di
ME09	06/20/2000	06/27/2000	6.4	6.4	6.9	43.8	B	W	d
ME09	06/27/2000	07/04/2000	51.4	51.4	8.4	433.8	B	W	h
ME09	07/04/2000	07/11/2000	12.1	12.1	15.1	182.6	B	W	d
ME09	07/11/2000	07/18/2000	10.4	10.4	13.5	140.4	B	W	d
ME09	07/18/2000	07/25/2000	21.6	21.6	11.4	245.4	B	W	d
ME09	07/25/2000	08/01/2000	0.0	0.0	--	0.0	A	D	
ME09	08/01/2000	08/08/2000	0.2	--	48.2	7.2	B	T	i
ME09	08/08/2000	08/15/2000	7.7	7.7	7.4	56.7	B	W	d
ME09	08/15/2000	08/22/2000	19.9	--	7.8	154.5	B	W	m

ME09	08/22/2000	08/29/2000	13.3	13.3	8.7	116.6	B	W	d
ME09	08/29/2000	09/05/2000	32.3	32.3	7.0	224.2	B	W	d
ME09	09/05/2000	09/12/2000	0.0	0.0	--	0.0	B	T	d
ME09	09/12/2000	09/19/2000	14.6	14.6	5.8	84.9	B	W	hd
ME09	09/19/2000	09/26/2000	7.1	7.1	7.9	56.1	B	W	hd
ME09	09/26/2000	10/02/2000	2.8	2.8	2.4	6.6	B	W	d
ME09	10/03/2000	10/10/2000	38.5	38.5	--	--	C	W	ufd
ME09	10/10/2000	10/17/2000	2.1	--	2.1	4.4	B	W	dm
ME09	10/17/2000	10/24/2000	13.8	13.8	1.9	26.3	B	W	d
ME09	10/24/2000	10/31/2000	29.0	29.0	2.5	72.5	B	W	d
ME09	10/31/2000	11/07/2000	5.1	5.1	0.7	3.8	B	W	d
ME09	11/07/2000	11/14/2000	1.7	1.7	6.1	10.0	B	W	di
ME09	11/14/2000	11/21/2000	38.4	38.4	1.3	49.2	B	W	hd
ME09	11/21/2000	11/28/2000	30.6	30.6	1.5	47.2	B	W	d
ME09	11/28/2000	12/05/2000	0.0	0.0	--	0.0	B	T	m
ME09	12/05/2000	12/12/2000	4.4	4.4	4.1	18.2	B	W	dm
ME09	12/12/2000	12/19/2000	84.2	84.2	7.1	597.6	B	W	d
ME09	12/19/2000	12/26/2000	18.8	18.8	1.2	22.7	B	W	d
ME09	12/26/2000	01/02/2001	19.3	19.3	2.1	41.0	A	W	

National Atmospheric Deposition Program/MDN

Weekly Mercury Concentrations and Depositions

FREEPORT ME96

Site	Date On	Date Off	Subppt	Pptrec	HgConc	HgDep	QR	Sample Type	Notes
			mm	Mm	ng/L	ng/m²			
ME96	12/28/1999	01/04/2000	3.8	3.8	15.4	58.5	B	W	d
ME96	01/04/2000	01/11/2000	47.5	47.5	4.5	214.4	B	W	d
ME96	01/11/2000	01/18/2000	12.4	12.4	5.2	64.0	B	W	d
ME96	01/18/2000	01/25/2000	0.0	0.0	--	0.0	A	D	
ME96	01/25/2000	02/01/2000	34.3	34.3	2.7	90.9	B	W	dh
ME96	02/01/2000	02/08/2000	0.0	0.0	--	0.0	B	D	dh
ME96	02/08/2000	02/15/2000	40.1	40.1	4.9	196.0	B	W	d
ME96	02/15/2000	02/22/2000	9.5	9.5	4.9	46.9	B	W	d
ME96	02/22/2000	02/29/2000	9.7	9.7	3.6	34.4	B	W	d
ME96	02/29/2000	03/07/2000	8.3	8.3	7.5	62.0	B	W	d
ME96	03/07/2000	03/14/2000	41.7	41.7	7.3	302.4	B	W	dh
ME96	03/14/2000	03/21/2000	13.7	13.7	4.6	63.1	B	W	d
ME96	03/21/2000	03/28/2000	9.4	9.4	8.2	76.9	B	W	dm
ME96	03/28/2000	04/04/2000	67.4	67.4	8.3	560.0	B	W	m
ME96	04/04/2000	04/11/2000	24.8	24.8	3.7	91.3	B	W	d
ME96	04/11/2000	04/18/2000	3.2	3.2	8.7	27.5	B	W	dh
ME96	04/18/2000	04/25/2000	99.7	99.7	2.5	247.1	B	W	d
ME96	04/25/2000	05/02/2000	3.9	3.9	15.7	61.9	B	W	d
ME96	05/02/2000	05/09/2000	10.8	--	21.4	229.6	B	W	m
ME96	05/09/2000	05/16/2000	32.8	32.8	10.2	334.9	B	W	d
ME96	05/16/2000	05/23/2000	8.4	8.4	14.3	119.6	A	W	
ME96	05/23/2000	05/30/2000	36.1	36.1	8.0	288.0	B	W	h
ME96	05/30/2000	06/06/2000	1.0	1.0	23.0	23.4	B	W	di
ME96	06/06/2000	06/13/2000	39.0	39.0	7.7	298.4	A	W	
ME96	06/13/2000	06/20/2000	17.2	17.2	7.5	129.9	B	W	d
ME96	06/20/2000	06/27/2000	16.8	16.8	7.7	129.4	B	W	d
ME96	06/27/2000	07/05/2000	37.0	37.0	14.5	538.6	A	W	
ME96	07/05/2000	07/11/2000	15.5	--	10.9	168.9	B	W	hm
ME96	07/11/2000	07/18/2000	49.4	49.4	3.4	165.6	B	W	d
ME96	07/18/2000	07/25/2000	17.1	17.1	22.0	376.3	B	W	d
ME96	07/25/2000	08/01/2000	15.9	15.9	7.3	115.6	A	W	
ME96	08/01/2000	08/08/2000	8.9	8.9	5.9	52.8	B	W	d
ME96	08/08/2000	08/15/2000	37.5	37.5	9.3	347.4	B	W	dh
ME96	08/15/2000	08/22/2000	1.6	--	19.7	31.6	B	W	dm
ME96	08/22/2000	08/29/2000	4.5	--	6.2	27.7	B	W	m

ME96	08/29/2000	09/05/2000	13.7	13.7	4.6	62.5	B	W	dh
ME96	09/05/2000	09/12/2000	0.0	0.0	--	0.0	A	D	
ME96	09/12/2000	09/19/2000	26.2	26.2	7.5	195.0	B	W	d
ME96	09/19/2000	09/26/2000	36.9	36.9	5.7	210.3	B	W	d
ME96	09/26/2000	10/03/2000	1.0	1.0	1.4	1.3	B	W	i
ME96	10/03/2000	10/10/2000	39.2	39.2	5.2	206.0	B	W	d
ME96	10/10/2000	10/17/2000	6.4	6.4	4.5	29.1	A	W	
ME96	10/17/2000	10/24/2000	30.6	30.6	4.1	126.0	B	W	d
ME96	10/24/2000	10/31/2000	24.8	24.8	1.2	28.8	A	W	
ME96	10/31/2000	11/07/2000	9.8	9.8	7.4	72.5	B	W	h
ME96	11/07/2000	11/14/2000	35.6	35.6	6.4	228.5	A	W	
ME96	11/14/2000	11/21/2000	37.5	37.5	3.1	115.0	B	W	d
ME96	11/21/2000	11/28/2000	42.7	42.7	1.8	75.7	B	W	h
ME96	11/28/2000	12/05/2000	0.0	0.0	--	0.0	A	T	
ME96	12/05/2000	12/12/2000	6.3	6.3	6.7	42.3	B	W	h
ME96	12/12/2000	12/19/2000	91.2	91.2	9.3	847.6	B	W	d
ME96	12/19/2000	12/26/2000	11.9	11.9	3.8	45.2	B	W	d
ME96	12/26/2000	01/02/2001	16.0	16.0	3.1	49.4	B	W	d

National Atmospheric Deposition Program/MDN

Weekly Mercury Concentrations and Depositions

ACADIA NATIONAL PARK ME98

Site	Date On	Date Off	Subppt	Pptrec	HgConc	HgDep	QR	Sample Type	Notes
			mm	Mm	ng/L	ng/m²			
ME98	12/28/1999	01/04/2000	14.2	14.2	20.2	286.6	B	W	dh
ME98	01/04/2000	01/11/2000	70.2	70.2	3.5	247.7	B	W	d
ME98	01/11/2000	01/18/2000	16.3	16.3	0.3	4.2	B	W	d
ME98	01/18/2000	01/25/2000	9.2	9.2	5.0	46.1	A	W	
ME98	01/25/2000	02/01/2000	22.2	22.2	5.9	131.1	B	W	hx
ME98	02/01/2000	02/08/2000	0.0	0.0	--	--	C	T	fd
ME98	02/08/2000	02/15/2000	59.9	59.9	8.1	486.1	B	W	dh
ME98	02/15/2000	02/22/2000	10.2	10.2	--	--	C	W	fvd
ME98	02/22/2000	02/29/2000	1.3	1.3	--	--	C	W	vm
ME98	02/29/2000	03/07/2000	19.1	19.1	2.7	51.0	B	W	dm
ME98	03/08/2000	03/15/2000	38.4	38.4	7.4	283.6	B	W	dh
ME98	03/14/2000	03/21/2000	10.7	10.7	--	--	C	W	vd
ME98	03/21/2000	03/28/2000	20.3	20.3	9.6	194.3	B	W	d
ME98	03/28/2000	04/04/2000	33.3	33.3	13.8	457.9	B	W	d
ME98	04/04/2000	04/11/2000	11.8	11.8	11.5	136.0	B	W	dh
ME98	04/11/2000	04/18/2000	8.8	8.8	7.3	64.4	B	W	d
ME98	04/18/2000	04/25/2000	177.7	177.7	5.1	913.0	A	W	
ME98	04/25/2000	05/02/2000	3.4	3.4	--	--	C	W	fvd
ME98	05/02/2000	05/09/2000	14.9	14.9	13.3	198.0	B	W	dh
ME98	05/09/2000	05/16/2000	49.4	49.4	7.6	376.7	A	W	
ME98	05/16/2000	05/23/2000	30.1	30.1	12.4	374.2	A	W	
ME98	05/23/2000	05/30/2000	31.6	31.6	6.0	189.0	B	W	h
ME98	05/31/2000	06/06/2000	0.0	0.0	--	0.0	A	T	
ME98	06/06/2000	06/13/2000	24.8	24.8	7.4	183.1	B	W	h
ME98	06/13/2000	06/20/2000	11.3	11.3	35.8	404.2	B	W	dh
ME98	06/20/2000	06/27/2000	11.4	11.4	9.2	105.7	B	W	h
ME98	06/27/2000	07/03/2000	3.6	3.6	52.4	186.3	B	W	d
ME98	07/03/2000	07/11/2000	26.0	26.0	15.5	404.3	B	W	d
ME98	07/11/2000	07/18/2000	59.7	59.7	5.8	343.2	B	W	dh
ME98	07/18/2000	07/25/2000	3.9	3.9	23.8	92.1	B	W	d
ME98	07/25/2000	08/01/2000	5.1	5.1	7.2	36.4	A	W	
ME98	08/01/2000	08/08/2000	5.1	5.1	9.4	47.9	B	W	d
ME98	08/08/2000	08/15/2000	4.2	4.2	19.2	80.3	B	W	h
ME98	08/15/2000	08/22/2000	9.0	9.0	11.8	106.6	B	W	h
ME98	08/22/2000	08/29/2000	7.0	7.0	3.5	24.5	A	W	

ME98	08/29/2000	09/05/2000	15.3	15.3	7.7	118.3	B	W	m
ME98	09/05/2000	09/12/2000	0.0	0.0	--	0.0	A	D	
ME98	09/12/2000	09/19/2000	34.2	34.2	6.5	221.3	B	W	h
ME98	09/19/2000	09/26/2000	20.8	20.8	9.0	186.6	B	W	m
ME98	09/26/2000	10/03/2000	0.0	0.0	--	0.0	A	D	
ME98	10/03/2000	10/10/2000	61.8	61.8	4.7	290.6	B	W	d
ME98	10/10/2000	10/17/2000	0.3	0.3	--	0.0	A	T	
ME98	10/17/2000	10/24/2000	21.7	21.7	4.6	100.6	A	W	
ME98	10/24/2000	10/31/2000	36.6	36.6	1.7	61.0	B	W	dh
ME98	10/31/2000	11/07/2000	31.3	31.3	2.9	90.2	B	W	h
ME98	11/07/2000	11/14/2000	1.0	1.0	6.8	6.9	B	W	hi
ME98	11/14/2000	11/21/2000	31.6	31.6	4.5	142.0	B	W	dh
ME98	11/21/2000	11/28/2000	62.7	62.7	1.7	104.0	B	W	d
ME98	11/28/2000	12/05/2000	2.7	2.7	9.3	24.7	A	W	
ME98	12/05/2000	12/12/2000	19.2	19.2	4.2	80.8	B	W	dh
ME98	12/12/2000	12/19/2000	64.7	64.7	6.8	440.1	B	W	h
ME98	12/19/2000	12/26/2000	27.7	27.7	9.8	271.7	B	W	dh
ME98	12/26/2000	01/02/2001	21.3	21.3	2.3	49.4	B	W	mh

Methyl Mercury in Precipitation at the Acadia National Park Station, Mercury Deposition Network

Terry A. Haines

Methyl mercury determination was added to the suite of analytes at the Acadia National Park station of the Mercury Deposition Network for the period July 2000 to June 2001. Samples were collected weekly, and there were 50 usable samples for total mercury and 47 for methyl mercury, however I dropped the methyl mercury data for July 1 as the results seemed unreasonably high (Table 1). For the weekly samples, methyl mercury concentration was about 0.9% of total mercury, 0.086 ng/L versus 9.29 ng/L. The annual volume-weighted mean concentrations were 6.32 ng/L for total mercury and 0.08 ng/L for methyl mercury, giving 1.27% methyl mercury. The Acadia MDN site has had annual volume-weighted mean total mercury concentration ranging from 6.0 to 6.8 ng/L during the period 1996-1999. Glass and Sorensen (1999) found a mean annual (volume-weighted) total mercury concentration in precipitation at six locations in the upper midwest during 1990-1995 to be 10.9 ng/L. The individual locations had mean total mercury concentrations ranging from 9.1 to 11.9 ng/L. Methyl mercury concentration, determined in only 36 weekly samples, averaged 0.18 ng/L.

Total mercury concentration was highest in rain samples, intermediate in snow, and lowest in mixed precipitation samples; however, methyl mercury concentration was highest in snow, intermediate in mixed, and lowest in rain samples (Table 1). In two small streams at Acadia National Park (Cadillac Brook and Hadlock Brook), mean total mercury concentrations were 0.6 and 1.5 ng/L, respectively, and methyl mercury averaged 0.05 and 0.07 ng/L, respectively, which represents 10% and 5% of total mercury in the two streams (K. Johnson and T. Haines, unpublished data). The estimate for Cadillac Brook methyl mercury may be high, as 0.05 ng/L is the detection limit for the analysis, and many samples were below detection. The mean was calculated by assuming that below detection results were half the detection limit. Concentrations of both total and methyl mercury were lower in stream water than in precipitation, but methyl mercury was higher relative to total mercury in the streams as compared to precipitation. This indicates that either there are sources of methyl mercury production in the watersheds, or that methyl mercury is less well retained in the watersheds relative to inorganic mercury.

Total mercury concentration in precipitation tends to be higher when precipitation volume is lower (Figure 1), which is probably related to wash-out of particulate mercury from the atmosphere. This pattern is not evident for methyl mercury (Figure 2) and in fact the arithmetic and volume-weighted annual mean concentrations are the same, suggesting that methyl mercury in the atmosphere may not be associated with particulate matter. In general, concentrations of total and methyl mercury follow the same temporal patterns (Figure 3), with high concentrations of total mercury normally accompanied by higher concentrations of methyl mercury. Although the source(s) of methyl mercury in the atmosphere is unknown, similar temporal trends in deposition suggests that at least some of the sources may be similar to those of inorganic mercury.

The annual deposition of total mercury for the period during which methyl mercury was determined was 6.65 $\mu g/m^2$, and deposition of methyl mercury was 0.082 $\mu g/m^2$, about 1.2%. During the period 1996-1999, total mercury deposition at the Acadia

site ranged from 7.7 to 9.0 µg/m^2, so this year was the lowest on record. Glass and Sorensen (1999) determined mean annual deposition of total mercury at six sites to be 7.4 µg/m^2, with individual sites ranging from 5.9 to 8.9 µg/m^2, which agree well with our findings. Methyl mercury was determined in only 36 weekly samples from the seven sites. Samples for methyl mercury analysis were collected once monthly during June to October in 1993, and not all stations were sampled each month. The mean concentration of methyl mercury was 0.18 ng/L, and the calculated annual deposition from these samples was 0.18 µg/m^2. The calculated total mercury annual deposition from these samples was 13.99 µg/m^2. These values are much higher than the Maine data, and may be artifacts of projecting 36 weekly samples to an annual rate. In Sweden, mean annual total mercury deposition was 7 µg/m^2 at Svartberget and 10 µg/m^2 at Gårdsjön during the period 1994-1998 (Lee *et al*. 2000), which are also similar to results in Maine. Methyl mercury deposition was 0.08 and 0.12 µg/m^2 respectively for the two Swedish watersheds during this time, agreeing well with our findings. At Little Rock Lake, Wisconsin, total mercury mean annual wet deposition was 6.8 µg/m^2 from 1988-1992, somewhat lower than in Maine, and methyl mercury deposition was 0.1 µg/m^2, somewhat higher than in Maine (Watras *et al*. 1994).

The weekly deposition of total and methyl mercury followed a similar temporal pattern (Figure 4), as was the case for concentration. The highest weekly methyl mercury deposition generally occurred in the winter, which is to be expected inasmuch as the concentration of methyl mercury was highest in snow samples.

Atmospheric deposition of methyl mercury at the Acadia site was generally similar to that determined at other locations as reported in the scientific literature. Methyl mercury is present in wet deposition, and generally amounts to about 1% of total mercury. Although the source of methyl mercury is unknown, the similarity in pattern to total mercury deposition suggests similar sources. It is likely that atmospheric deposition is not a significant source of methyl mercury in aquatic environments in Maine. Production of methyl mercury in the environment from deposited inorganic mercury is probably much more important as a source of contamination to aquatic biota. I do not recommend continuation of the determination of methyl mercury in precipitation as the data collected indicate that it is probably not important in Maine, and also because it was very difficult to deal with the analytical laboratory to obtain the data.

Literature Cited

Glass, G., and J. Sorensen. 1999. Six-year trend (1990-1995) of wet mercury deposition in the upper midwest, U.S.A. Environ. Sci. Technol. 33: 3303-3312.

Lee, Y., K. Bishop, and J. Munthe. 2000. Do concepts about catchment cycling of methylmercury and mercury in boreal catchments stand the test of time? Six years of atmospheric inputs and runoff export at Svartberget, northern Sweden. Sci. Tot. Environ. 260: 11-20.

Watras, C., and others. 1994. Sources and fates of mercury and methylmercury in Wisconsin lakes. Pages 153-177 in C. Watras and J. Huckabee (editors), Mercury Pollution Integration and Synthesis. Lewis Publishers, Boca Raton, Florida.

Table 1. Summary statistics for total mercury and methylmercury in precipitation at Acadia National Park.

Variable	Sample Type	Number	Mean	Std. Dev.	Minimum	Maximum
Total Hg, ng/L	All Samples	50	9.29	9.33	0	52.38
	Rain	30	11.51	10.41	1.36	52.38
	Snow	10	7.08	8.73	0	27.46
	Mixed	10	4.82	2.18	1.67	9.79
Methyl Hg, ng/L	All Samples	46	0.086	0.141	0	0.82
	Rain	27	0.065	0.078	0	0.36
	Snow	10	0.15	0.27	0	0.82
	Mixed	10	0.075	0.059	0.006	0.18
Percent Methyl	All Samples	47	0.93			
	Rain	27	0.56			
	Snow	10	2.12			
	Mixed	10	1.56			

Figure 1. Plot of total mercury concentration versus precipitation volume for the Acadia National Park Mercury Deposition Network site for the period July 2000 to June 2001.

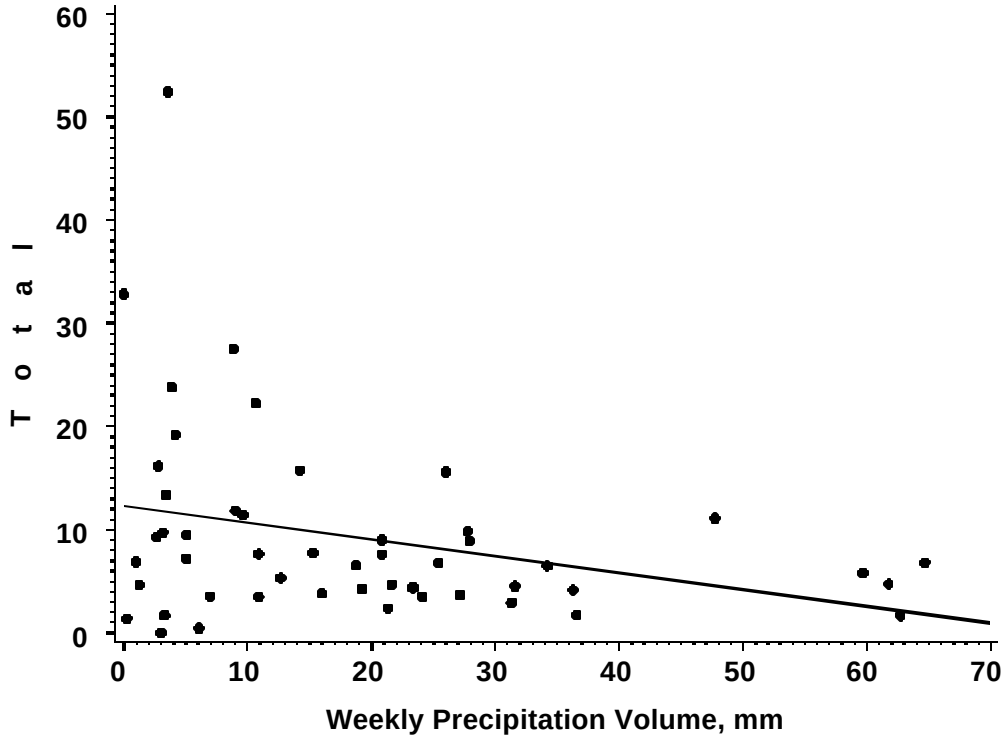


Figure 2. Plot of methyl mercury concentration versus precipitation volume for the Acadia National Park Mercury Deposition Network site for the period July 2000 to June 2001.

