APPLICATION OF THE REGIONAL MERCURY CYCLING MODEL (RMCM) TO PREDICT THE FATE AND REMEDIATION OF MERCURY IN ONONDAGA LAKE, NEW YORK

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Abstract. Onondaga Lake exhibits elevated concentrations of total mercury (Hg_T) and methyl-mercury (MeHg) in the water column, sediments and fish tissue due to industrial inputs, wastewater discharge and urban runoff. The steady-state Regional Mercury Cycling Model (RMCM) was calibrated to Onondaga Lake and applied to evaluate various remediation scenarios. Because of detailed data available for Onondaga Lake, the RMCM was effectively calibrated. Model predictions of water column and fish concentrations of Hg generally agreed with measured values. The model underestimated concentrations of Hg in sediments. Mass balance calculations show that inputs of Hg_T largely originate from tributary and wastewater inflows to the lake. In contrast, MeHg is largely derived from internal production. Model calculations suggest that elimination of Hg inputs from wastewater effluent and of drainage from a former chlor-alkali facility could greatly decrease Hg concentrations in fish tissue.

Keywords: budget, mass balance, mercury, regional model, Onondaga Lake

1. Introduction

Contamination of soil, sediment or water with mercury (Hg) is a serious environmental problem. Mercury bioaccumulates in aquatic food chains to the extent that consumption of contaminated fish may be hazardous to birds, mammals, and humans. Monomethyl Hg (MeHg) is the form of Hg that is absorbed readily from water by phytoplankton and subsequently bioconcentrates via the aquatic food chain (Mason *et al.,* 1995).

A crucial determinant of potential effects of Hg in aquatic ecosystems is the cycling of various species of Hg. Cycling consists of inputs to and outputs from the ecosystem, and the transport and transformations of Hg within the ecosystem. These processes are affected by site-specific factors, such as water chemistry, sediment transport and hydrodynamics. Inputs of Hg to aquatic ecosystems could originate from surface and ground water, industrial discharge or wastewater facilities, sediments and atmospheric deposition. Inorganic Hg ($Hg^{2+}{}_T$) is the predominant form entering these ecosystems and may form complexes with inorganic ligands (e.g., Cl^-, OH^-), bind with dissolved organic carbon, or sorb to particulate matter (Driscoll *et al.,* 1994). Inorganic Hg can be reduced to elemental Hg (Hg^o), which can volatilize to the atmosphere or be methylated to MeHg abiotically (Lee *et al.,* 1985) or by sulfate reducing bacteria in anoxic zones (Rudd, 1995). Monomethyl Hg can be further methylated to dimethyl Hg or can be demethylated by microbial processes.

Onondaga Lake received Hg discharges from a chlor-alkali manufacturer from 1946 to 1970 (USEPA 1973) and was closed to fishing in 1970 due to Hg contamination of fish tissue. Although the degree of contamination has decreased, a large fraction of the legal-size fish continues to exceed the U.S. Food and Drug Administration (FDA) limit of 1 $\mu g\ g^{-1}$ (NYSDEC, 1987). The lake has been reopened to recreational angling (since 1986), but an advisory for the consumption of fish taken has remained in effect through the present. Recent studies have found high concentrations of total Hg (Hg_T) and MeHg in the lake water, with Hg_T concentrations (2-35 ng L^{-1}; Bloom and Effler, 1990) exceeding values reported for uncontaminated surface waters (0.9-1.9 ng L^{-1} for Wisconsin seepage lakes; Fitzgerald and Watras, 1989) by about an order of magnitude. Lake sediments are highly contaminated with Hg_T (Klein and Jacobs, 1995). Concern has been expressed that atmospheric deposition may be an important source of Hg to Onondaga Lake.

The primary objective of this study was to evaluate the cycling of Hg in Onondaga Lake under ambient conditions and various remediation scenarios of external Hg loading. The specific objectives were to: 1) determine external inputs of Hg_T and MeHg to the lake and compare these values to the internal rates of accumulation of Hg_T and MeHg in the hypolimnion of the lake, 2) use the Regional Mercury Cycling Model (RMCM; Tetra Tech Inc., 1996) to simulate mass fluxes of Hg_T and MeHg into, within and out of the lake, and 3) simulate the effects of various remediation strategies of reduced external loads on the concentrations of Hg species in water, sediments and fish.

2. Materials and Methods

2.1. Site Description

Onondaga Lake (lat. 43°06'54"N; long. 76°14'34"W) is located adjacent to the city of Syracuse, in Onondaga County, New York (Figure 1). The lake (1.4×10^8 m^3 volume, 12 m mean depth, 20.5 m maximum depth) is a hypereutrophic, dimictic lake and it flushes rapidly, 2.5 to 5.0 times annually (Devan and Effler, 1984). The lake water is well buffered with a pH of between 7 to 8.5 (Driscoll *et al.,* 1993). The major hydrologic inputs are Ninemile Creek and Onondaga Creek, which together account for about 62 % of the annual inflow to the lake, the Onondaga County Metropolitan Wastewater Treatment Plant (METRO), which is the third largest source of water to the lake making up 19 % of the annual inflow, Ley Creek, Harbor Brook, and Bloody Brook.