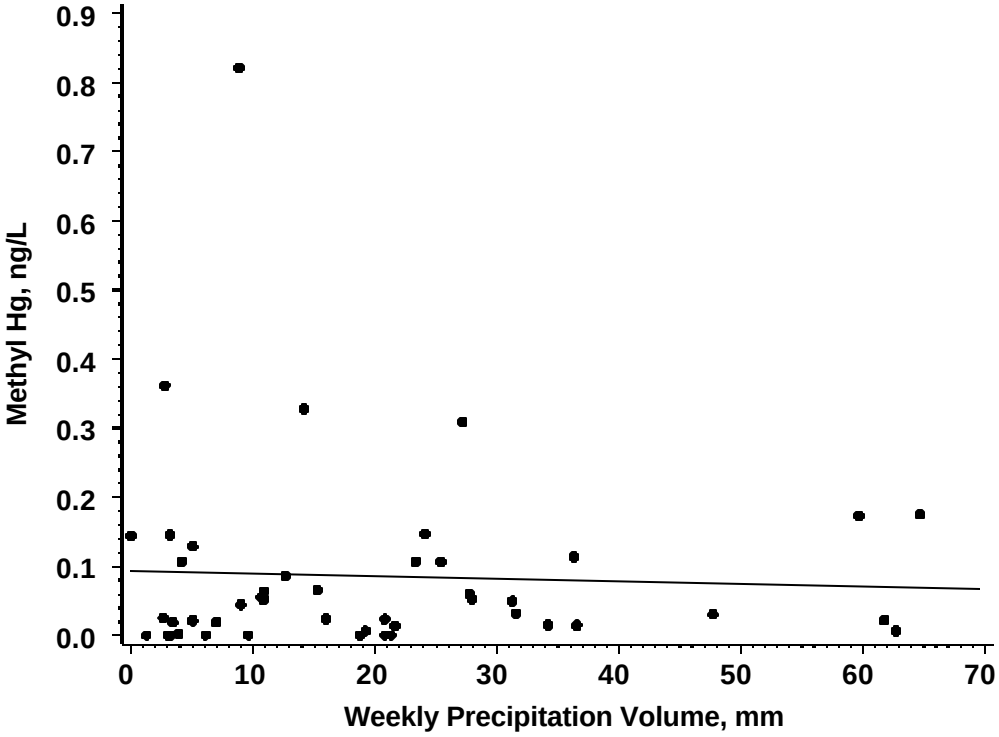


Figure 3. Plot of total and methyl mercury concentration for the Acadia National Park Mercury Deposition Network site for the period July 2000 to June 2001. Hg = solid line, MeHg = dashed line.

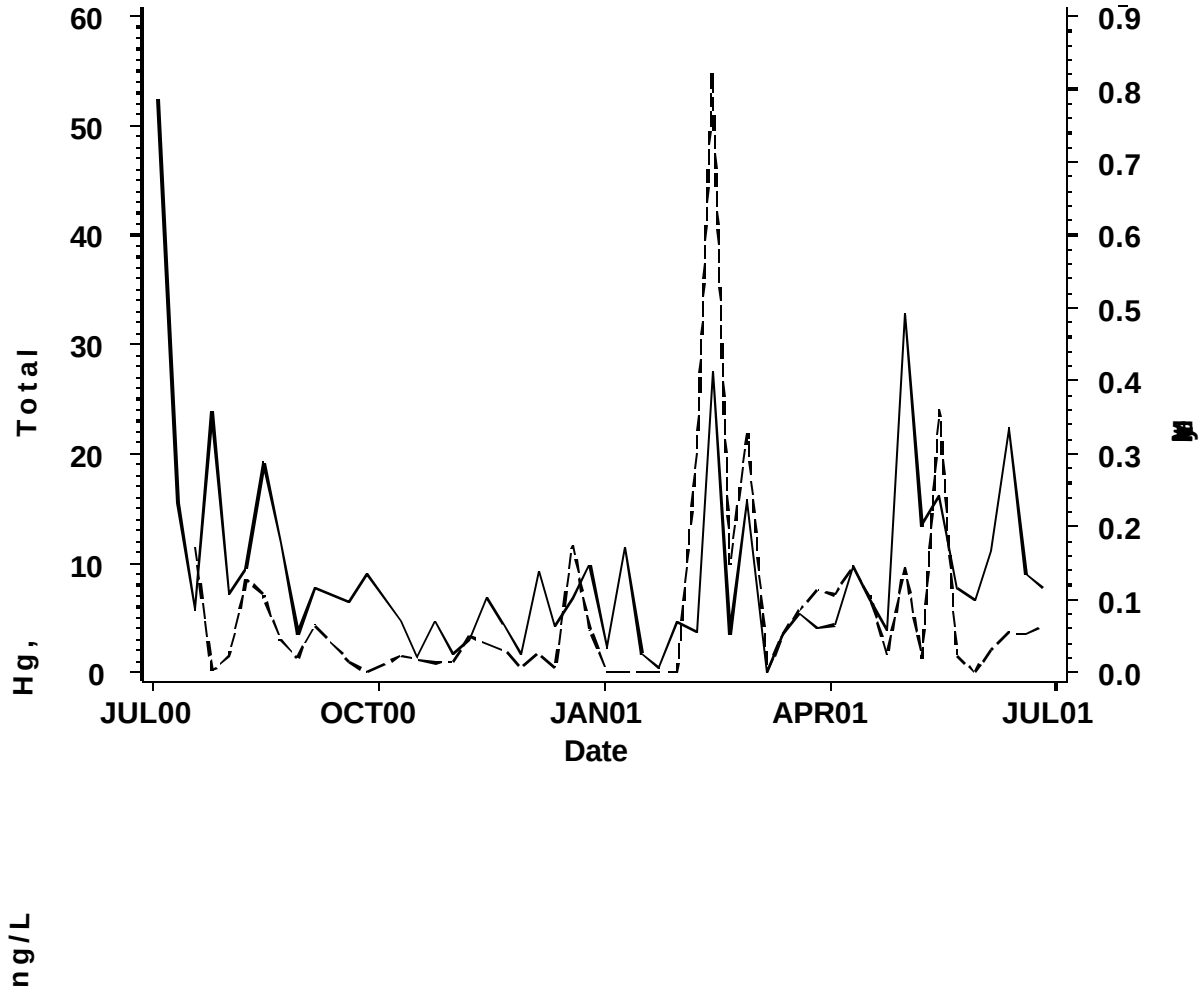
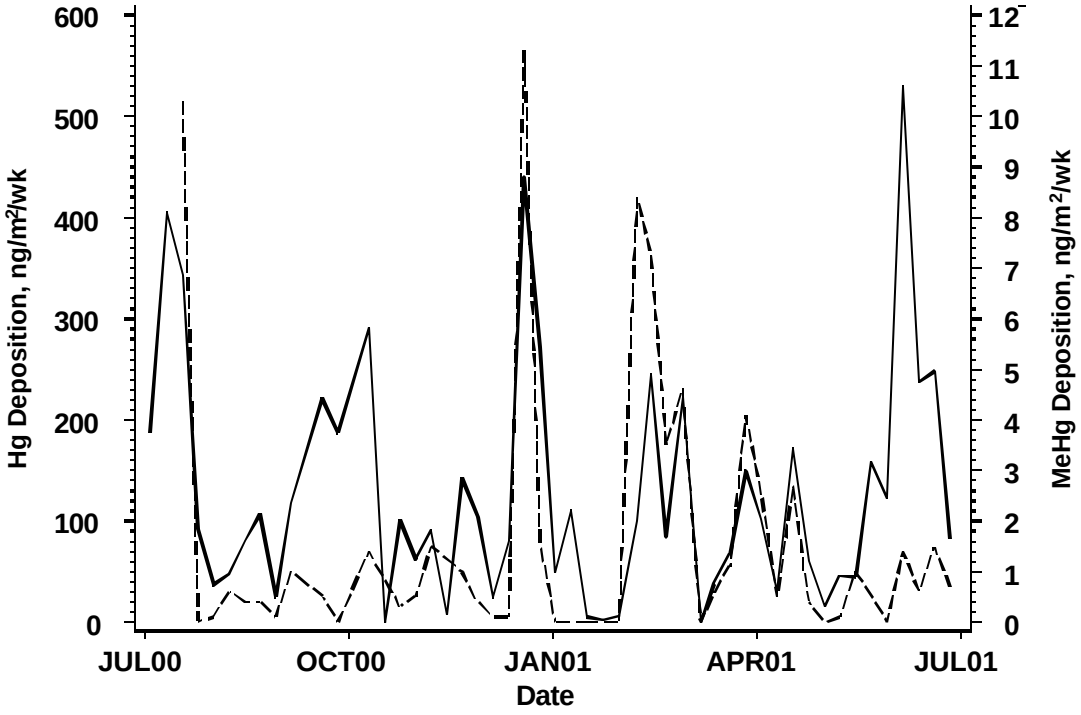


Figure 4. Plot of total and methyl mercury deposition for the Acadia National Park Mercury Deposition Network site for the period July 2000 to June 2001. Hg = solid line, MeHg = dashed line.



METHYLMERCURY LEVELS IN PRECIPITATION AT ACADIA NATIONAL PARK ME98

Site ID	Collection End Date	Precip. Hg Conc.	Weekly Hg Deposition	Precip. MHg Conc	Weekly MHg Deposition
ME98	07/03/00	52.38 ng/L	186.3 ng/m2	LE	LE
ME98	07/11/00	15.53 ng/L	404.3 ng/m2	1.187 ng/L	30.8 ng/m2
ME98	07/18/00	5.75 ng/L	343.2 ng/m2	0.173 ng/L	10.3 ng/m2
ME98	07/25/00	23.79 ng/L	92.1 ng/m2	0.002 ng/L	0.0 ng/m2
ME98	08/01/00	7.17 ng/L	36.4 ng/m2	0.021 ng/L	0.1 ng/m2
ME98	08/08/00	9.42 ng/L	47.9 ng/m2	0.128 ng/L	0.6 ng/m2
ME98	08/15/00	19.15 ng/L	80.3 ng/m2	0.106 ng/L	0.4 ng/m2
ME98	08/22/00	11.82 ng/L	106.6 ng/m2	0.044 ng/L	0.4 ng/m2
ME98	08/29/00	3.51 ng/L	24.5 ng/m2	0.019 ng/L	0.1 ng/m2
ME98	09/05/00	7.73 ng/L	118.3 ng/m2	0.065 ng/L	1.0 ng/m2
ME98	09/12/00	0.00 ng/L	0.0 ng/m2	0.000 ng/L	0.0 ng/m2
ME98	09/19/00	6.47 ng/L	221.3 ng/m2	0.014 ng/L	0.5 ng/m2
ME98	09/26/00	8.96 ng/L	186.6 ng/m2	0.000 ng/L	0.0 ng/m2
	Quarterly Sum:		**1847.7 ng/m2**		**44.1 ng/m2**
	Vol. Weighted Ave:	**9.84 ng/L**		**0.243 ng/L**	

Site ID	Collection End Date	Precip. Hg Conc.	Weekly Hg Deposition	Precip. MHg Conc	Weekly MHg Deposition
ME98	10/03/00	NA	NA	NR	0.0 ng/m2
ME98	10/10/00	4.70 ng/L	290.6 ng/m2	0.022 ng/L	1.4 ng/m2
ME98	10/17/00	1.36 ng/L	0.3 ng/m2	NR	0.0 ng/m2
ME98	10/24/00	4.64 ng/L	100.6 ng/m2	0.013 ng/L	0.3 ng/m2
ME98	10/31/00	1.67 ng/L	61.0 ng/m2	0.014 ng/L	0.5 ng/m2
ME98	11/07/00	2.88 ng/L	90.2 ng/m2	0.049 ng/L	1.5 ng/m2
ME98	11/14/00	6.83 ng/L	6.9 ng/m2	NR	0.0 ng/m2
ME98	11/21/00	4.49 ng/L	142.0 ng/m2	0.031 ng/L	1.0 ng/m2
ME98	11/28/00	1.66 ng/L	104.0 ng/m2	0.006 ng/L	0.4 ng/m2
ME98	12/05/00	9.26 ng/L	24.7 ng/m2	0.025 ng/L	0.1 ng/m2
ME98	12/12/00	4.20 ng/L	80.8 ng/m2	0.006 ng/L	0.1 ng/m2
ME98	12/19/00	6.80 ng/L	440.1 ng/m2	0.175 ng/L	11.3 ng/m2
ME98	12/26/00	9.79 ng/L	271.7 ng/m2	0.059 ng/L	1.6 ng/m2

Quarterly Sum:	**1612.9 ng/m2**		**18.1 ng/m2**

	Vol. Weighted Ave:	**4.27 ng/L**		**0.044 ng/m2**	
Site ID	**Collection End Date**	**Precip. Hg Conc.**	**Weekly Hg Deposition**	**Precip. MHg Conc**	**Weekly MHg Deposition**
ME98	01/02/01	2.32 ng/L	49.4 ng/m2	0.00 ng/L	0.0 ng/m2
ME98	01/09/01	11.44 ng/L	110.4 ng/m2	0.00 ng/L	0.0 ng/m2
ME98	01/16/01	1.69 ng/L	5.6 ng/m2	0.00 ng/L	0.0 ng/m2
ME98	01/23/01	0.42 ng/L	2.5 ng/m2	0.00 ng/L	0.0 ng/m2
ME98	01/30/01	4.62 ng/L	5.9 ng/m2	0.00 ng/L	0.0 ng/m2
ME98	02/07/01	3.68 ng/L	99.9 ng/m2	0.31 ng/L	8.4 ng/m2
ME98	02/13/01	27.46 ng/L	244.1 ng/m2	0.82 ng/L	7.3 ng/m2
ME98	02/20/01	3.47 ng/L	83.8 ng/m2	0.15 ng/L	3.5 ng/m2
ME98	02/27/01	15.70 ng/L	223.3 ng/m2	0.33 ng/L	4.6 ng/m2
ME98	03/07/01	0.00 ng/L	0.0 ng/m2	0.00 ng/L	0.0 ng/m2
ME98	03/13/01	3.45 ng/L	37.7 ng/m2	0.05 ng/L	0.6 ng/m2
ME98	03/20/01	5.28 ng/L	67.1 ng/m2	0.09 ng/L	1.1 ng/m2
ME98	03/27/01	4.13 ng/L	149.8 ng/m2	0.11 ng/L	4.1 ng/m2
	Quarterly Sum:		**1079.5 ng/m2**		**29.6 ng/m2**
	Vol. Weighted Ave:	**5.21 ng/L**		**0.148 ng/L**	
Site ID	**Collection End Date**	**Precip. Hg Conc.**	**Weekly Hg Deposition**	**Precip. MHg Conc**	**Weekly MHg Deposition**
ME98	04/03/01	4.35 ng/L	101.6 ng/m2	0.11 ng/L	2.5 ng/m2
ME98	04/10/01	9.70 ng/L	30.8 ng/m2	0.14 ng/L	0.5 ng/m2
ME98	04/17/01	6.73 ng/L	171.0 ng/m2	0.11 ng/L	2.7 ng/m2
ME98	04/24/01	3.82 ng/L	61.1 ng/m2	0.02 ng/L	0.4 ng/m2
ME98	05/01/01	32.78 ng/L	15.5 ng/m2	0.14 ng/L	0.0 ng/m2
ME98	05/08/01	13.33 ng/L	45.7 ng/m2	0.02 ng/L	0.1 ng/m2
ME98	05/15/01	16.11 ng/L	45.0 ng/m2	0.36 ng/L	1.0 ng/m2
ME98	05/22/01	7.60 ng/L	158.3 ng/m2	0.02 ng/L	0.5 ng/m2
ME98	05/29/01	6.53 ng/L	122.7 ng/m2	0.00 ng/L	0.0 ng/m2
ME98	06/05/01	11.08 ng/L	529.3 ng/m2	0.03 ng/L	1.4 ng/m2
ME98	06/12/01	22.26 ng/L	237.4 ng/m2	0.06 ng/L	0.6 ng/m2
ME98	06/19/01	8.88 ng/L	248.1 ng/m2	0.05 ng/L	1.5 ng/m2
ME98	06/26/01	7.62 ng/L	83.2 ng/m2	0.06 ng/L	0.7 ng/m2
	Quarterly Sum:		**1849.7 ng/m2**		**11.7 ng/m2**
	Vol. Weighted Ave:	**8.63 ng/L**		**0.054 ng/L**	

2.2

FISH CONSUMPTION ADVISORIES

LAKE TROUT

NORTHERN PIKE

CHAIN PICKEREL

DDT

We had hoped we could identify an indicator fish species and avoid the need to test multiple species for mercury contamination. However, our review of the data from the 'Indicator Species Study' in previous years does not appear to support this approach. The range of the ratios of mercury levels for the different species sampled does not seem consistent enough to identify a reliable predictor fish species, though this conclusion is somewhat compromised by the small number of lakes sampled.

Therefore, we are back to looking at obtaining data at the individual species level. Collapsing data into cold water versus warm water fish species is problematic because lake trout and brown trout have mercury levels more similar to warm water fish species than other cold water species, such as brook trout or landlocked salmon. Another important determinant of data needs is our desire to estimate a high percentile lake average fish-mercury concentration rather than the statewide mean. Anglers do not necessarily fish lakes randomly or fish a large number of water bodies (if they did, the mean would be the appropriate statistic). Rather, they may have one or just a few lakes or ponds they primarily fish (especially for those people living on a lake). Consequently, we believe we need to evaluate the likelihood that individuals may routinely consume fish from a high-end lake. To do this, we need sufficient data to estimate the statewide distribution for fish species routinely consumed and to estimate high percentile lakes (e.g., 75^{th} to 95^{th} percentile lake).

Based on the white perch data, we think this will require data on about 50 to 60 lakes (current data suggests percent relative standard deviations for lake averages for fish species generally ranging from 30 - 60%; white perch has a %RSD of 50%). For some important species such as lake trout (important based on angler consumption surveys and a survey of women of childbearing age), our current database is very limited (N=8). Brown trout (N=8) and pickerel (N=7) are other species that have very limited data. We have no data on pike and very little data on black crappie. Consequently, these species are our priorities for obtaining additional data.

As a general approach to obtaining additional data, we propose that we select a specific species or two for a focussed sampling program in a given year (e.g., this year we sampled lake trout, pike, and pickerel for study). We propose to continue this program until we reach our goal of 50 lakes per major fish species than re-evaluate where we are. This will likely take several years to accomplish.

Lake Trout Study. Current data for lake trout are very limited, and 90th and 95th percentile lake averages cannot be estimated with the desired degree of confidence. Importantly, this fish species was reported to be the second most commonly consumed fish species based on the preliminary results from our random survey of women of childbearing age. We currently have an 8 lake random sample from the REMAP study, and a 7 lake study where lakes were selected based on the presence of 4 predator species (warm and cold water species). Statistical tests indicate that these two data sets should not be pooled (underlying statistical distributions appear significantly different). Consequently there is a strong need to expand the current database to better characterize the statewide distribution.

To this end, DEP requested that DIFW collect lake trout in performance of its own duties, noting the following: a) which lakes they intended to sample in 2000, b) which lakes are considered to have significant angler pressure, and c) which lakes have primarily stocked versus natural populations. DIFW successfully provided samples of lake trout from 11 lakes and a sample of splake from 1 lake.

Mean size did not vary much among the lakes and was not correlated with mean mercury concentrations. Concentrations varied considerably, but the mean (0.36 mg/kg, n=11) (Table 2.2.1) was

not significantly different from the REMAP data (0.46 mg/kg, n=8) or the indicator species (0.60 mg/kg, n=4) data for skinless fillets (Mann Whitney U test, p=0.05).

Northern Pike. There were no data on pike that are apparently found in a number of waters in central Maine, such as the Belgrade Lakes, Sabattus Lake, and the Annabessacook - Cobbosseeconte Chain. Pike are big predatory fish and would be expected to have higher mercury concentrations than even pickerel. The goal was to catch 5 pike from the Belgrade Lakes region, Sabattus Lake, and Annabessacook. Lake. We were able to capture pike from only Great Pond in Belgrade and Sabattus Pond in Sabattus. Concentrations were greatly different, being much higher in Great Pond and surprisingly low in Sabattus, even though those fish were smaller (Table 2.2.1). Collection of pike from Sabattus Pond was repeated in 2001.

Chain Pickerel. There are mercury data from only 7 lakes sampled for chain pickerel , which appear to be high in mercury, though standard deviations are low. More data were needed to get a better sense of the underlying distribution, but it was unclear whether new data would have much of an effect on the advisory. DEP asked DIFW to collect 5 pickerel from each of 5 lakes in the course of their normal duties. We received a sample from only Great Pond in Belgrade and the concentration was much lower than the previous data for chain pickerel from the REMAP project and also lower than from the pike in Great Pond. The study was repeated in 2001 and results will be reported in the 2001 report.

Confirming REMAP DDT analysis. From the 1993-94 REMAP study of Maine lakes, 15 lake/species samples were identified as having fish with elevated total DDT that exceeded Bureau of Health fish tissue action level (FTAL=64 ppb) in edible filets. Attempts were made to collect 5 fish for each of these combinations to be analyzed for total DDT. A total of seven samples of fish were captured from a total of 5 lakes. We were unable to capture some species from some lakes, other lakes were not visited. Total DDT concentrations were much lower than those from the REMAP project (Table 2.2.2). Most of the REMAP data were flagged for some sort of quality assurance exceedance. None of the 2000 samples exceeded the FTAL.

Table 2.2.1 Mercury concentrations in 2001 fish samples from some Maine lakes.

WATER	MIDAS No.	TOWN	SPECIES CODE	HG mg/kg
Auburn Lake	3748	Auburn	LKT	0.15
Allagash L	9787	T8R14 WELS	LKT	0.61
Eagle Lake	1634	Eagle Lake	LKT	0.37
E Musquash L	1088	Topsfield	LKT	0.63
Haymock L	2814	T8R11WELS	LKT	0.24
Hurd Pond	2064	T2R10WELS	LKT	0.24
Kezar Lake	0097	Lovell	LKT	0.38
Millimagasett L	3004	T7R8	LKT	0.44
Mattagamon Lake	4260	Trout Brook Twp	LKT	0.53
Nickerson Lake	1036	New Limerick	LKT	0.26
Pleasant Pond, Island Falls	0224	Caratunk	LKT	0.13
Thissell Pond	2726	T5R11WELS	SPK	0.24
Sabattus Pond	3796	Greene	PIK	0.06
Great Pond	5274	Belgrade	PIK	0.45
Great Pond	5274	Belgrade	PKL	0.28

DEP Sample ID	Length mm	HG mg/kg
LAKE TROUT		
Allagash L		
LK-9787-LKT-1	479	0.482
LK-9787-LKT-2	500	0.389
LK-9787-LKT-3	465	0.389
LK-9787-LKT-4	502	0.634
LK-9787-LKT-5	445	0.429
LK-9787-LKT-6	552	1.23
LK-9787-LKT-7	500	0.654
LK-9787-LKT-8	479	0.519
LK-9787-LKT-9	525	0.674
LK-9787-LKT-10	517	0.676
MEAN	**496**	**0.61**
Auburn L		
LK-3748-LKT-1	456	0.071
LK-3748-LKT-2	520	0.185
LK-3748-LKT-3	505	0.143
LK-3748-LKT-4	530	0.133
LK-3748-LKT-5	535	0.193
LK-3748-LKT-6	500	0.159
MEAN	**508**	**0.15**
Eagle Lake		
LK-1634-LKT-1	503	0.329
LK-1634-LKT-2	475	0.383
LK-1634-LKT-3	480	0.364
LK-1634-LKT-4	478	0.346
LK-1634-LKT-5	559	0.429
MEAN	**499**	**0.37**
E Musquash L		
EMQ-LKT-01	551	0.684
EMQ-LKT-02	596	0.785
EMQ-LKT-03	535	0.552
EMQ-LKT-04	460	0.499
MEAN	**536**	**0.63**
Haymock Lake		
LK-2814-LKT-1	605	0.225
LK-2814-LKT-2	551	0.262
LK-2814-LKT-3	617	0.265
LK-2814-LKT-4	618	0.211
LK-2814-LKT-5	615	0.354
LK-2814-LKT-6	564	0.299
LK-2814-LKT-7	443	0.161
LK-2814-LKT-8	427	0.18
LK-2814-LKT-9	492	0.133
LK-2814-LKT-10	510	0.299
MEAN	**544**	**0.24**

DEP Sample ID	Length mm	HG mg/kg
Hurd Pond		
LK-2064-LKT-1	355	0.143
LK-2064-LKT-2	383	0.3
LK-2064-LKT-3	404	0.183
LK-2064-LKT-4	410	0.259
LK-2064-LKT-5	424	0.307
MEAN	**395**	**0.24**
Kezar Lake		
LK-0097-LKT-1	446	0.321
LK-0097-LKT-2	582	0.544
LK-0097-LKT-3	506	0.326
LK-0097-LKT-4	515	0.303
LK-0097-LKT-5	482	0.415
MEAN	**506**	**0.38**
Mattagamon Lake		
LK-4260-LKT-1	491	0.542
LK-4260-LKT-2	423	0.517
LK-4260-LKT-3	444	0.316
LK-4260-LKT-4	574	0.679
LK-4260-LKT-5	486	0.589
MEAN	**484**	**0.53**
Millimagassett Lake		
LK-3004-LKT-1	530	0.356
LK-3004-LKT-2	663	0.451
LK-3004-LKT-3	620	0.522
LK-3004-LKT-4	536	0.442
LK-3004-LKT-5	557	0.44
MEAN	**581**	**0.44**
Nickerson Lake		
LK-1036-LKT-1	578	0.47
LK-1036-LKT-2	497	0.188
LK-1036-LKT-3	557	0.45
LK-1036-LKT-4	318	0.121
LK-1036-LKT-5	330	0.174
LK-1036-LKT-6	330	0.136
MEAN	**435**	**0.26**

DEP Sample ID	Length mm	HG mg/kg
Pleasant Pond, Caratunk		
LK-0224-LKT-1	482	0.069
LK-0224-LKT-2	536	0.117
LK-0224-LKT-3	516	0.107
LK-0224-LKT-4	547	0.154
LK-0224-LKT-5	516	0.103
LK-0224-LKT-6	559	0.148
LK-0224-LKT-7	502	0.102
LK-0224-LKT-8	523	0.115
LK-0224-LKT-9	570	0.256
LK-0224-LKT-10	510	0.127
MEAN	**526**	**0.130**
Thissell Pond		
LK-2726-SPK-1	395	0.209
LK-2726-SPK-2	425	0.26
LK-2726-SPK-3	425	0.237
LK-2726-SPK-4	391	0.191
LK-2726-SPK-5	467	0.31
MEAN	**421**	**0.24**
NORTHERN PIKE		
Sabattus Pond		
SPS-PKE-1	461	0.056
SPS-PKE-2	410	0.047
SPS-PKE-3	448	0.054
SPS-PKE-4	446	0.06
SPS-PKE-5	447	0.064
MEAN	**442**	**0.06**
Great Pond, Belgrade		
GRT-PIK-01	728	0.749
GRT-PIK-02	697	0.382
GRT-PIK-03	666	0.378
GRT-PIK-04	670	0.299
GRT-PIK-05	653	0.459
MEAN	**683**	**0.45**
CHAIN PICKEREL		
Great Pond, Belgrade		
GRT-PKL-01	393	0.204
GRT-PKL-02	380	0.261
GRT-PKL-03	465	0.347
GRT-PKL-04	497	0.313
MEAN	**434**	**0.28**

Table 2.2.2 Total DDT concentrations in 2000 fish samples from some Maine lakes

Location	Station Code	Species	Total DDX nd=1/2 mdl
Eagle Lake			
Eagle Lake	LK1634	LKT	2.9
Little Ossipee Pond			
Waterboro	LOW	LLS	3.0
Lovewell Pond			
Fryeberg	LPF	BNT	15.9
Round Pond	RPL	SMB	6.8
Livermore		WHS	61.9
Lower Range Pond	LRP	BNT	4.1
Poland		WHS	27.6

DEP ID#	DL	LK1634-LKT-1	LK1634-LKT-2	LK1634-LKT-3	LK1634-LKT-4	LK1634-LKT-5
Compound	**ng/kg**					
2,4-DDE	1.0	<DL	<DL	<DL	<DL	<DL
4,4-DDE	1.0	0.25	0.19	0.31	0.56	0.47
2,4-DDD	1.0	0.34	0.65	0.22	0.48	0.79
4,4-DDD	1.0	<DL	<DL	<DL	<DL	<DL
2,4-DDT	1.0	<DL	<DL	<DL	<DL	<DL
4,4-DDT	1.0	<DL	<DL	<DL	<DL	<DL
Total DDX		0.59	0.84	0.53	1.04	1.26
TCMX (% rec.	65-125	77.5	81.6	89.4	82.3	77.1
Sample weight (g)		24.9	25.1	24.9	25.0	24.9

DEP ID#	DL	LPF-BNT-1	LPF-BNT-2	LPF-BNT-3	LPF-BNT-4	LPF-BNT-5
Compound	**ng/kg**					
2,4-DDE	1.0	<DL	<DL	<DL	<DL	<DL
4,4-DDE	1.0	<DL	<DL	0.88	1.28	0.75
2,4-DDD	1.0	2.87	1.59	2.66	10.93	8.15
4,4-DDD	1.0	1.32	1.15	1.44	3.20	2.66
2,4-DDT	1.0	3.24	2.97	2.36	4.24	3.87
4,4-DDT	1.0	4.48	3.88	4.87	3.32	4.01
Total DDX		11.92	9.59	12.21	22.97	19.44
TCMX (% rec.	65-125	77.1	81.4	70.2	80.2	91.0
Sample weight (g)		25.0	25.0	25.0	25.0	25.1

DEP ID#	DL	LOW-LLS-1	LOW-LLS-2	LOW-LLS-3	LOW-LLS-4	LOW-LLS-5
Compound	**ng/kg**					
2,4-DDE	1.0	<DL	<DL	<DL	<DL	<DL
4,4-DDE	1.0	<DL	<DL	<DL	<DL	<DL
2,4-DDD	1.0	<DL	<DL	<DL	<DL	<DL
4,4-DDD	1.0	<DL	<DL	<DL	<DL	<DL
2,4-DDT	1.0	<DL	<DL	<DL	<DL	<DL
4,4-DDT	1.0	<DL	<DL	<DL	<DL	<DL
Total DDX		0.00	0.00	0.00	0.00	0.00
TCMX (% rec.	65-125	75.5	88.7	70.3	70.0	75.3
Sample weight (g)		25.0	24.9	25.0	25.1	25.1

DEP ID#	DL	LRP-BNT-1	LRP-BNT-2	LRP-BNT-3	LRP-BNT-4	LRP-BNT-5
Compound	**ng/kg**					
2,4-DDE	1.0	<DL	<DL	<DL	<DL	<DL
4,4-DDE	1.0	0.32	0.41	<DL	<DL	0.28
2,4-DDD	1.0	<DL	<DL	<DL	<DL	<DL
4,4-DDD	1.0	0.52	0.56	<DL	<DL	0.52
2,4-DDT	1.0	2.12	1.95	<DL	<DL	<DL
4,4-DDT	1.0	1.80	<DL	<DL	<DL	1.83
Total DDX		4.76	2.92	0.00	0.00	2.63
TCMX (% rec.	65-125	75.8	75.5	89.3	89.7	93.5
Sample weight (g)		25.0	25.0	25.1	25.0	25.1

DEP ID#	DL	RPL-SMB-1	RPL-SMB-2	RPL-SMB-3	RPL-SMB-4	RPL-SMB-5
Compound	**ng/kg**					
2,4-DDE	1.0	<DL	<DL	<DL	<DL	<DL
4,4-DDE	1.0	0.40	<DL	0.69	0.28	0.34
2,4-DDD	1.0	<DL	<DL	<DL	<DL	<DL
4,4-DDD	1.0	0.48	<DL	1.18	0.48	0.51
2,4-DDT	1.0	2.04	1.77	4.73	1.96	2.18
4,4-DDT	1.0	<DL	1.81	4.53	1.88	2.01
Total DDX		2.92	3.58	11.13	4.60	5.04
TCMX (% rec.	65-125	84.2	76.2	72.4	65.4	68.4
Sample weight (g)		25.0	24.9	10.2	25.0	23.4

DEP ID#	DL	LRP-WHS-1	LRP-WHS-2
Compound	**ng/kg**		
2,4-DDE	1.0	<DL	<DL
4,4-DDE	1.0	24.7	15.72
2,4-DDD	1.0	1.68	1.24
4,4-DDD	1.0	4.13	2.03
2,4-DDT	1.0	<DL	<DL
4,4-DDT	1.0	1.84	1.84
Total DDX		32.39	20.82
TCMX (% rec.	65-125	77.6	71.5
Sample weight (g)		24.9	25.0

DEP ID#	DL	RPL-WHS-1	RPL-WHS-2	RPL-WHS-3	RPL-WHS-4	RPL-WHS-5
Compound	**ng/kg**					
2,4-DDE	1.0	<DL	<DL	3.96	<DL	<DL
4,4-DDE	1.0	6.21	16.0	128	15.7	4.46
2,4-DDD	1.0	4.25	18.8	49.5	13.6	2.27
4,4-DDD	1.0	1.26	3.43	16.6	<DL	1.04
2,4-DDT	1.0	2.03	10.3	<DL	<DL	5.82
4,4-DDT	1.0	<DL	<DL	<DL	<DL	<DL
Total DDX		13.75	48.57	198.49	29.28	13.59
TCMX (% rec.	65-125	79.6	86.3	87.6	68.9	69.1
Sample weight (g)		25.0	25.1	25.0	25.1	25.1

2.3

LOON EFFECTS STUDY

Assessing the impacts of methylmercury on piscivorous wildlife: as indicated by the Common Loon, 1998-2000

(Report BRI00-01)

2000 Final Report

Submitted to:

Maine Department of Environmental Protection
Surface Water Ambient Toxic Monitoring Program
State House Station 17
Augusta, Maine 04333

Submitted by:

David C. Evers, Chris De Sorbo, and Lucas Savoy
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23 March 2001

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Executive Summary:

Anthropogenic inputs of mercury (Hg) into the environment have significantly increased in the past few decades. In conjunction, the current availability of methylmercury (MeHg) in aquatic systems has increased to levels posing risks to human and ecological health. Risk levels vary considerably in response to MeHg availability, which is affected by lake hydrology, biogeochemistry, habitat, topography, and proximity to airborne sources. We selected the Common Loon as the most suitable bioindicator of aquatic Hg toxicity, based on ecological, logistical, and other criteria, including public valuations of natural resources. Opportunistic and probability-based sampling efforts from 1994-2000 indicate New England's breeding loon population is at unacceptable levels of risk to Hg contamination, particularly in Maine. Based on risk categories developed from the literature and *in situ* studies by BioDiversity Research Institute and their collaborators, 30% of the breeding loon population in Maine is estimated to be at risk, while 46% of the eggs laid are potentially impacted.

Because results from national sampling indicated loons were at most risk from Hg in New England (particularly Maine), we identified several individual- and population-level parameters to better understand the extent of mercury toxicity across Maine. Between 1994-00, we collected 139 abandoned eggs as well as blood and feather samples from 253 adult and 103 juvenile wild loons captured in Maine. The Hg concentrations in these samples were used to characterize sublethal impacts of Hg on egg development, behavior, developmental stability, immunosuppression, individual survival, and overall reproductive success. In the Rangeley Lakes Study Area, a total of 185 loon territories were monitored on 43 lakes during 1998-00. Current monitoring efforts and historical data comprise 515 territory-years measured. Behavioral observations were conducted for over 1,500 hours on 16 lakes with 38 loon territories from 1998 to 2000.

Several reproductive measures significantly declined for loon pairs at high risk to prey MeHg availability, thereby corroborating studies in high-risk sites in Nova Scotia and Wisconsin that show Hg impacts reproductive success. Based on 223 loon territories representing 748 territory-years surveyed we found that extra-high risk pairs fledged 37% fewer young than pairs at low risk to Hg. We also found similar significant patterns of lower productivity on high and extra-high risk territories compared to low and moderate risk territories for other reproductive measures. We view the implication of long-term declines in these reproductive measures are serious and contend they would not be detected by traditional survey techniques.

Insight into why loons are facing Hg-based population declines can be seen through our hazard assessment process that is based on a weight of evidence approach. Physiological impacts of Hg are measured through two key biomarkers: corticosterone stress hormone levels and flight feather asymmetry. Circulating corticosterone hormone levels are strongly linked with increasing blood Hg levels and are not related to capture and handling stress. Corticosterone hormone levels increase on an average of 14.6% for every one ppm of increase in blood Hg levels (n=239). This indicates that loons with high blood Hg levels have higher rates of chronic stress and may therefore have compromised immune systems. Asymmetry measurements provide insights into developmental stability and potentially reproductive fitness. Three years of flight feather measurements have shown annual agreement that loon breeding populations with greater exposure to Hg have significantly greater asymmetry than populations at low risk (n=227). Greater asymmetry may indicate disruptions from stressors on their embryonic

development and current physiological status as well as a potential decline in reproductive fitness.

Many behavioral impacts that appear to be related to the neurotoxic effects of MeHg can rarely be observed in the field. We found adult loons in high risk situations left eggs uncovered 14% of the time, compared to 1% in controls. Several cases of direct field observations indicate that adult loons with high Hg body burdens avoid incubating their eggs and display atypical behaviors such as patrolling in front of, or sitting next to the nest. We documented a significant negative relationship between adult blood Hg and foraging behavior, and a significant positive relationship between adult blood Hg and brooding behavior. Recategorizing our data according to energy demands revealed a significant inverse relationship between blood Hg and time spent in high energy behaviors. Our findings are consistent with other studies linking Hg and lethargy, reduced motivation to hunt prey, and compromised foraging abilities.

Current levels of Hg in Maine's lacustrine ecosystems also appear to be impacting individual survival of adult and juvenile loons. Recaptured adult loons exhibit a significant annual increase of Hg (9% in males, 5.6% in females) that we predict will significantly reduce lifetime individual performance. A model of this impact indicates a decline of 13 to 8 young produced over a loon's lifetime. Further, juveniles from high-risk territories have significantly increasing blood Hg levels of 3% per day during the summer, potentially reaching dangerous levels after the final feather molt at 11 weeks of age.

Characterization of the risk imposed by MeHg bioavailability in aquatic systems to high trophic level obligate piscivores such as the Common Loon indicates negative population level impacts in Maine. Although the impacts of Hg on loons are varied, complex, and not yet fully understood, the combination of high exposure to a significant part of the breeding population and the "bottom-line" impact of reducing overall reproductive success to 37%, has created an aquatic landscape that is not sustainable for the Common Loon in Maine.

Current models indicate a negative population growth rate. Because of the loon's life history strategy (i.e., long lived, slow maturing, and low fecundity) the annual and continual impacts of this type of stressor causes an erosion of the non-breeding or buffer population that serves as a natural cushion to catastrophic events. Once this buffer population is exhausted, the occupancy of established territories will shrink and it will be more obvious that loon populations are declining. However, the realization of shrinking loon populations at that stage will require drastic and potentially expensive efforts to reverse the decline. Models based on a 25-year, statewide comprehensive monitoring effort in New Hampshire show approximately half of Maine's buffer population has been exhausted. Certain areas in Maine, such as the Allagash area that may be particularly impacted from Hg, may already exhibit exhaustion of the buffer population and a shrinking number of territorial pairs.

Continued refinement of model parameters and either a probability-based sampling scheme or new sampling efforts in northern Maine will provide higher confidence in our estimates that will therefore assist in state-based policy efforts as well as national regulations that reflect the ecological injury Hg is currently having on the freshwater landscape.

The full report is available with the 2000 SWAT report separately at
http://www.state.me.us/dep/blwq/monitoring.htm

2.4

WILDLIFE CONTAMINANTS

Investigation of mercury exposure in Maine's Mink and River Otter: Final 2000 Report

(Report BRI-2001-06)

Submitted to:

Wally Jakubas, Maine Inland Fisheries and Wildlife and
Barry Mower, Maine Department of Environmental Protection &
Drew Major, U.S. Fish & Wildlife Service

Submitted by:

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Submitted on: 6 December 2001

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APPENDICES

OBJECTIVE

Obtain mink (*Mustela vison*) and river otter (*Lutra canadensis*) carcasses from trappers to analyze liver, brain, muscle, and fur for total mercury.

INTRODUCTION

Mercury and other aquatic-based persistent bioaccumulative toxins are prevalent in Maine's freshwater and marine environments (Maine DEP 1998). Methylmercury (MeHg) availability to fish and wildlife varies geographically and is strongly influenced by hydrology and biogeochemical factors (Evers and Reaman 1998, Evers et al. 1998b). To interpret exposure levels in wildlife established benchmarks are needed. Therefore, standardized sampling of target biosentinels provides a method for making informed comparisons and definitive interpretations, thereby helping to assess risks to wildlife and allow landscape-level extrapolations of the hazards.

The mink and the river otter are both widely distributed in New England and Maine. Both species have diets that include fish and crayfish, although mink are known generalists. Because of their high metabolism and piscivorous diet, both mink and river otters are highly susceptible to elevated levels of environmental MeHg.

Context

Lab-based, dose-response studies of mink (Wobeser et al. 1976) and otter (O'Connor and Neilson 1980) have shown that terminal total Hg concentrations occur at 25 ppm (ww) in the liver and kidney. Thompson (1996) estimated that 30 ppm (ww) of total Hg in the liver or kidney is at least sublethal and potentially lethal. He also reported that dietary MeHg concentrations of 2 to 6 ppm (ww) were "sufficient to cause mercury intoxication."

Although fish total Hg levels over 2 ppm occur in Maine, they are relatively rare. However, fish total Hg levels greater than 1 ppm are common. Evers and Reaman (1998) found fillets from Land-locked Salmon (*Salmo salar*)in Pierce Pond (1.06 ppm, ww for a 53 cm fish), Yellow Perch (*Perca flavescens*) in Mooselookmeguntic (1.11 ppm, ww for a 34 cm fish), and Yellow Perch in Flagstaff (1.26 ppm, ww for a 29 cm fish) exceeded these levels. They also found fillet Hg levels to significantly increase as fish size (indexed by length x weight) increased for Land-locked Salmon, Smallmouth Bass (*Micropterus dolomieu*), and Yellow Perch. Nearly all Hg in fish is MeHg (Wiener and Spry 1996).

As evidenced by empirical studies conducted by BioDiversity Research Institute (BRI) in Maine and comparisons with other studies (Table 1 and 2), mink and river otter are likely exposed to sufficient quantities of dietary Hg to cause sublethal impacts. Evers et al. (1998a) found Common Loon (*Gavia immer*) Hg levels to show an increasing west-east trend. Mean juvenile loon blood mercury levels from Maine were 4.5x higher than Alaska and 2x higher than the upper Great Lakes and Ontario. On several of Maine's reservoirs (e.g., Flagstaff Lake), juvenile loon blood Hg levels were up to 10x higher than Great Lakes sites.

Table 1. Concentrations of total Hg (ppm,ww) in river otter from various study sites. All values in parentheses are ranges and single values are means.

Site / Tissue	*Muscle*	Brain	Liver	Kidney	Fur*	Source
Ireland			(0.15-17.03)			Mason 1993
Denmark			(0.03-12.37)			Mason 1992
Britain			(0.17-4.33)	(0.08-2.02)		Mason 1988
New York			2.35			Mayack 1994
Ontario 1	36	30	96	58	47	Wren 1985
Ontario 2	0.89		2.97	1.05		Wren 1980
Ontario 3			(1.0-3.5)			Wren 1988
Wisconsin	1.44	0.74	3.44	8.47	6.47	Sheffy 1982

* fresh weight

Table 2. Concentrations of total Hg (ppm,ww) in mink from various study sites. All values in parentheses are ranges and single values are means.

Site	Muscle	Brain	Liver	Kidney	Fur*	Source
Wisc	1.26	0.46	2.08	2.33	7.61	Sheffy 1982
Conn			(1.1-8.47)			Major 1991
Mass			(0.008-1.92)			Major 1991
New York			2.35			Mayack 1994
Ohio			0.135			Lynch 1973
Quebec 1	1.87	0.83	9.23			Desai-Greenway 1976
Quebec 2	2.4 (0.41-6.2)		8.34 (2.21-20.0)			Langis 1999
Saskatchewan			58.2	31.9		Wobeser 1976

* Fresh weight

Because the otter and mink prey base is similar to the loon's, their body burdens of Hg may be comparable. For example, if a young loon has blood Hg levels 10x higher than

same-aged loons in the Great Lakes, then otter from Wisconsin (i.e., liver mercury was 3.44 ppm, ww) (Table 1) should be 10x higher on high Hg lakes in Maine (i.e., 34.4 ppm, ww). Similarly, mink with 2.08 ppm of mercury in their liver in Wisconsin (Table 2) could potentially have up to 20.8 ppm, ww in Maine (Table 3). As mentioned, 30 ppm, ww in the liver or kidney is considered lethal (Wren (1985) and Wobeser (1976)).

STUDY AREA & METHODS

Study area

Previous mercury-based studies in Maine and throughout New England provided extensive information on known hotspots (Evers et al. 1998a), aquatic scenarios prone to enhanced MeHg availability (Evers and Reaman 1998), and species most at risk (Evers et al. 1998b). Flagstaff Lake, the North Branch of the Dead River and its watershed including Chain-of-Ponds, and the Dead River outflow from the Flagstaff dam have some of the highest levels of biotic Hg in the country. Because of this known hotspot and background information on the fish and crayfish mercury levels we focused collection of otter and mink carcasses from this area. Another focus area was the Seboomook Lake region, where trappers have reported extirpations of mink. However, trappers in the Seboomook Lake region took no animals. Collection of carcasses from other areas in Maine was opportunistic and based on availability (Appendix 1).

Sample collection and processing

We collected 8 river otter and 24 mink carcasses from licensed fur trappers during the 2000-2001 trapping season (Appendix 2 and 3). Carcasses were stored on-site in freezers and regularly retrieved by BRI staff. Brain, femoral muscle and liver tissue were removed using stainless-steel instruments and placed into I-CHEM® jars. Fur was taken from the foot of the animal using stainless-steel instruments then cleaned and placed into sealed envelopes. The tissues, once harvested, were refrozen until they were sent to the lab. The tissue samples were harvested at the University of Southern Maine's Biology lab using techniques according to Tufts University Animal Wildlife clinic protocols (M. Pokras, pers. com.).

Fur, brain and liver tissues were analyzed for total mercury using Cold Vapor Atomic Absorption (CVAA) methods. Laboratory analysis was conducted by Texas A&M Trace Element Research Lab (TERL). Femoral muscle tissue were archived for future analysis. TERL has conducted BRI's mercury analysis for bird tissues (blood, feathers, and eggs), fish, and crayfish for the past three years. Mercury level results are given as fresh weight for fur and wet weight for liver and brain. Methylmercury levels were not analyzed.

Contacts for retrieving carcasses

Dave Yates discussed logistics of carcass retrieval with the following trappers: Jim Arsenault of Dresden, Chester Brewer of Boothbay, Bobby Cercena of Eustis, Jerry Le Beau of North Anson, Yukkies Taxidermy of Stratton, Oscar Cronk of Wiscassett, and Brett Damm of Sumner. He also met trappers in the Boothbay area during a trapper safety course sponsored by Inland Fisheries and Wildlife (where he received his trapping certificate # METS-025-00-006).

RESULTS AND DISCUSSION

River Otter

Fur Hg concentrations ranged between 5 ppm in otters from Wiley Pond to 30.5 ppm on the St. John River. Otter fur Hg levels indicate individuals from several sites are elevated (Table 3). Brain total Hg levels ranged from 0.08 to 0.69 ppm while liver total Hg levels ranged from 0.24 to 4.74 ppm (Table 3).

Wren (1985) showed that Ontario river otters with mean fur Hg levels of 47 ppm had on average 30 ppm and 96 ppm total Hg in the brain and liver respectively. Lethal levels are considered 30 ppm total Hg in the liver (Thompson 1996) and 19 ppm total Hg in the brain (Mierle et al. 2000). Although our fur Hg levels approach lethal levels, brain and liver Hg levels indicate lower than expected exposure.

Table 3. Total Hg levels (ppm) in fur from river otters collected in Maine during 2000 trapping season.

Site	Sex	Total Fur Hg (ppm, fw)	Total Brain Hg (ppm, ww)	Total Liver Hg (ppm, ww)
Boothbay-Wiley Pond	Male	5.0	0.08	0.24
Boothbay-Wiley Pond	Male	5.2	0.09	-
Boothbay Harbor - Lewis Cove	Female	18	0.37	2.61
Flagstaff - Turner	Female	33.7	0.60	4.01
St. John River-T15 R11	Male	28.1	0.57	2.57
St. John River-T15 R11	Female	22.7	0.64	4.69
St. John River-T 15 R11	Female	30.5	0.69	4.74
Wiscasset-Dresden Bog	Male	29.6	0.54	2.13

Fur Hg levels reflect the total body burden bioaccumulated over time, particularly for individuals with high exposure. Consequently the animal's age may be a confounding factor in interpreting fur Hg results. Mierle et al. (2000) found that Hg concentrations in fur changed with age. It increased during the first four years in Ontario otters, but then declined. However, fur Hg levels in the Ontario study did not exceed 15 ppm in known age otters, and it is likely the animals were able to demethylate their Hg body burden. In our study, several otters had relatively high fur Hg levels, therefore it is not clear if these animals would be able to demethylate their body burden. Blood Hg levels reflect recent dietary uptake and would help explain fur Hg concentrations.

Mink

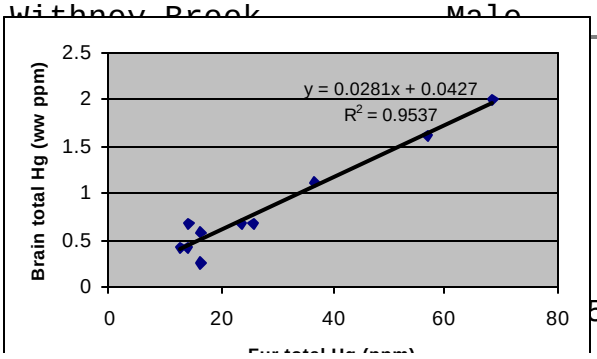
We analyzed 24 fur samples, and 10 brain and liver samples. Mink fur Hg concentrations ranged from 9.2 ppm on Adams Pond near Boothbay to 68.5 ppm on Dead River, Flagstaff Lake. Mink brain and liver Hg ranged from 0.26 (brain) and 0.77 (liver) to 2.0 (brain) and 8.0 ppm (liver) from Bog Brook in Hebron and Dead River respectively (Table 4).

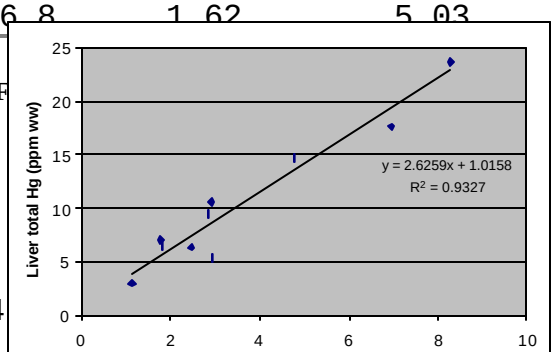
There does not appear to be a relationship between the size and sex of the animal and tissue Hg levels, however sample size is limiting. All liver samples were below the lethal levels of 25 ppm as reported by Wobeser et al. (1976), although extrapolating

findings from controlled lab experiments to wild populations are difficult. Additionally, liver total Hg levels are best used for historical comparisons. Recent work has shown the percentage of MeHg in the liver reaches an upper limit and does not correlate with total Hg levels (D. Evans, pers. com.). Therefore, evaluating the impact of Hg toxicity only using liver Hg levels is not recommended. There is a strong correlation between fur and brain (r^2=0.95) (Figure 1), and brain and liver (r^2=0.93) total Hg in mink (Figure 2).

Table 4. Total Hg levels (ppm) in mink fur, brain, and liver samples collected in Maine during 2000 trapping season.

Site	Sex	Total Fur Hg (ppm, fw)	Total Brain Hg (ppm, ww)	Total Liver Hg (ppm, ww)
Boothbay - Adams Pond	Male	9.2	0.15	0.049
Boothbay - Adams Pond	Female	13.4	0.26	0.60
Bog Brook	Male	16.3	0.26	0.77
Dresden Bog	Male	19.0	0.29	0.92
Flagstaff - Turner	Female	36.9	1.11	4.40
Flagstaff - Turner	Female	68.5	2.00	8.03
Boothbay - Cross River	Male	18.3	0.33	2.71
Boothbay - Lewis Cove	Male	22.9	0.52	1.82
Boothbay - Pleasant Cove	Male	11.2	0.13	1.06
Little Androscoggin River	Male	14.4	0.68	1.46
Little Androscoggin River	Female	25.9	0.68	2.61
Nezinscot River	Male	16.5	0.58	1.78
Nezinscot River	Male	10.5	0.20	1.17
Nezinscot River	Male	24.3	0.54	1.65
Nezinscot River	Male	32.6	1.20	4.79
Nezinscot River	Female	27.4	0.37	1.56
Nezinscot River	Male	17.6	0.48	1.49
Nezinscot River	Male	29.5	0.75	2.14
Sherman Lake	Male	27.8	0.49	1.25
St. John River	Female	23.7	0.67	3.01
St. John River	Female	34.1	1.06	6.29
St. John River	Male	14	0.42	2.06
West Branch Nezinscot River	Female	12.8	0.42	1.96
Withney Brook	Male	56.8	1.62	5.03





RECOMMENDATIONS

Because few trappers operate in the Flagstaff and Seboomook regions we recommend live trapping in these areas. Capturing a live animal permits blood sampling. Analysis of blood samples allow more meaningful comparisons among different sites and regions, because (1) blood Hg levels reflect a recent or short term Hg exposure of a piscivorous mammal and (2) should be independent of age. Because >95% of Hg in the blood is in the methyl form, measuring total Hg provides insight into the recent dietary uptake of MeHg. Collecting blood samples from recently killed animals is difficult because blood rapidly loses moisture after death; therefore, blood clots and whole blood Hg likely do not correlate (based on studies with loons). Conversely, much of the Hg in organs is inorganic. By sampling and analyzing fur and blood from live mammals we hope to establish a relationship between the two matrices that can be applied to future studies for Hg interpretation of live or dead animals. Because animals can be live-trapped in areas of low density, we avoid potential population impacts and provide a comparative template for other studies that cannot afford removing animals.

Live trapping also adds another matrix of Hg measurement that can be related to other compartments such as fur, liver, kidney, and brain. Each matrix provides different information. Mercury levels in fur are an indicator of long-term body burden and organs generally demethylate Hg and do not necessarily provide an accurate assessment on toxicity to the individual. There is now evidence that the brain can demethylate Hg (particularly in the otter, D. Evans pers. com.) so that compartment may not be helpful for chronic Hg loads. Sampling certain matrices, such as muscle or fur (since fur would likely reflect remobilization of MeHg in the muscle) can provide better insights into the lifetime body burden for the animal. This is crucial part of this investigation because the bioaccumulation rate of MeHg is one of the most important aspects of its toxicity to a population.

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Appendix 1. Otter carcasses provided by Maine trappers, 2001.

Date Trapped	Location	Sex	Body of Water	Body Weight (kg)	Length (cm)	Latitude	Longitude
11/10/00	Boothbay	Male	Wiley Pond	4500	60.5	43 54.072	69 38.218
11/10/00	Boothbay	Male	Wiley Pond	5800	69.5	43 54.072	69 38.218
11/5/00	Boothbay	Female	Lewis Cove	5200	65.5	43 51.266	69 36.793
11/23/00	Dead River	Female	Flagstaff - Turner	5400	68.5	45 8.226	70 10.188
12/1/00	T15 R11	Male	St. John River	5600	67.4	46 44.372	69 37.244
12/6/00	T16 R11	Female	St. John River	2800	60	46 45.644	69 34.734
11/15/00	T16 R11	Female	St. John River	4200	64.1	46 39.876	69 44.189
11/13/00	Wiscasset	Male	Dresden Bog	6700	87.5	44 5.720	69 41.154

Appendix 2. Mink carcasses provided by Maine trappers, 2001.

Date Trapped	Location	Sex	Body of Water	Body Weight (g)	Length (cm)	Latitude	Longitude
11/10/00	Boothbay	Male	Pleasent Cove	603.8	41	43 53.886	69 36.161
11/14/00	Boothbay	Female	Adams Pond	452.2	36	43 53.544	69 37.872
11/14/00	Boothbay	Male	Cross River	766.2	41.5	43 55.860	69 36.956
11/6/00	Boothbay	Male	Adams Pond	883.6	43	43 53.544	69 37.872
11/3/00	Boothbay	Male	Lewis Cove	940.3	44.8	43 51.266	69 36.793
11/6/00	Buckfield	Male	Nezinscot River	584	35	44 15.895	70 19.686
11/11/00	Buckfield	Male	Nezinscot River	635.5	38	44 15.895	70 19.686
11/9/00	Buckfield	Female	Nezinscot River	475.1	36.7	44 15.895	70 19.686
11/20/00	Dead River	Female	Flagstaff - Turner	422.1	36	45 8.226	70 10.188
11/20/00	Dead River	Female	Flagstaff - Turner	562.4	39.2	45 8.226	70 10.188
11/2/00	Dresden	Male	Dresden Bog	707.8	40.5	44 5.720	69 41.154
11/2/00	Hebron	Male	Bog Brook	769	37.5	44 13.651	70 20.754
11/7/00	Hebron	Female	L. Androscoggin R.	390	32.8	44 13.651	70 34.453
11/14/00	Newcastle	Male	Sherman Lake	828.7	41	44 0.351	69 35.589
11/2/00	Sumner	Female	Nezinscot R. - W. Br.	373	31	44 23.939	70 27.749
11/3/00	Sumner	Male	Nezinscot River	603	36	44 20.612	70 25.771
11/6/00	Sumner	Male	Nezinscot River	683	36.5	44 20.612	70 25.771
11/11/00	Sumner	Male	Nezinscot River	772.8	40.5	44 20.612	70 25.771
11/11/00	Sumner	Male	Nezinscot River	576.1	37.8	44 20.612	70 25.771
11/29/00	T12 R16	Female	St. John River	270.3	32	46 41.091	69 47.508
12/3/00	T12 R16	Female	St. John River	541.7	37	46 41.091	69 47.508
11/23/00	T12 R16	Male	St. John River	583.4	38	46 41.091	69 47.508
12/3/00	T5 R15	Male	Withney Brook	459.4	36.2	46 6.281	69 39.745
11/6/00	West Paris	Male	L. Androscoggin R.	410	32	44 19.404	70 34.453

SHARP-TAILED SPARROW STUDY

This report includes studies funded by SWAT for 2000 and 2001.

Mercury Exposure Profile for Sharp-tailed Sparrows Breeding in Coastal Maine Salt Marshes

(BRI 2002-11)

BioDiversity Research Institute is a Maine-based nonprofit research group dedicated to progressive environmental research and education that furthers global sustainability and conservation policies. Fundamental studies involve avian conservation and aquatic toxicology. We believe high trophic level piscivorous wildlife are vital indicators of aquatic integrity.

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Hg Exposure Profile for Sharp-tailed Sparrows Breeding in Coastal Maine Salt Marshes

(BRI 2002 – 11)

Submitted to:
Maine Department of Environmental Protection
Surface Water Ambient Toxic Monitoring Program
State House Station 17
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19 April, 2002

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INTRODUCTION

Sharp-tailed sparrows (*Ammodramus* spp.) inhabit wet meadows, marshes, and salt marshes of central and eastern North America. The taxonomy, distribution, and evolutionary history of this group has been debated for over a century. In 1995, based on morphological and genetic evidence, the American Ornithologists Union committee on classification and nomenclature voted to separate this single species with five known sub-species into two species: a northern species, *Ammodramus nelsoni*, with 3 sub-species (*A. n. nelsoni, A. n. alterus*, and *A. n subvirgatus*) and a southern species, *A. caudacutus* with two sub-species (*A. c. caudacutus and A. c. diverus*), limited to coastal wetlands. *A. n. subvirgatus* (hereafter Nelson's Sparrow) and *A. c. caudacutus* (hereafter Saltmarsh Sparrow) are sympatric in coastal Maine, New Hampshire, and the northeast shore of Massachusetts.

The biomagnification of mercury (Hg) in aquatic biota is well known (Watras and Huckabee 1994), however its expression in insectivorous birds is not well studied (see review in Thompson 1996). Terrestrial species have recently been selected to serve as potential bioindicators of contaminants including Tree Swallows (*Tachycineata bicolor*) for Hg exposure (Gerrard and St. Louis 2001) and organochlorines (Secord et al. 1999) and American Robins (*Turdus migratorius*) for lead (Johnson et al. 1999).

We believe sharp-tailed sparrows are an appropriate indicator of methylmercury availability in coastal marshes. Our two target species spend their entire life-cycle in salt marsh habitats of the Atlantic coast. Their small breeding territories afford an excellent opportunity to determine contaminant exposure for target marshes and even specific areas within a marsh. Because of increasing urbanization surrounding these habitats a better understanding of contaminant ecological impacts has been identified and is of national interest (Newman et al. 2002).

The objectives of this study were to 1) determine the extent of Hg exposure in two species of sharp-tailed sparrows in coastal Maine salt marshes, 2) compare blood Hg between Saltmarsh and Nelson's sparrows, and 3) determine if there were differences in Hg exposure among five Maine salt marshes.

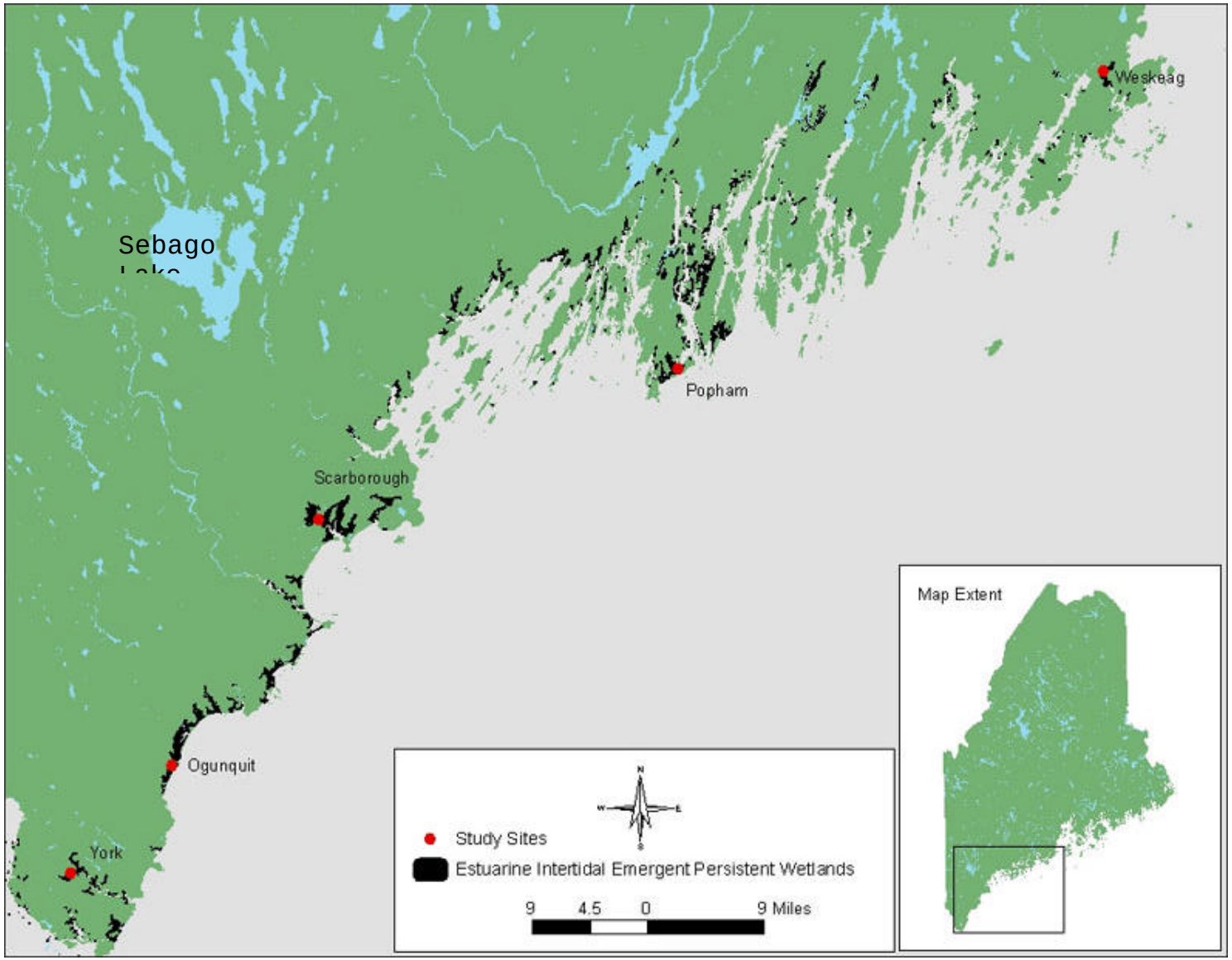
STUDY AREA & METHODS

We sampled sharp-tailed sparrows from 5 marshes along the Maine coast during the breeding seasons (15 June-1 August 2001) of 2000 and 2001 (Figure 1). We used mist nets to capture sparrows and attached a U.S. Fish and

Wildlife Service band and three color-bands to each individual. We used a wing cord ruler to measure unbended wing cord and dividers to measure tarsus length. We weighed all sparrows using a spring scale to the nearest 0.25 gm. We collected 30 µl – 50 µl of blood from the cutaneous ulnar vein for Hg contamination analysis using a micro-pipette. Micro-pipettes were stored in a test-tube and placed in a cooler immediately after collection. All samples were frozen on the day of collection and were maintained at $<25^{o}$ (F) until contamination analyses were conducted. Blood Hg levels are generally not compromised by body burden Hg levels during the breeding season (Evers et al. 1998).

We used independent *t* tests to determine differences in blood Hg levels between species and sex. If differences were significant between species or sex we then conducted further analyses separately. We used ANOVA with Tukey's post-hoc tests to determine if differences existed in blood Hg levels among the 5 sites. If there were differences among sites we then used ANOVA to determine if there were weight (g) or wing cord (mm) differences between high and low Hg level sites. All means are presented $\pm$ 1 SE.

Figure 1. Study sites with estuarine wetlands.



RESULTS

We captured and drew blood from 81 sharp-tailed sparrows (28 Nelson's and 54 Saltmarsh) in 5 marshes on the Maine coast (Table 1). Saltmarsh Sparrows (mean = 0.69 $\pm$ 0.03) had 41% greater blood Hg levels than Nelson's Sparrows (mean = 0.41 +/- 0.03) (t = 6.338, df = 79, P < 0.001, Figure 2). There was no difference in blood Hg levels between males and females for either species (Nelson's t = 1.69, df = 23, P = 0.171; Saltmarsh t = 0.848, df = 48, P = 0.401). We detected a difference in blood Hg levels among sites for both species (Nelson's F = 7.402, df = 4, P = 0.001; Saltmarsh F = 6.154, df = 4, P < 0.001, Figure 3 A and B). Popham beech and Ogunquit were highest in blood Hg for both species (Figure 3A and B). Sparrow weight and wing cord did not differ between high and low Hg level sites for either species (Nelson's weight F = 0.128, df = 1, P = 0.723, Nelson's wing cord F = 4.097, df =1, P = 0.053; Saltmarsh weight F = 1.219, df = 1, P = 0.275, Saltmarsh wing cord F = 1.542, df = 1, P = 0.220). There was a significant difference in weight between sparrow species.

Figure 2. Differences in blood Hg between Nelson’s Sparrow and Saltmarsh Sparrow. Saltmarsh Sparrows had significantly more blood Hg than Nelson’s Sparrow.

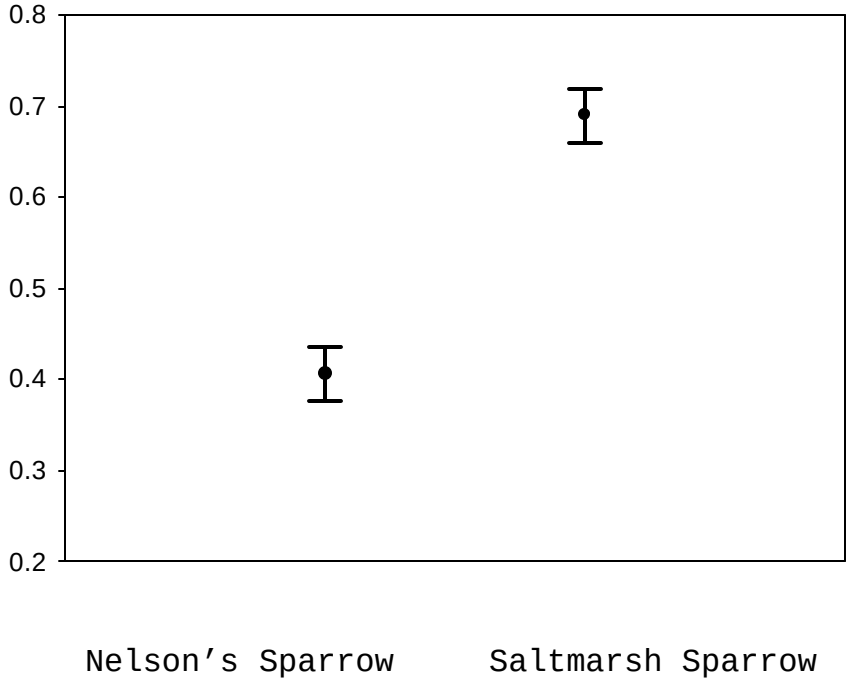


Figure 3. Differences in blood Hg between sites for A) Nelson's Sparrow and B) Saltmarsh Sparrow. Blood Hg levels were highest at Popham and Ogunquit for both species.

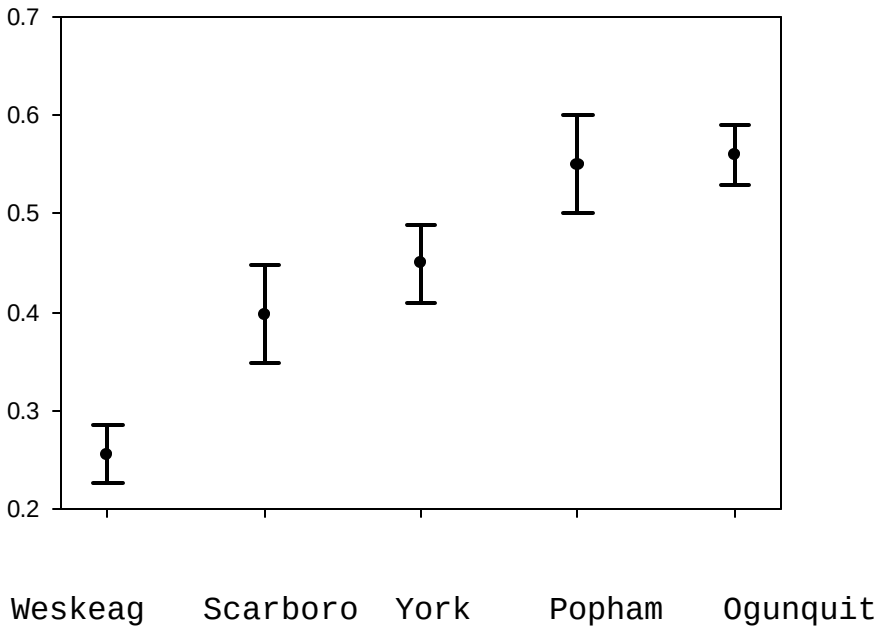


Table 1. Sampling locations, sample sizes and mean weight and wing cord for Saltmarsh and Nelson's Sharp-tailed Sparrows in coastal Maine (2000-2001).

		Saltmarsh Sharp-tailed Sparrow					Nelson's Sharp-tailed Sparrow				
Site	Lat / Long	Male	Female	Juvs	Mean Weight (g)	Mean Wing Cord (mm)	Males	Females	Juvs.	Mean Weight (g)	Mean Wing Cord (mm)
Weskeag	N 44 04.680 W 69 08.625	4	1	0	21.1 (0.6)	57.9 (2.2)	6	0	3	18.0 (0.8)	57.1 (1.1)
Popham	N 43 44.37 W 69 48.247	6	0	0	22.6 (0.5)	59.8 (0.8)	4	2	0	19.3 (0.7)	55.9 (1.6)
Scarborough	N 43 33.90 W 70 21.67	16	6	0	20.3 (1.6)	57.2 (1.3)	6	2	0	17.7 (1.7)	57.3 (2.1)
Ogunquit	N 43 17.02 W 70 34.92	7	4	0	20.3 (1.6)	57.6 (2.7)	3	0	0	18.3 (1.5)	56.8 (1.0)

York	N 43 09.64 W 70 44.01	6	1	3	19.2 (1.9)	56.9 (2.1)	2	0	0	18.4 (0.9)	57.0 (1.4)
TOTAL		39	12	3	20.7 +/- 1.3	57.9 +/- 1.1	21	4	3	18.3 +/- 0.6	56.8 +/- 0.5

DISCUSSION

We found nearly twice the Hg blood levels in Saltmarsh Sparrows than we did in Nelson's Sparrows at all five sites. This pattern was not predicted as both species spend their entire life-cycle in salt marsh habitat, presumably exposed to the same levels of contamination. Differential prey selection by sparrows could explain differences in the observed blood Hg levels. If Saltmarsh Sparrows, which are larger and have larger beaks, selected carnivorous prey while the smaller Neslon's Sparrows selected herbivorous prey, then we would expect to see higher levels of blood Hg in Saltmarsh Sparrows. Because these sparrows were recently split into two separate species (1995), little is known about dietary differences between them that may explain differences in blood Hg levels we found during this study.

We also found differences among the five salt marshes we sampled; indicating that blood Hg levels in sharp-tailed sparrows may be used as an index to Hg contamination in the salt marshes. This finding was supported by the similar pattern in Hg levels within each species across the five sites. For both species, blood Hg levels were highest in Popham and Ogunquit, intermediate at York, and lowest in Scarborough and Weskeag. This consistency in blood Hg levels in the two species across the five sites indicates that theses sparrows may be potential indicators of salt marsh and estuarine Hg contamination.

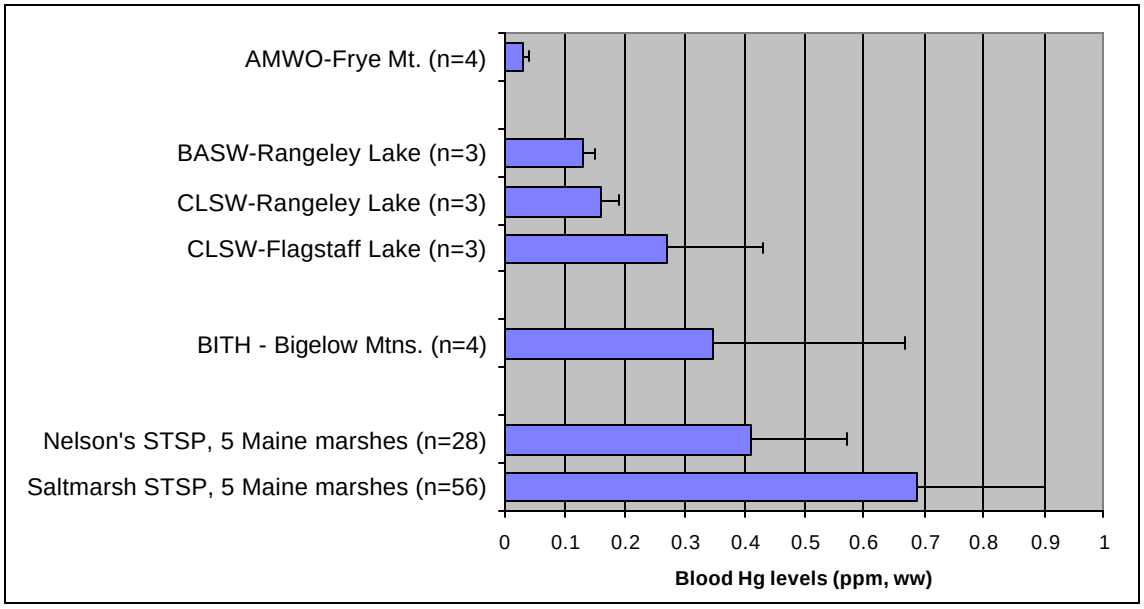
Comparing our sparrow blood Hg levels with other related species is difficult. The handful of terrestrial bird Hg studies are not based on blood, rather their assessments use whole body analysis and/or organs (i.e., lethal sampling). However, our non-lethal sampling strategy for this project is comparable with other such collection efforts with insectivorous birds in Maine. BioDiversity Research Institute staff have sampled terrestrial birds including American Woodcock (*Scolopax minor*) (AMWO), Barn Swallow (*Hirundo rustica*) (BASW), Cliff Swallow (Petrochelidon pyrrhonota) (CLSW), and Bicknell’s Thrush (Catharus bicknelli) (BITH) (Figure 4).

The sampling efforts with the swallows are particularly informative as a reference for Hg exposure. Swallows were sampled from two lakes that have thorough biotic Hg risk assessments based on fish and the Common Loon (*Gavia immer*) (Evers et al. 2002). Because swallow sample sizes are minimal statistical comparisons were not attempted. Barn and Cliff Swallows from Rangeley Lake, a low Hg risk system, had mean blood Hg levels considerably less than those found from both sharp-tailed sparrow species in each of the five marshes. Assuming a

relationship exists between fish Hg levels and associated emerging insects, reference blood Hg levels for insectivorous birds are possibly less than 0.20 ppm (ww). Flagstaff Lake is well known for its elevated biotic Hg levels (Evers et al. 2002). Cliff Swallow blood Hg levels tended to be less on Flagstaff Lake than sharp-tailed sparrow blood Hg levels.

Further efforts with swallow species in areas with known biotic Hg assessments as well as at the sharp-tailed sparrow locations will provide further context for assessing hazards related to Hg levels in coastal Maine's salt marshes.

Figure 4. Blood Hg levels in selected insectivorous birds in New England



RECOMMENDATIONS

1. Determine Hg exposure for sharp-tailed sparrows in other Maine coastal marshes with large breeding populations;
2. Determine Hg exposure for Tree Swallows with breeding territories in coastal marshes with sharp-tailed sparrows at some locations for comparative purposes;
3. Determine Hg exposure for swallow species with breeding territories in areas with known biotic Hg levels;
4. Determine prey base of sharp-tailed sparrows and analyze prey items for Hg;
5. Measure levels of other contaminants including polychlorinated biphenyls in sharp-tailed sparrows.

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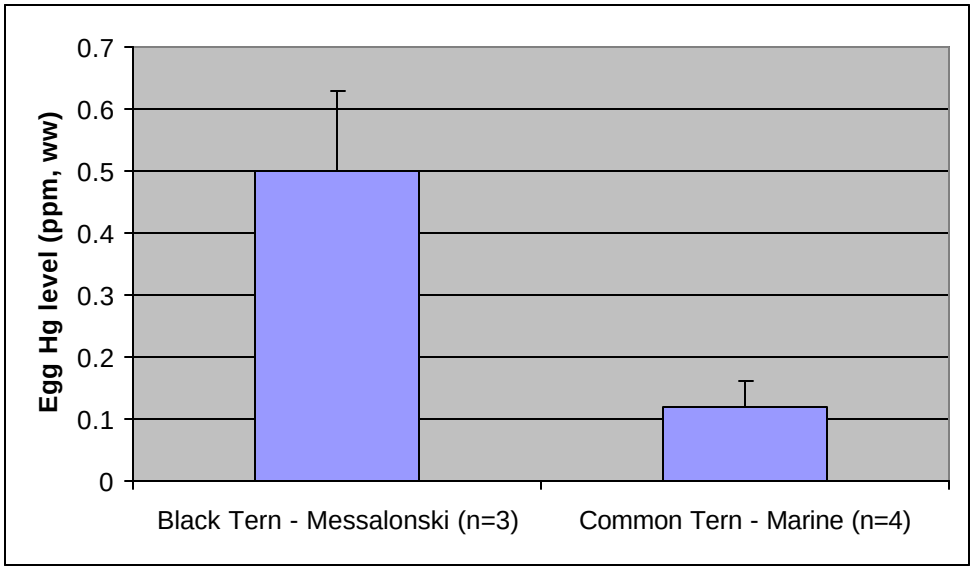
Watras, C. J. and J. W. Huckabee. 1994. Mercury pollution: Integration and synthesis. Lewis Publ., Boca Raton, Florida.

Black Terns

The Black Tern (*Chlidonias niger*) is a endangered species in Maine. It is a colonial nesting species using open emergent wetlands. In June 2001, three abandoned eggs were collected at one of the largest colonies in the state at Messalonskee Lake. These eggs were analyzed at Texas A&M Trace Element Research Lab for mercury (Hg). The mean egg Hg level was 0.50 +/- 0.13 ppm (wet weight) (Figure 1). Compared to Common Tern (*Sterna hirundo*) egg Hg levels from Stratton Island (a marine nesting site), Black Tern egg Hg levels were over four times greater. A similar pattern between marine and freshwater habitats has been demonstrated in the Belted Kingfisher (*Ceryle alcyon*) as well.

Although some evidence indicates impacts to bird reproduction and health occur when egg Hg levels exceed 0.50 ppm (ww), egg Hg levels in piscivorous birds likely need to approach 1.0 ppm (ww) before impacts occur. However, because this species is listed as endangered in Maine, further collections of tissues for Hg analysis is prudent.

Figure 1. Egg mercury levels for the Black Tern in Maine.



2.5

MERCURY TRENDS
(1999)

Temporal Changes in Fish Mercury Concentration in Maine Lakes

Final Report

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Maine Department of Environmental Protection

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Introduction

Previous studies have demonstrated that many lakes in Maine contain fish with high mercury concentrations, commonly above 1 µg/g. We hypothesize that these levels are not natural but rather result from atmospheric transport and deposition of mercury from urban/industrial areas to the south and west of Maine to these waters. Sediment core studies from several lakes and bogs in Maine indicate that mercury deposition to sediment was relatively low and constant prior to about 1900, and then increased greatly to levels 2 to 5 times higher by 1970. At some locations, primarily in southern and coastal Maine, sediment mercury deposition then declines to the present. At other locations, primarily in northern and inland Maine, mercury deposition to sediment continues to increase to the present. One explanation for this is that mercury emissions from large point sources (coal-fired electric generating stations, solid waste incinerators) were reduced by implementation of the Clean Air Act in 1972, resulting in reduced mercury deposition from these sources. In more remote areas (northern and inland Maine) mercury deposition may result more from the global reservoir of mercury, which will likely decline very slowly. Further, the balance of precipitation between rain and snow is markedly different away from the coast, perhaps favoring less mercury deposition at inland sites. Lastly, air masses impinging on the coast during precipitation typically have storm tracks up the eastern U. S. seaboard, whereas northern Maine is more under the influence of Canadian air masses. At present there is no information to indicate if fish mercury concentration in Maine lakes is changing over time.

The earliest reliable fish mercury data in Maine are from a study of several species of fish from three lakes in the Allagash region that were sampled in 1978 (Akielaszek and Haines 1981). Several other studies, conducted from 1982-1984, determined mercury in brook trout (*Salvelinus fontinalis*) and white suckers (*Catostomus commersoni*) from nine small lakes in central and southern Maine (Haines *et al.* 1987; Hamilton and Haines1989; Haines and Brumbaugh 1994). Several of these lakes were cored for historical sediment analysis (stable lead isotopes, trace metals not including mercury, and diatoms) at the time fish were collected for mercury analysis.

The objective of this study was to determine if there has been any consistent, systematic change in fish mercury concentration in Maine lakes over the period from 1978 to the present. To accomplish this, we screened the list of lakes for which historical fish mercury data were available and selected candidate lakes that represented the various species of fish and physical and chemical conditions available. Fish of different food habits or life history may respond differently to changes in mercury supply or availability. Lakes of different size and location may have different mercury inputs, retention, or methylation rate, which may in turn affect fish mercury content. We also collected a sediment core from one lake, so that historical changes in sediment mercury accumulation could be compared to changes in fish mercury over the same time period.

Methods

Several species of fish from a number of Maine lakes were analyzed for mercury in the late 1970s to early 1980s (Akielaszek and Haines 1981; Haines and Brumbaugh 1994; Haines *et al*. 1987; Hamilton and Haines 1989). We screened this data set to identify suitable lakes to be resurveyed. Selection criteria included: no major change in

access, watershed development, or fish management practices; relative ease of access; presence of robust fish population to be resampled; and a time interval between sample periods of at least 15 years. Seven lakes were selected to be resurveyed. They ranged in size from 2 to more than 2000 ha and included three species of fish: lake trout (*Salvelinus namaycush*, long-lived, carnivorous), brook trout (short-lived, omnivorous), and white sucker (long-lived, bottom-feeder). For the original survey, detailed field and laboratory records were available from data archives. These records described collection methods, locations, and times, fish species, number, and size collected, and laboratory analytical methods and quality assurance used to determine mercury content.

For the present survey, fish were collected by the same methods and in the same locations as in the original survey. The primary collection method was gill netting with Swedish experimental (graded mesh) nets, except that lake trout from Big Eagle Lake were collected by angling in 2001 and by both gill net and angling in 1978. The collection goal was 10 fish per species within a similar size range to those previously analyzed. Fish were placed in plastic bags, on ice, and returned to the laboratory as soon as practical. In the laboratory, fish were weighed (nearest g), measured (total length, mm), individually wrapped in plastic bags, and frozen. For analysis, the fish were thawed, and processed following the same procedure used to produce the prior mercury data. In the case of Big Eagle, Cliff, and St. Froid lakes (lake trout and brook trout), a strip of dorsal muscle tissue extending from just behind the head to the tail was removed from each fish, skinned, and homogenized. In the case of Mountain Pond (Coburn) and Mountain Pond (Rangeley) (brook trout), the whole fish was homogenized. And in the case of Green Lake and Horseshoe Pond (white sucker), the fish were first eviscerated and then homogenized.

In the original surveys, the fish tissue homogenate was wet-digested in acid, the precise nature of which varied with the survey, and analyzed by atomic absorption spectrometry, the standard method at the time. The moisture content of the homogenate was determined in some studies but not in others. In the Akielaszek and Haines (1981) study, the fish were analyzed at the University of Maine. For the other studies, the fish were analyzed at the National Fisheries Contaminant Research Center (now the Environmental Research Center) in Columbia, Missouri. In all cases a full quality assurance program was followed to ensure data precision and accuracy, including reagent and digestion blanks, sample duplicates, matrix spikes, and certified reference materials. For the recent samples, moisture content was determined on a subsample of the homogenate by use of a moisture balance. Total mercury was determined by digestion of approximately 0.5 g of homogenate in a 1:1 mixture of concentrated nitric acid and hydrogen peroxide with microwave energy, and analysis by automated atomic fluorescence spectrometry. The quality assurance program included the same components as that in the original surveys. Although these methods differed from those used in the original survey, the quality assurance programs in effect demonstrate that the results obtained were within accepted standards of accuracy and precision, and thus are comparable.

A sediment core was retrieved from Cliff Lake from a point just west of the deep hole, in water 58 feet deep, using a stationary piston coring head and acrylic core barrel with a 10 cm diameter. The 45 cm core was extruded and sectioned in the field as follows: 0 to 1 cm, 1 to 10 cm depth in 0.5 cm intervals, 10-30 cm in 1.0 cm intervals,

and 30-42 cm in 2.0 cm intervals. Sediment was stored cold in Whirl-Pak™ bags prior to processing in the laboratory. Water concentration and organic matter concentration were determined for calculations associated with ^{210}Pb dating of the sediment. Water concentration was determined by heating the sediment at 35^{o}C to constant weight. Previous experience has demonstrated that no mercury is lost when heating at temperatures below 70 ^{o}C. Organic matter was determined by ramped heating of an aliquot of dried sample to 550^{o}C, for 3 hours (constant weight). ^{210}Pb gamma-ray activity was determined using the 46.52 keV emission. We used a Canberra germanium well detector (1 by 4 cm) with 22.5% efficiency for ^{60}Co. Data were processed using *GammaTrac* software (Oxford Instruments). Dried sediment in capped 1 by 4 cm polyethylene vial was counted for 43,200 to 259,200 seconds. Data were analyzed by Compton continuum subtraction of the peaks. Calibration of the detector was done using U.S. EPA National Exposure Research Laboratory aqueous standards (^{210}Pb, ^{241}Am, ^{226}Ra, ^{137}Cs, and ^{60}Co) in the same geometry as the sediment samples. The $^{210}Pb_u$(unsupported) activity was estimated by subtracting the constant background ^{210}Pb activity, deep in the core, from total $^{210}Pb_t$. The (integrated) $\Sigma^{210}Pb_u$ ($Bq^{210}Pb_u/cm^2$/core), necessary for dating, also assesses sediment focusing. Calculation of age of interval mid-points was based on the CRS model of Appleby and Oldfield (1983). We used linear interpolation between interval mid-point ages to determine ages of interval boundaries and thus the years represented by an interval.

This analytical method concurrently may measure other radionuclides of interest, including ^{134}Cs, ^{137}Cs, ^{241}Am, and ^{40}K. The first three have been commonly used to corroborate the ^{210}Pb method, which gives continuous dating of the sediment with depth. The age calculation was based on the CRS (**c**onstant **r**ate of **s**upply) model of Appleby and Oldfield (1978); however, we also tested the CIC model (**c**onstant **i**nitial **c**oncentration).

Separate aliquots of sediment intervals were analyzed for total Hg by atomic fluorescence spectrometry. QA/QC measures included: analysis of duplicate aliquots, multiple measurements of the sample aliquot, analysis of reagent blanks, and analysis of standard reference materials. Data from the chemical and radionuclide analyses were used to generate the relationship between flux of the metals to the sediment versus chronological time. This output can be compared to the history of the drainage basin to establish causal relationships between chemical changes and anthropogenic activities, and to the mercury concentration in fish at known time intervals.

The net accumulation rate for total Hg (ng Hg/cm^2/yr) equals:

Hg_T = [(mass of sediment/interval/cm^2)(concentration of Hg in interval)]/(years/interval) (1)

This total flux is composed of three components. (1) The natural background flux of Hg, Hg_B. Commonly, variations in % organic matter cause variations in Hg concentration. However, LOI does not vary appreciably in the background portion of the core and therefore we have made no correction for this effect. We use pre-1880 sediment for this estimate. (2) Variations in the gross sedimentation rate caused by human activities in the watershed cause variations in the flux of Hg, Hg_V. This variation is estimated from pre-1880 sediment using the ratio (sedimentation rate for any sediment interval [g/m^2/yr])/(pre-1880 sedimentation rate [g/m^2/yr]). (3) Variations in deposition of

anthropogenic Hg directly to the lake and from leaching of Hg from the watershed to the lake, Hg_A. Thus:

$$\text{Total Hg (ng Hg/cm}^2\text{/yr)} = Hg_B + Hg_V + Hg_A \qquad (2)$$

Results

The seven lakes that were resurveyed ranged in surface area from 2 to 2153 ha, in maximum depth from 3 to 38 m, in elevation from 139 to 871 m, in watershed area from 13 to 104,600 ha, and in acid neutralizing capacity from 6 to 520 µeq/L (Table 1). The locations ranged from central Hancock County to northern Aroostook County (Figure 1). For two of the lakes resurveyed, only brook trout were collected, for an additional two only lake trout were collected, for one lake both brook trout and lake trout were collected, and for two lakes only white suckers were collected (Table 2). The fish collected in the recent survey were generally similar in size to those collected previously (Figure 2), and linear regressions of weight on length for each collection year for each species and lake were generally not different for slope or intercept (test for common regression, Freese 1967). However, the recent collections of white suckers from Green Lake and brook trout from Mountain Pond (Coburn) were significantly different for intercept but not slope, indicating that the relation between weight and length is similar, but that the recently-collected fish are significantly lighter in weight than fish of the same length in the previous survey.

Fish mercury concentration generally increased as fish size increased (Figure 3), although the regressions were not always statistically significant, especially for the collections with smaller sample sizes. Regressions of fish mercury concentration on either length or weight were similar, so tests of difference in fish mercury concentration among years were adjusted for difference in fish size by using length as the covariate. For brook trout, fish mercury concentration was significantly higher in the recent collection for all three lakes (Table 3), even in Mountain Pond (Coburn), where the fish in the recent collection were significantly smaller than in the earlier collection. Fish mercury concentration also increased over time for lake trout, but the difference was statistically significant only for Cliff Lake (Table 3). For white sucker, fish from both Green Lake and Horseshoe Pond were significantly lower in mercury concentration for the recent collection (Table 3). Although for Green Lake the fish in the recent collection were also smaller than in the earlier collection, which could contribute to the lower mercury concentration in the fish in the recent collection, this was not the case for Horseshoe Pond. These differences are large and statistically significant whether adjusted for fish size or not.

Dating of the sediment is based on a 12-point analysis of ^{210}Pb. There is a suggestion of some mixing of sediment or accelerated sediment accumulation in the upper 5-7 cm, indicated by a slightly less steep downward decrease in the activity of ^{210}Pb. Application of the CRS dating model yields a nearly linear decrease in age with increasing depth. The maximum activity of ^{137}Cs occurs in the sediment interval dated at 1958 (spanning 1956-1960), reasonably consistent with the known maximum atmospheric deposition caused by atmospheric nuclear bomb testing in 1963. ^{137}Cs occurs considerably deeper than sediment dated as 1963 and also persists to the surface. The former is likely caused by some downward bioturbation of early 1960s sediment as well as downward diffusion of ionic Cs. The latter is caused by upward bioturbation and

diffusion from the maximum sediment activity as well as post 1960s wash-in from the watershed. ^{241}Am was too low in activity for use as a second independent estimate of the depth of 1963. We judge the chronological control to be very good in the last 50 years. The counting error at about 1900 yields an age estimate error of ±15 yr.

The concentration of Hg in sediment ranges from background values of approximately 185 µg/g prior to 1800 (poorly estimated age) to a maximum of about 335 µg/g in the 1980-1995 period. Concentration increased slightly in the first half of the 19th century and then linearly with time from 1850 to about 1915; it decreased slightly to about 1950 and then increased to about 300± until the time of coring in 2000 (Figure 4). The early onset of the increase (1850), while small in absolute value, is persistent and the earliest clear increase we have observed in Maine. The overall increase is modest, never reaching twice background, and is comparable to other remote lakes in northern Maine. The total flux of Hg to the coring site ranges from slightly over 2 ng/cm^2/yr prior to 1850 to nearly 10 ng/cm^2/yr (averaging the two widely disparate samples at the top of the core (Figure 5). The increase is relatively constant with little indication of a persistent change in slope. The error in analysis of the ^{210}Pb for any particular measurement is probably the poorest in precision of any parameter. These errors translate through the calculations for age of sediment intervals, length of time represented by intervals, and thus the flux of Hg. The running accumulation rate for total Hg based on 3-adjacent intervals yields a smooth increase in the total Hg flux over the last 130 years. The accumulation rate for the anthropogenic component of the total Hg flux (correcting for background and varying sediment accumulation rate) was determined with the assumption that background is the flux prior to 1875 (i.e., the mean anthropogenic flux prior to 1875 is 0 ng/cm^2/yr). Thus the flux increases from 0 to approximately 3 ng/cm^2/yr, using a 3-point running average to reduce the section to section variation (Figure 6). The increase is relatively constant, possibly with a plateau from 1920 to 1950. This is a typical profile for lakes in northern Maine.

The anthropogenic mercury accumulation rate during the time period for which fish mercury data are also available increases consistently (Figure 7), with the accumulation rate in 2000 being about 35% higher than in 1978. Fish mercury concentration in this lake has also increased over this time interval, by a factor of 2.5 for brook trout and 1.8 for lake trout.

Discussion

The lakes surveyed for this project were relatively undisturbed by human activity. There are no known local sources of mercury to these lakes, so atmospheric deposition is presumed to be the major source. At most there are a few seasonal roads and dwellings in the watersheds, and timber harvesting is the major watershed disturbance. Management of the fish populations has generally not changed, although brook trout were stocked into Horseshoe Pond annually from 1997 to 1999 (no trout were caught in the gill nets). There was evidence that angling pressure had increased significantly at Mountain Pond (Rangeley) between the two collection dates. At the time of the recent survey there was a very well-used ATV trail leading to the lake, and a large number of boats and canoes on the shore of the lake. At the time of the earlier survey there was only a little-used hiking trail to the lake, and a very small number of boats.

The same species and sizes of fish that were collected in the original survey were obtained in the recent survey. Fish were not aged because they had not been aged in the

original survey, so changes in size-age relationships could not be determined. However, in two cases the length-weight regressions indicated that fish in the recent survey were lighter than fish of the same length in the original survey. Inasmuch as there had been no change in human activity in the watershed between collections, the change in size probably resulted from a change in population density or food supply, which then resulted in a decline in growth rate. The declines were small and probably had little effect on the mercury concentration results.

Although the amount of mercury on earth has not changed since the planet was formed, it is generally accepted that human activities have increased the amount of mercury cycling through the biosphere and that this increase is reflected in the mercury content of biota (USEPA 1997). Our results document an increase in mercury input to Maine lakes, as shown by the sedimentary record, and a concomitant increase in mercury concentration in some lake-dwelling fish. Mercury concentration in brook trout increased on average at the rate of 4.2 ng/g/yr, and in lake trout at 3.3 ng/g/yr (although for two out of the three populations the increase was not statistically significant). These findings are similar to those reported elsewhere. However, mercury concentration of white suckers decreased on average at the rate of 9.8 ng/g/yr. There were no previous studies of change in mercury content of this species in the literature, so it is unknown if this is a typical response for this species.

In Minnesota, Swain and Helwig (1989) surveyed the change in mercury concentration of northern pike (*Esox lucius*) in 9 lakes between 1970 and 1988, with time between sampling of 5 to 16 years, and found a mean increase of 17 ng/g/yr, from a mean concentration of 360 ng/g to 470 ng/g over an average time interval of 7.2 years. Håkanson (1991) determined mercury concentration in northern pike in 73 lakes in Sweden at various time intervals. Change in fish mercury concentration with time ranged from –135 ng/g/yr to +180 ng/g/yr, with 11% of the lakes having a decreasing trend, 45% having no change, and 44% having an increasing trend. Fabris *et al*. (1999) compared mercury concentration in black bream (*Acanthopagrus butcheri*) from brackish lakes in southeastern Australia with values obtained in another survey 18 years previously. Mercury concentration increased from 110 to 180 ng/g (least square mean size adjusted), for a rate of 3.9 ng/g/yr.

Several authors have compared mercury concentration in museum fish specimens to that in recently collected fish from the same location. Amrhein and Geis (2001) compared muscle mercury concentration in yellow perch (*Perca flavescens*) collected from five lakes in northern Wisconsin 1927-28 with that in 1988. There was a significant increase in mercury concentration in fish from two lakes (from 220 to 760 ng/g dry weight in one lake and from 370 ng/g to 670 ng/g in another), no change in two lakes, and a significant decrease in one lake (from 530 ng/g dry weight to 260 ng/g). There were some differences in size and age of fish between the two collection dates, which were not controlled in the analysis, making these results somewhat questionable. Swain and Helwig (1989) determined mercury concentration in northern pike and walleye (*Stizostedion vitreum*) collected in 1935-36 from six lakes in northern Minnesota and compared them with measurements for the same species and size of fish in 1983-86. Fish mercury concentration increased over time in four lakes, at rates ranging from 2 to 9 ng/g/yr, did not change in one lake, and decreased in one lake at a rate of 2 ng/g/yr. In contrast, Kelly *et al*. (1975) found little difference in mercury concentration of walleyes

in Michigan when museum specimens collected between 1865 and 1936 were compared with specimens collected in 1971 from the same lakes. They noted that variation in fish mercury concentration was greater among locations than between collection periods. There are concerns with mercury results from analysis of museum specimens, chiefly regarding the loss of moisture and lipids to the storage medium (normally alcohol), but the above studies controlled for most of these problems, and the findings are consistent with other studies.

The only cases where fish mercury concentration declined over time were for white suckers. In both Green Lake and Horseshoe Pond, fish mercury concentration declined by a similar percentage: 34% in Green Lake and 28% in Horseshoe Pond. The rate of decrease was -5.8 ng/g/yr for Green Lake and -13.7 ng/g/yr for Horseshoe Pond. In the two studies cited above for which decreases in fish mercury concentration over time were reported, the rates were 2 ng/g/yr for northern pike (Swain and Helwig 1989) and 4.4 ng/g/yr for yellow perch (Amrhein and Geis 2001). A recent study of mercury concentration changes over time in eastern mosquitofish (*Gambusia holbrooki*) in south Florida (Stober *et al*. 2001) found that mean mercury concentration declined from 163 ng/g in 1995-96 to 123 ng/g in 1999, a rate of -13.3 ng/g/yr. The authors believe that mercury emissions to the atmosphere from waste incinerators declined in this area during this time period, and water concentrations of total mercury also declined during the wet season, from 1.96 ng/L in 1995-96 to 1.43 ng/L in 1999 (water mercury concentrations increased during the dry season over this time interval). The Everglades ecosystem is very shallow and mosquitofish are very small and short-lived, so this system may respond rapidly to reductions in mercury inputs. However, the authors also state that other studies have documented declines in mercury concentration of largemouth bass (*Micropterus salmoides*) of 66% since 1990, and in great egret nestling feathers of 50% from 1994 to 2000, so mercury may be declining quite rapidly in the entire system (the references cited for this work are abstracts from workshop proceedings, which are not available for inspection).

The earlier collections of white suckers were analyzed for mercury at the National Fisheries Contaminant Research Center (now the Columbia Environmental Research Center) in Columbia, Missouri. However, full quality assurance procedures were followed by the Columbia laboratory, and the mercury results obtained are believed to be at least as accurate as the analyses that were performed at the University of Maine. Further, the brook trout from Mountain Pond (Coburn) and Mountain Pond (Rangeley) were also analyzed at the Columbia laboratory in the initial survey, and fish mercury increased over time in these cases. The mercury concentration of white suckers is in the same range as that of brook trout, so there is no reason to suspect that there is any analytical bias in the mercury results. White suckers are not stocked into lakes, and are not harvested by anglers, unlike brook trout and lake trout. It is not known if these differences could contribute to the different response in mercury accumulation by these fish.

The sediment core was of high quality with little disturbance of the sediment-water interface observed during coring and sectioning. The profile of water and LOI down-core were typical for lake sediment in Maine and indicated no major disturbance of the sedimentary regime, such as slumping or a dramatic change in the accumulation rate of sediment. The ^{210}Pb chronology is good and consistent with the ^{137}Cs data. Although

there is evidence of an increase in Hg concentration in the second half of the 19th century, and presumably an increase in the accumulation rate of Hg, the correction for this cannot be made because the limit of the dating is about 1850. The corrections would be slight and would not materially change the anthropogenic flux or trends. Sedimentation rate did increase rather abruptly about 1935+/- by nearly 100%. This may be related to a cycle of forest cutting in the catchment, with associated land scarification and erosion. This increase is factored into the results of Figures 5 and 6. Some unknown proportion of Hg associated with the increased sedimentation rate was derived from anthropogenic Hg deposited initially on the catchment, rather than directly on the lake. Thus Figures 5 and 6 combine anthropogenic Hg derived from three routes: direct deposition from the atmosphere to the lake, leakage of Hg from the terrestrial part of the catchment, and mechanical erosion of previously stored Hg.

Cliff Lake is relatively deep for Maine lakes and has a pronounced deep area. The result of this is a slight focusing of sediment at the coring site. This is most clearly seen in the integrated unsupported ^{210}Pb derived from atmospheric deposition. Approximately 17 pCi/cm^2 ($6.2*10^{-1}$ bq/cm^2) exists at the coring site, nearly 50% higher than is delivered by precipitation. This suggests that the flux of Hg is probably overestimated, but trends are unaffected. The observation that Hg accumulation (total and anthropogenic) increases to the present (2000) cannot be interpreted as an increase in deposition. Retention of recently deposited Hg in the catchment may be in the range of 90 to 95%. Consequently, a change in Hg deposition from the atmosphere may take 30-40 years to reach steady state with export of Hg from the watershed to the lake. Independent paleolimnological and soil evidence (Evans *et al*. 2000; Norton *et al*. 1997) indicates that Hg deposition from the atmosphere probably peaked in Maine in the 1970s. However, the most recent 30 years of sediment Hg accumulation in Cliff Lake (Figure 7) do not reflect such a decline.

The Mercury Deposition Network (MDN) has conducted measurements of mercury content of weekly precipitation since 1996 at the station in Acadia National Park, and since 1998 at the station in Greenville. During this rather short time period, mean annual mercury concentration has been relatively constant at both locations (Figure 8). The mean annual mercury deposition rate has been constant at Acadia National Park, but may have declined slightly at Greenville (Figure 9). Note, however, that the data point for 2001 is for the first six months of the year only, and that 2001 has been a drought year in Maine. The presumed decrease in atmospheric deposition may take a few more years to be detectable in precipitation measurements, and a few decades to be reflected in a measurable reduction in export of Hg from the catchment to the lake. Any paleolimnological record typically lags atmospheric changes and is smeared through time by bioturbation and as a consequence of the time necessary for fine-grained sediment to reach the lake and be mechanically winnowed into deeper water. Sediment deposited at the deep hole is a mixture of modern and reworked older sediment. In spite of these reservations, it is clear that the supply of Hg to the coring location has not diminished over the last 30 years. The sedimentary mercury record for this lake is thus in general agreement with the fish mercury record.

Summary and Conclusions

The available scientific evidence demonstrates that human activity has increased the amount of mercury cycling through the atmosphere of the earth, and being deposited to the earth's surface. Although the increase in atmospheric mercury may have halted recently, or even declined at some locations, the sediment core from Cliff Lake demonstrates that the mercury input to this remote lake began to increase above background in the mid-1800s and that this increase continues to the present. The fish mercury record is in general agreement with the sedimentary record, increasing over time at six of the eight locations surveyed, although the increase was statistically significant at only four of the six locations. The only cases where fish mercury concentration declined over time were for white suckers from two lakes. The results of this study are generally consistent with the literature, where increases in fish mercury concentration over recent time have been found for the majority of cases investigated. Fish mercury decreases have been reported for other species in other lakes in a minority of cases, and may reflect normal lake-to-lake variability. It is not known if there is some unique feature related to white suckers as a species or the lakes from which they were collected that could account for the observed decline in mercury in this species.

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Table 1. Physical and chemical characteristics of the lakes resurveyed.

MIDAS	Name	Township	County	Surface Area (ha)	Max Depth (m)	Elevation (m)	Watershed Area (ha)	ANC, µeq/L
0160	Mountain Pd. (Coburn)	Johnson Mtn	Somerset	2	3	871	13	45
1610	St. Froid Lk.	Winterville Plt.	Aroostook	972	35	185	104596	504
2780	Cliff Lk.	T9 R12 WELS	Piscataquis	228	20	307	2436	520
2858	Eagle Lk. (Big)	Eagle Lake	Piscataquis	2153	38	294	44013	286
3540	Mountain Pd. (Rangeley)	Rangeley Plt.	Franklin	17	12	733	145	13
4790	Green Lk. (#2)	T35 MD	Hancock	26	4	139	114	110
0858	Horseshoe Pd.	Willimantic	Piscataquis	26	4	162	194	6

Table 2. Size-adjusted mean fish mercury concentration by species and lake for the early and recent collection dates. Mercury values are least square means, ng/g wet weight, using length as the covariate.

Species	Lake	Year	Hg, ng/g	p
Brook trout	Cliff Lake	1978	73	0.01
	Cliff Lake	2000	182	
	Mountain (Coburn)	1979	25	0.0004
	Mountain (Coburn)	2001	74	
	Mountain (Rangeley)	1979	95	0.0005
	Mountain (Rangeley)	2000	218	
Lake trout	Big Eagle Lake	1978	531	0.44
	Big Eagle Lake	2001	594	
	Cliff Lake	1978	187	0.006
	Cliff Lake	2000	341	
	St. Froid Lake	1978	558	0.87
	St. Froid Lake	2000	569	
White sucker	Green Lake	1984	140	0.0001
	Green Lake	2000	47	
	Horseshoe Pond	1983	342	0.0031
	Horseshoe Pond	2001	96	

Figure 1. Map of the state of Maine showing the location of lakes surveyed, and the location of the Mercury Deposition Network stations in Greenville and Acadia National Park.

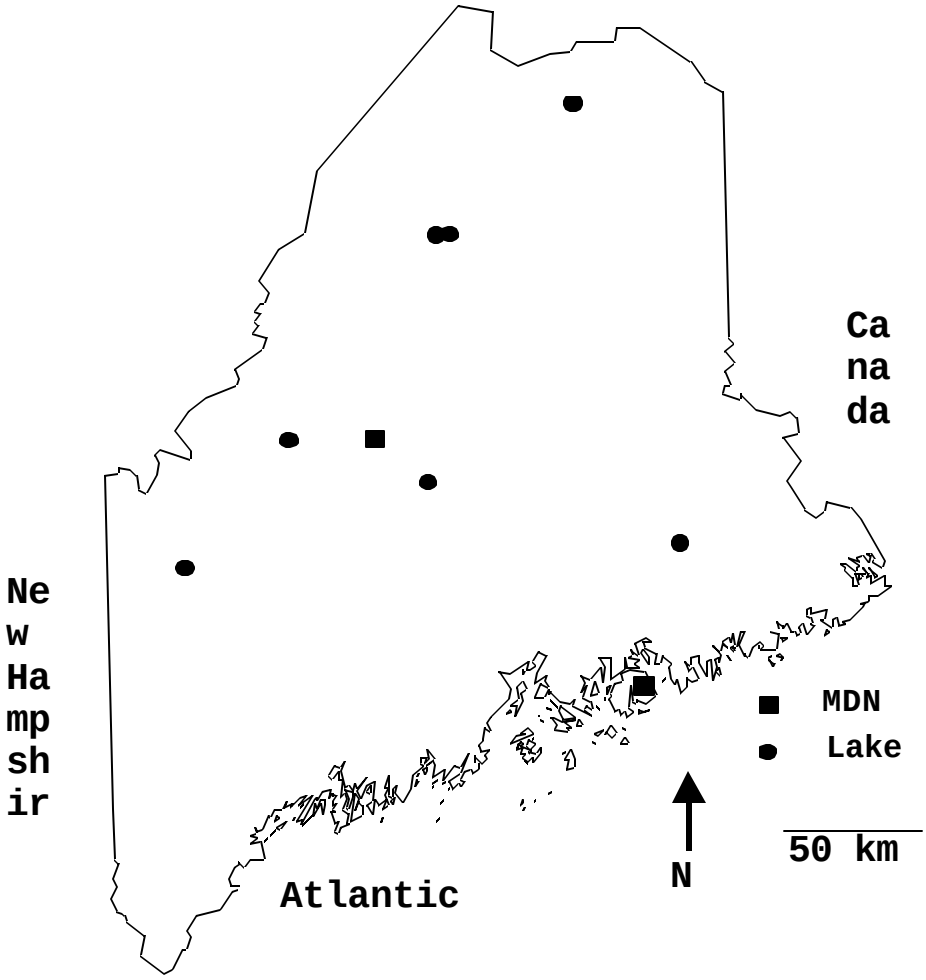
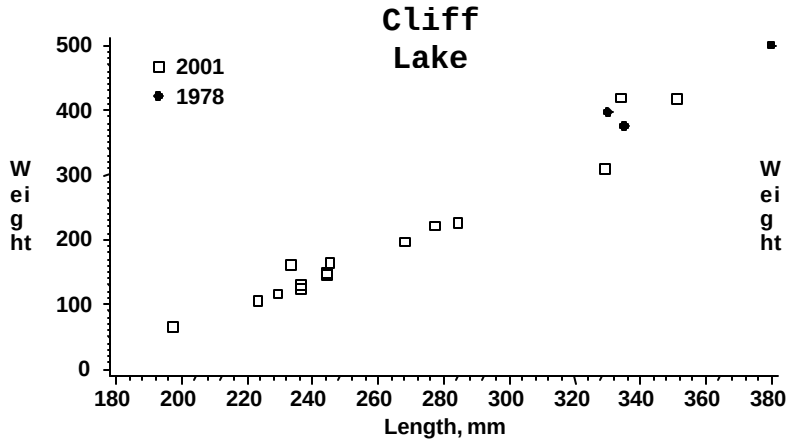
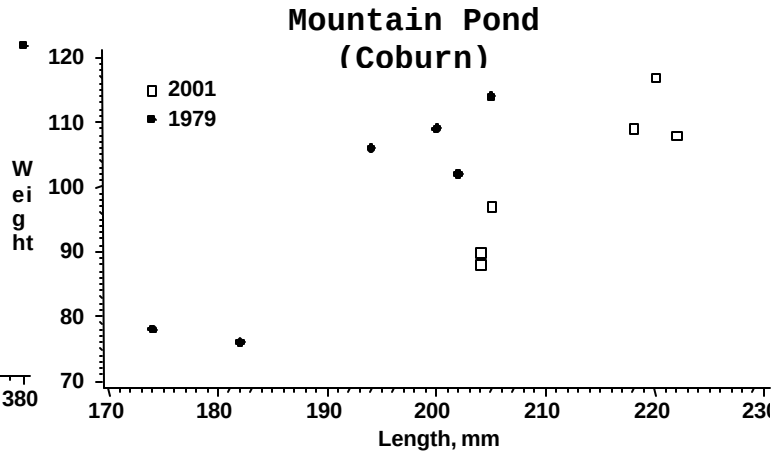
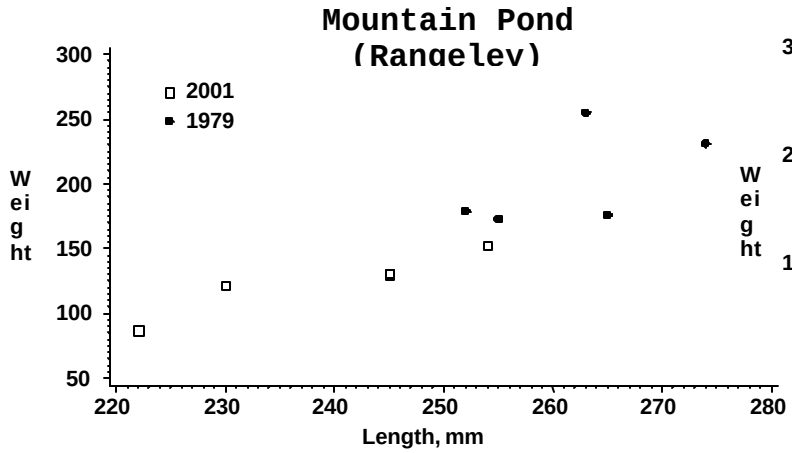


Figure 2a. Plot of fish weight on length for the two collection years. The regressions within each lake are not statistically different, except Mountain Pond (Coburn) are significantly different for intercept (Test of Common Regression, $F_{(1,9)} = 18.87$), but not for







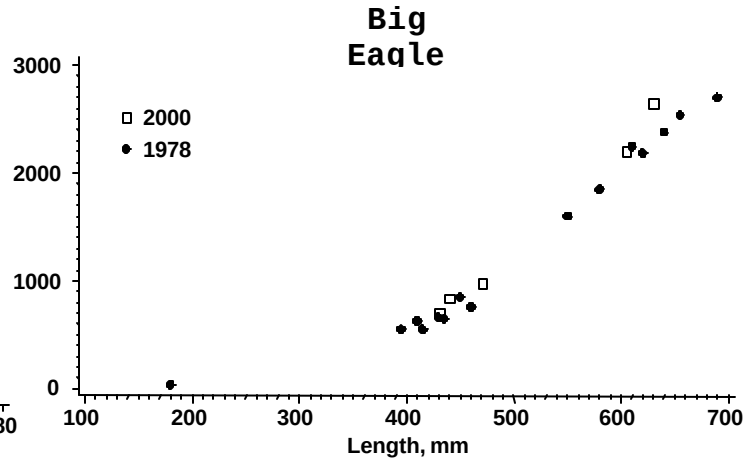


Figure 2b. Plot of fish weight on length for the two collection years. The regressions within each lake are not statistically different, except Green Lake are significantly different for

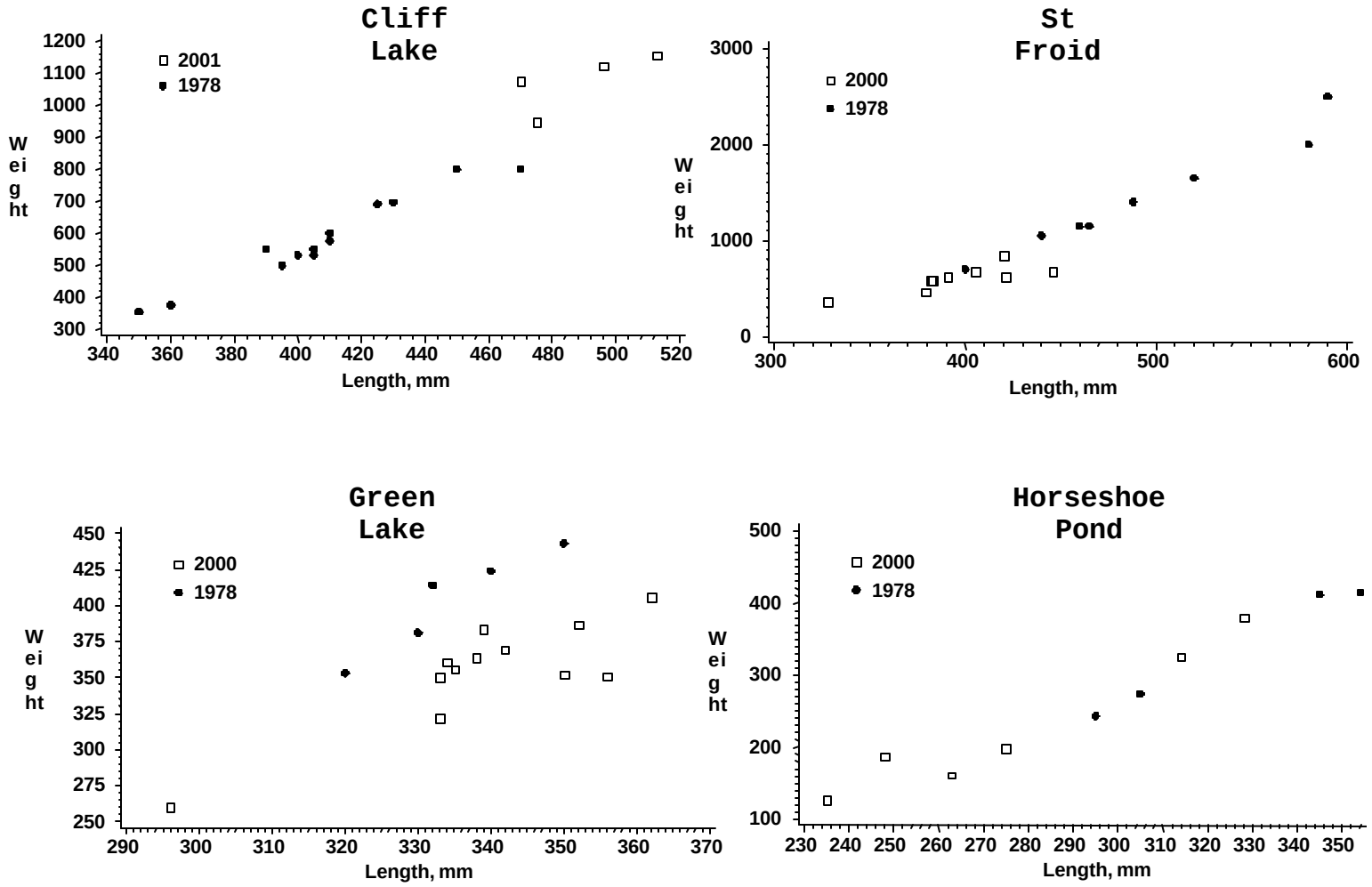
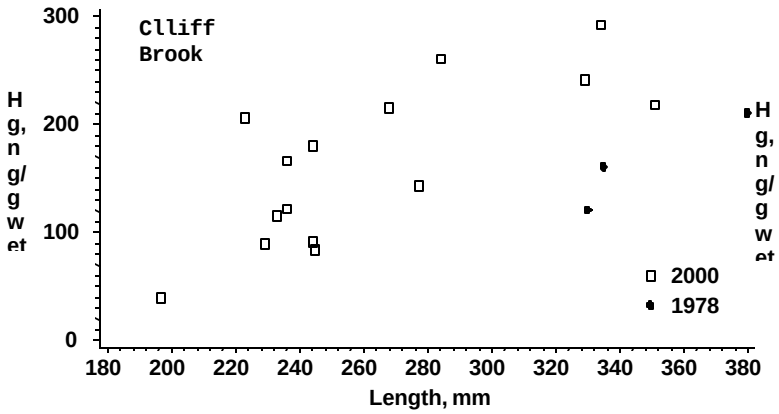
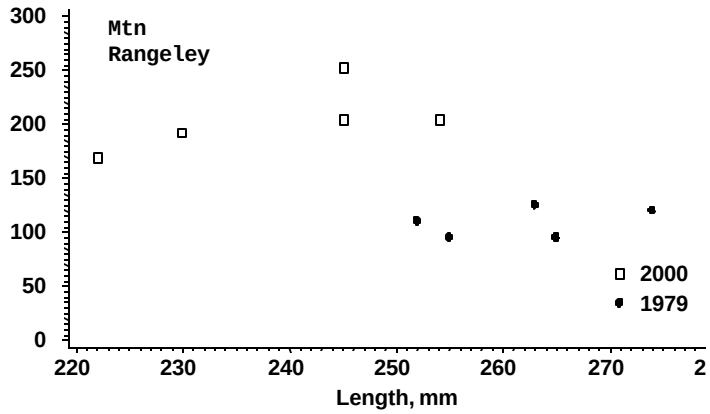
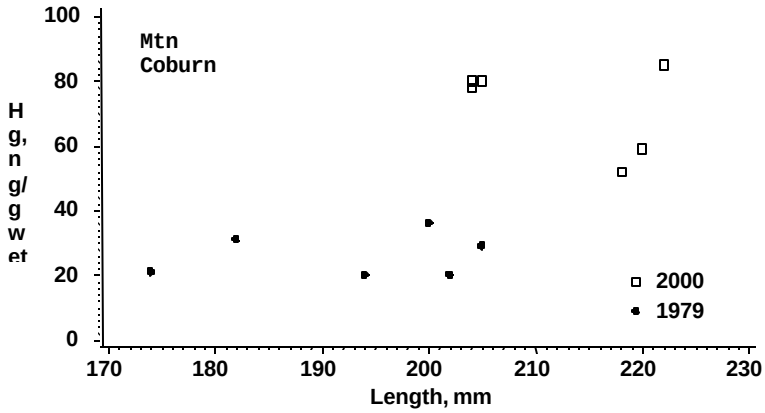


Figure 3a. Plot of fish mercury concentration on length for the two collection years by lake and species.







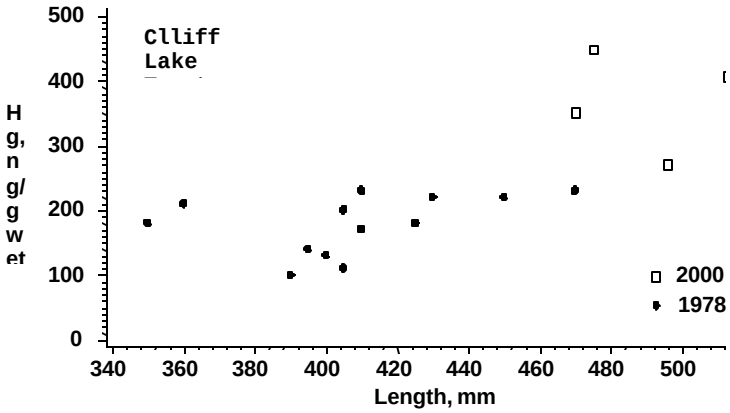


Figure 3b. Plot of fish mercury concentration on length for the two collection years by lake and species.

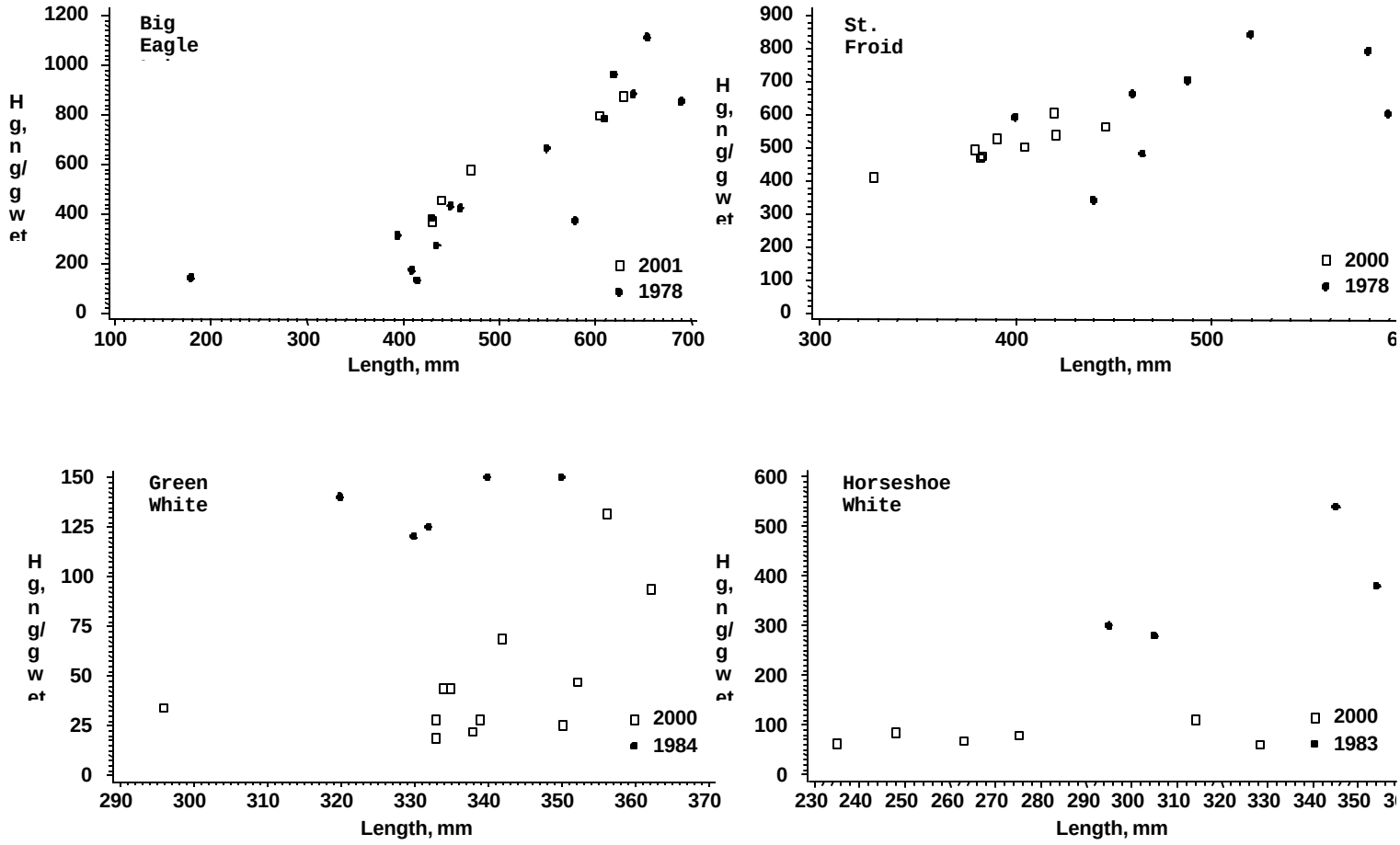


Figure 4. Total mercury concentration vs estimated age of sediment for Cliff Lake. Sediment age is estimated by ^{210}Pb dating.

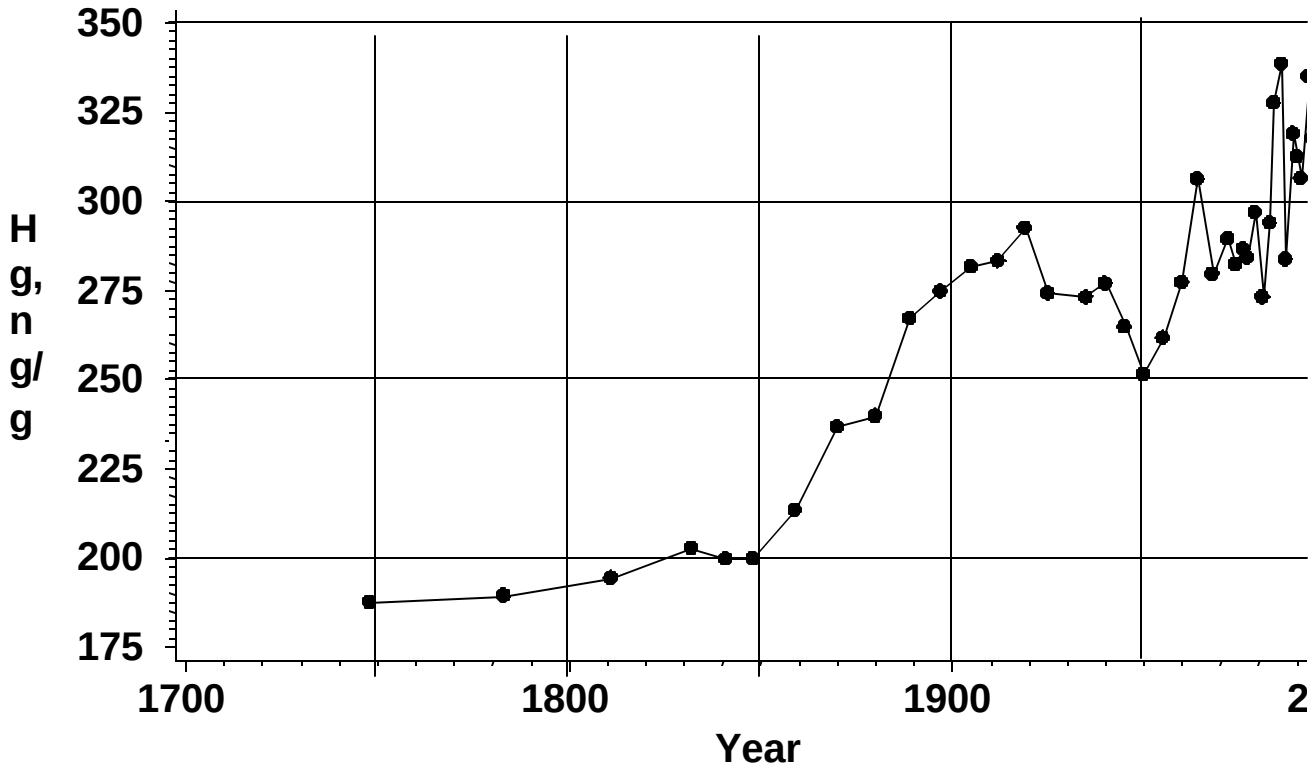


Figure 5. Total mercury accumulation rate vs estimated age of sediment for Cliff Lake. Sediment age is estimated by ^{210}Pb dating.

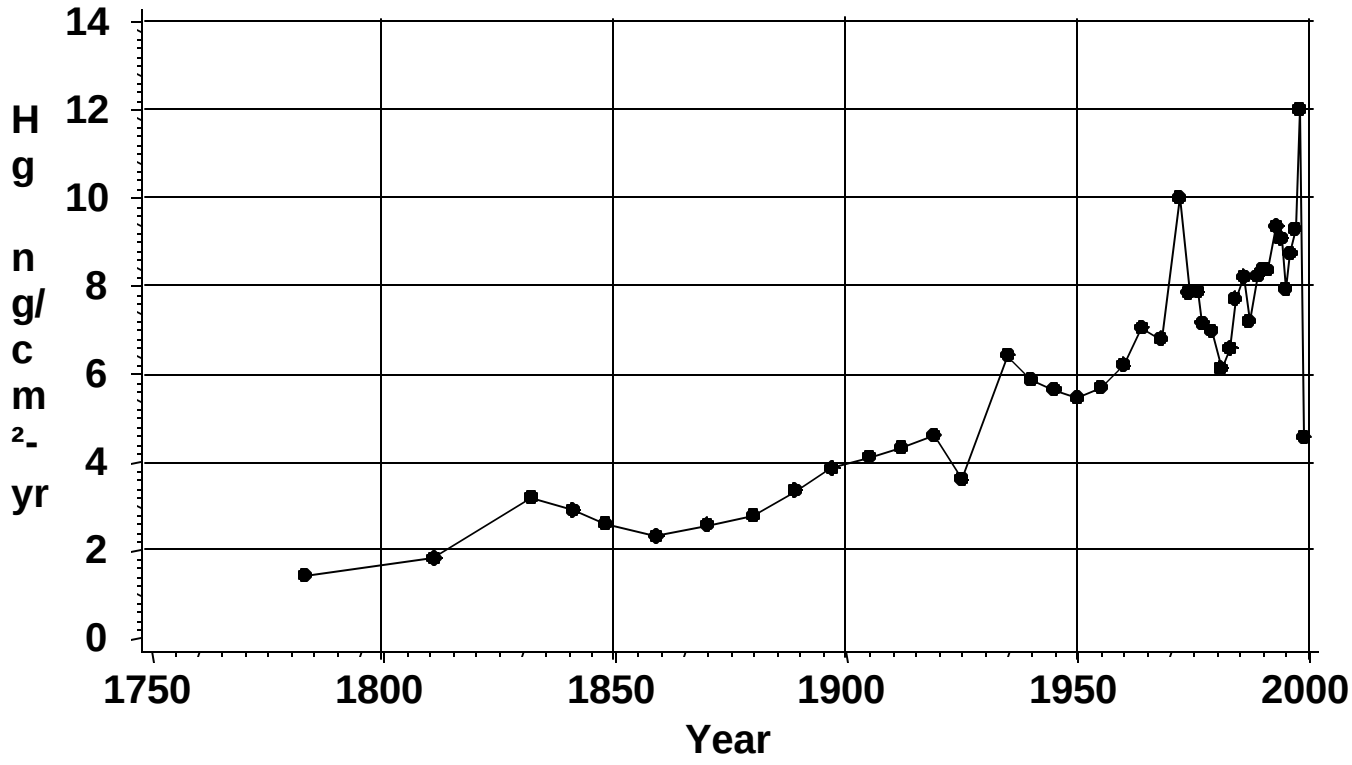


Figure 6. Anthropogenic mercury accumulation rate vs estimated age of sediment for Cliff Lake. Sediment age is estimated by ^{210}Pb dating. Anthropogenic accumulation rates are calculated by subtraction of the pre-1875 background rate and adjusting for changes in sediment accumulation rate. Raw data are plotted as calculated; 3-point average data are the mean of three consecutive measured rates centered on the plotted point.

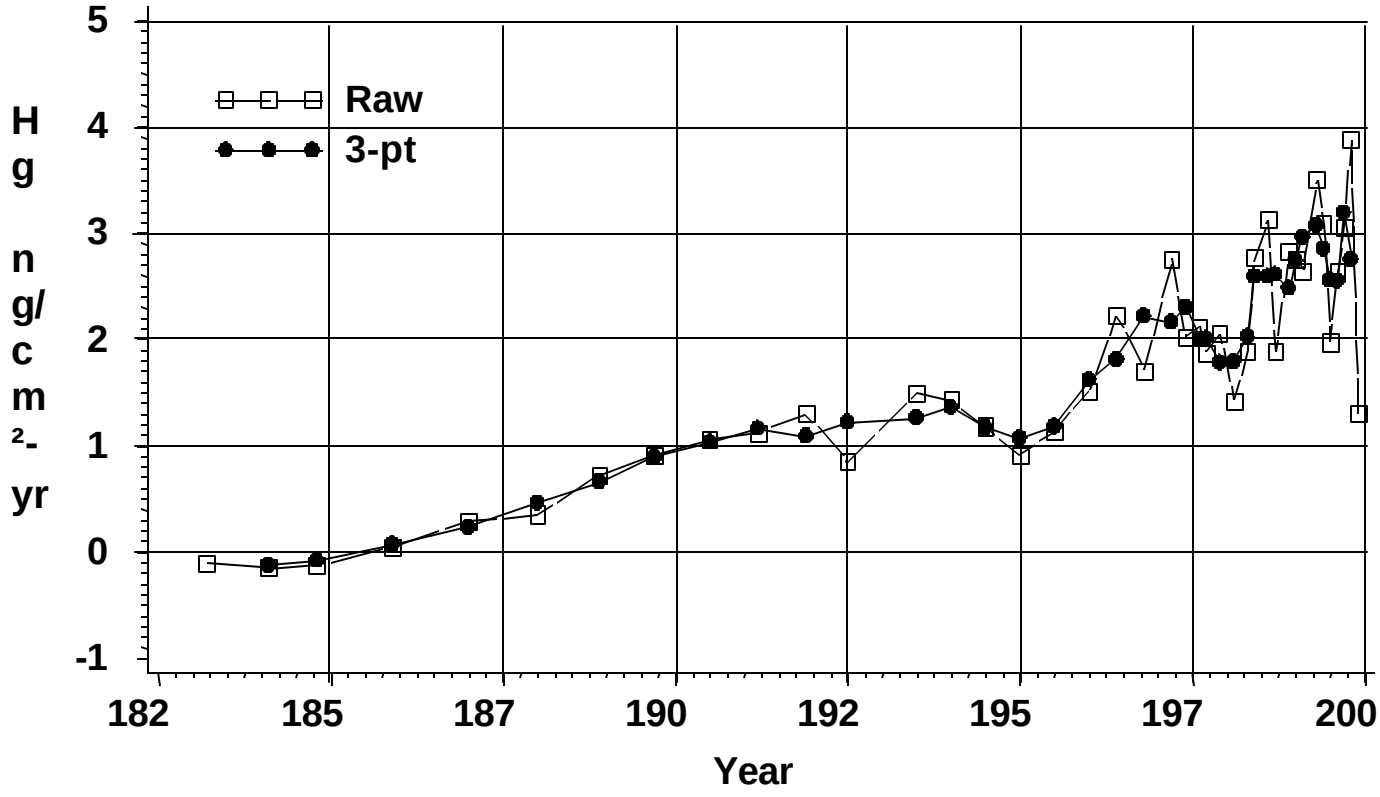


Figure 7. Linear regression of three-point running average anthropogeni accumulation rate vs estimated age of sediment for Cliff Lake for the pe during which fish mercury data are available. Sediment age is estimated dating. Anthropogenic accumulation rates are calculated by subtraction o 1875 background rate and adjusting for changes in sediment accumulation Three-point average data are the mean of three consecutive measured rate

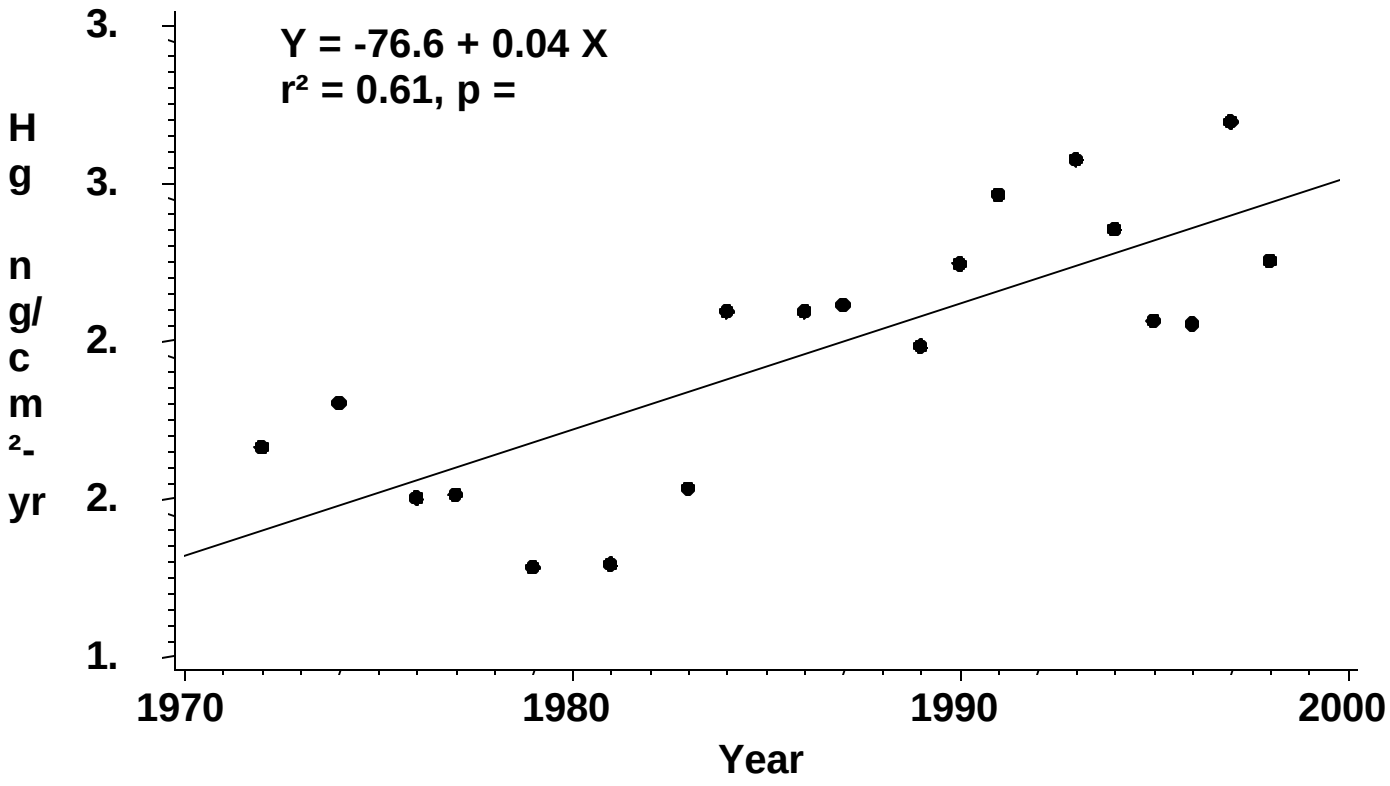


Figure 8. Annual mean mercury concentration at the Mercury Deposition Network stations at Acadia National Park and Greenville. *Data for 2001 are January to June only.

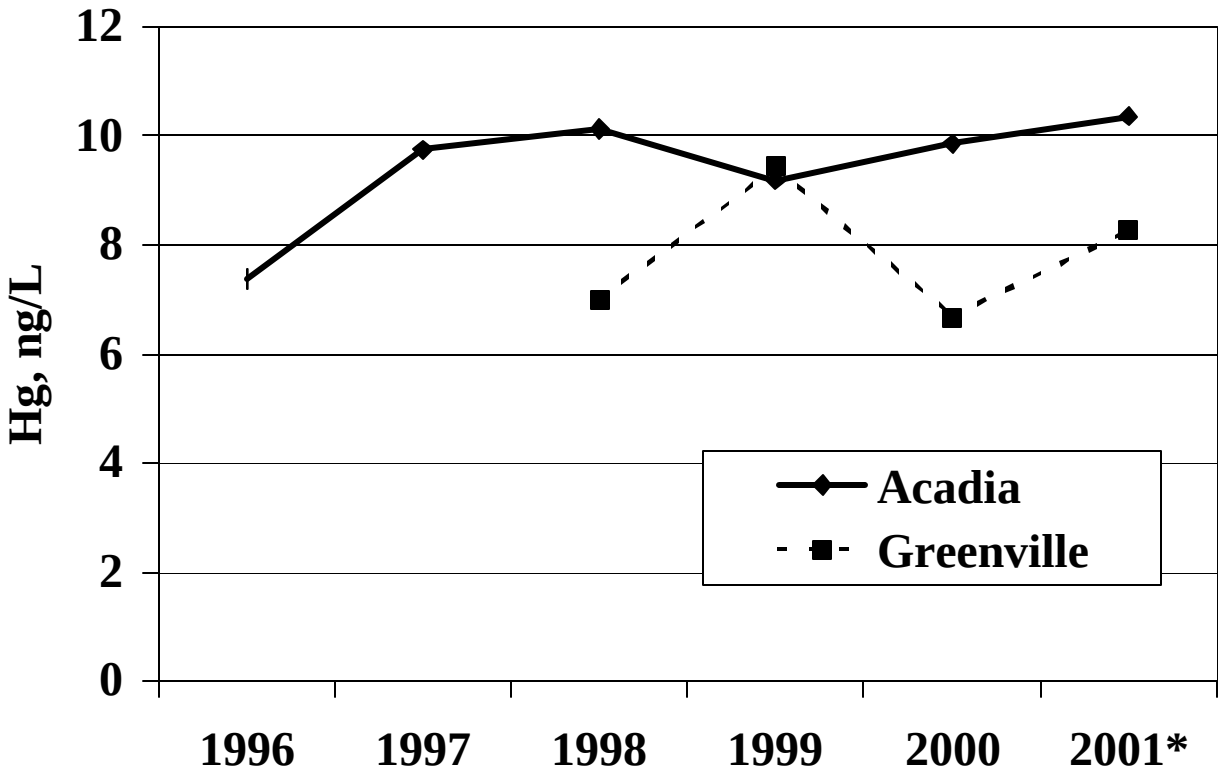
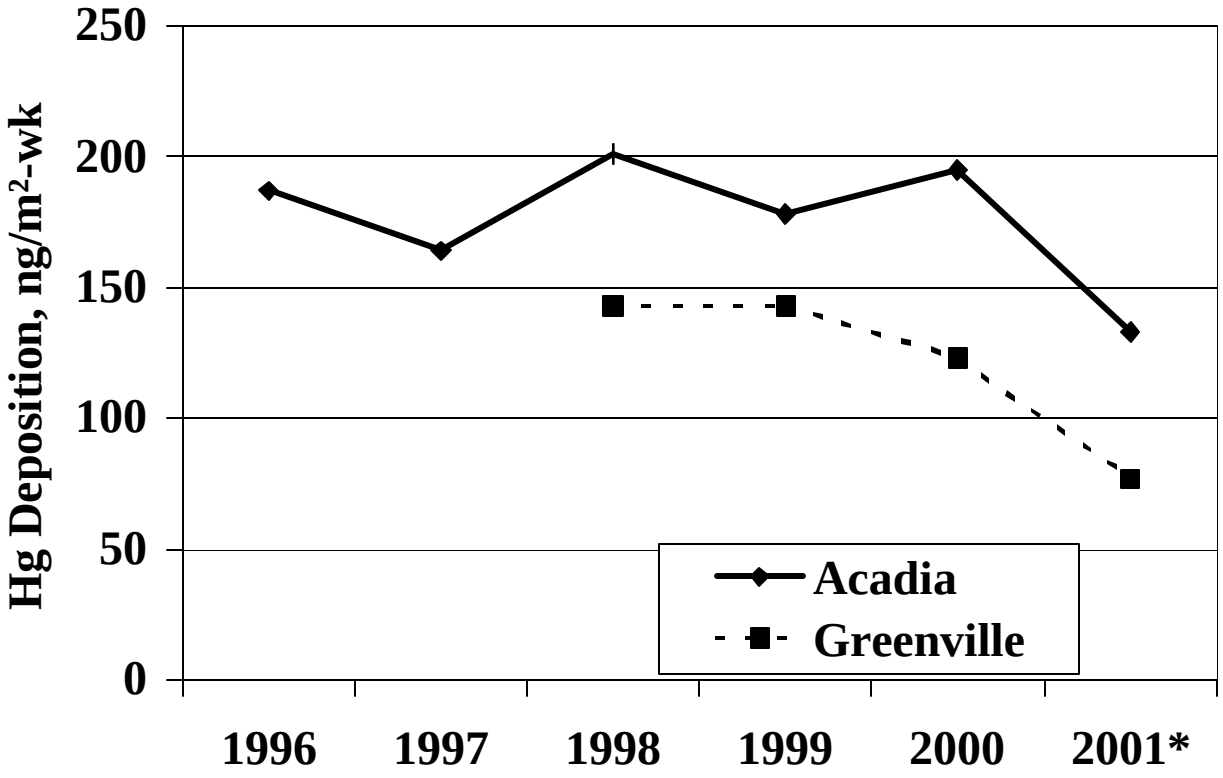


Figure 9. Annual mean mercury deposition at the Mercury Deposition Network stations at Acadia National Park and Greenville. *Data for 2001 are January to June only.



2.6

ANDROSCOGGIN LAKE SEDIMENTS

ANDROSCOGGIN LAKE SEDIMENTS

Monitoring of fish from Androscoggin Lake for dioxin as part of Maine's Dioxin Monitoring Program in 1996 documented concentrations of dioxins similar to those found in fish from the Androscoggin River nearby and higher than found in any other lake monitored in Maine (9 lakes). Since the Androscoggin River floods the lake one or more times each year, the river is the suspected source of dioxins to the fish in the lake. Additional fish samples collected in 1998, 1999, and 2000 have documented a continuing decline in dioxin concentrations to levels near background (Dioxin Monitoring Program Report, 2000 at http://www.state.me.us/dep/blwq/monitoring.htm).

In order to document the pathway, in 1999, surficial sediment samples were collected from 4 areas in the lake and analyzed for dioxins. Results were all below the detection limit (Table 2.6.1). To further explore the potential pathway, in 2000 sediment samples were collected at the lake outlet, as in 1999, at a station just upstream of the Dead River Dam and a station approximately half way between. Both surficial and subsurface samples were collected in order to determine historical and recent contamination. Results show that the lake outlet sample had significantly more dioxin than measured in 1999 and that both river stations also had measurable amounts. The difference between the 1999 and 2000 lake outlet concentrations may be due to the patchniness of sediments. Surficial sediment concentrations were slightly lower at the lake outlet and middle stations but much lower at the dam station than the subsurface samples, perhaps reflecting decreased discharges in recent years.

It is interesting that in 1999 the fish had more but the sediments had less than in 2000. The study should be repeated in 2002 to provide more documentation of sediment concentrations in the lake and river.

Table 2.6.1 Dioxin concentrations in Androscoggin Lake sediments.

Androscoggin Lake sediment DTE (ppt)

station	depth	1999 DTE	2000 DTE
L1	0-1"	0.1-0.7	7.6-8.1
	3-4"		8.0-8.2
L2	0-1"	0.03-0.7	
L3	0-1"	0.01-0.7	
L4	0-1"	0.06-0.7	
R1	0-1"		13.1-13.2
	2-3"		14.2-14.3
R2	0-1"		7.9-8.3
	1.5-2.5"		11.5-12.0

Ranges calculate for non-detects at 0 and at the detection limit.

DEP ID		ALW-SED-1	ALW-SED-2	ALW-SED-3	ALW-SED-4	ALW-SED-5	ALW-SED-6
Compound	**DL (ng/Kg, dry weight)**						
2378-tcdf	0.11	22.4	15.7	26.4	30.8	24.3	26.4
12378-pecdf	0.25	<DL	<DL	<DL	18.5	13.3	14.8
23478-pecdf	0.25	6.94	3.20	7.69	4.66	3.26	9.38
123478-hxcdf	0.25	4.22	2.88	6.29	6.29	7.69	12.7
123678-hxcdf	0.25	9.31	17.6	7.99	13.5	16.6	16.8
234678-hxcdf	0.25	<DL	<DL	<DL	<DL	2.11	1.99
123789-hxcdf	0.25	2.65	1.10	8.19	6.32	<DL	<DL
1234678-hpcdf	0.50	13.2	6.34	16.4	8.53	16.6	9.31
1234789-hpcdf	0.50	2.58	4.55	3.42	1.29	1.18	1.93
ocdf	0.50	8.75	37.5	8.45	8.12	10.6	6.63
2378-tcdd	0.10	<DL	<DL	1.03	1.36	<DL	<DL
12378-pecdd	0.25	<DL	0.13	2.93	3.63	<DL	<DL
123478-hxcdd	0.25	<DL	<DL	<DL	<DL	0.42	<DL
123678-hxcdd	0.25	<DL	<DL	<DL	<DL	1.18	<DL
123789-hxcdd	0.25	<DL	22.8	<DL	<DL	<DL	<DL
1234678-hpcdd	0.50	15.1	12.8	18.2	17.5	17.2	11.8
ocdd	0.50	94.8	105	129	104	108	82.6
Total TEQ (ND=0)		7.65	7.99	13.09	14.22	7.89	11.46
Total TEQ (ND=DL)		8.11	8.18	13.20	14.32	8.29	11.91
Sample weight (g dry weight)		45.3	48.9	51.9	50.2	50.4	50.2

Values less than the established MDLs are to be considered estimated values.

* = Values are influenced by the presence of diphenyl ethers and are estimated maximum concentrations.