2.2. Sampling and Analytical Procedures

Clean sampling techniques (Fitzgerald and Watras, 1989; USEPA, 1995a) were used to collect monthly water samples from October 1995 to September 1996 in selected tributaries including Ninemile Creek downstream at Lakeland and upstream at Amboy, Onondaga Creek, Harbor Brook, Ley Creek, and the effluent discharged from METRO. Analytical protocols described in USEPA (1995b) and Gill and Fitzgerald (1987) were used to analyze the tributary water samples for Hg_T. Methyl Hg analysis was done in accordance with Hovart *et al.* (1993) and Liang *et al.* (1994). Mean daily flow measurements for the major tributaries were obtained from the U.S. Geological Survey.

Because the minor tributaries, lake water and sediments, and groundwater inputs to the lake were not sampled during this period, Hg values obtained in recent studies (Bloom and Effler, 1990; Jacobs *et al.,* 1995; Klein and Jacobs, 1995; Wang and Driscoll, 1995) were used. The minor tributaries are not gauged, so flow rates for these tributaries were estimated using the empirical relationship in Effler (1996). Concentrations of Hg in fish for the sampling period were obtained from the New York State Department of Environmental Conservation (R. Sloan, 1997, NYSDEC, unpublished data).

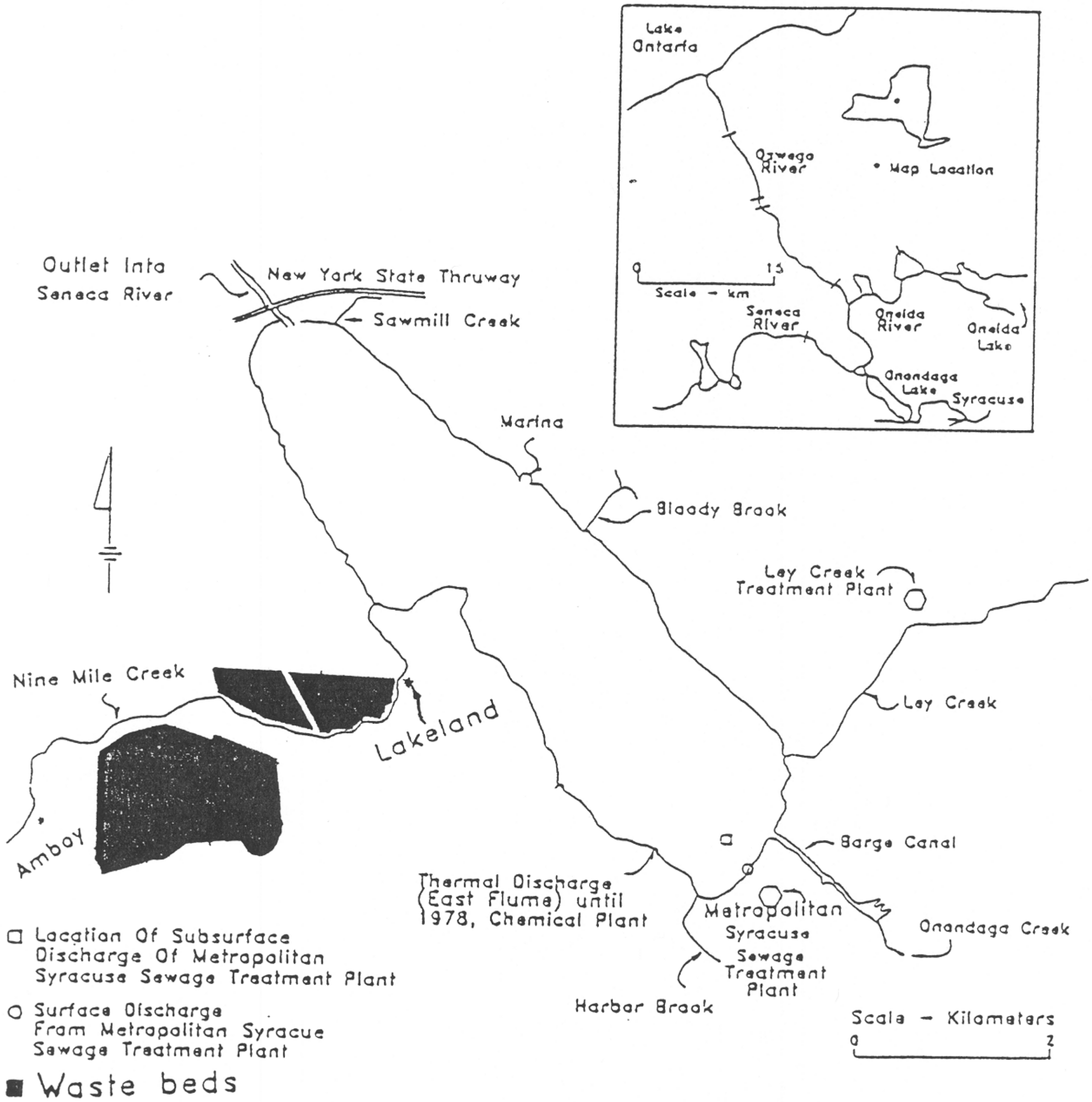

Fig. 1. Map of Onondaga Lake showing input waters (after Effler, 1996).

2.3. MODELING FLUX AND MODELING APPROACHES

Mass loadings from the input waters were estimated using the program FLUX (Walker, 1987). FLUX evaluates the flow/concentration relationships developed from the sample record onto the entire flow record to calculate total mass loading and associated error statistics. The areal mass fluxes of Hg_T and MeHg measured for Ninemile at Amboy were considered as background values for the Onondaga Lake watershed. The difference in mass fluxes of Hg_T and MeHg for Ninemile Creek at Lakeland and Amboy were assumed to be due to inputs from the former chlor-alkali facility, which was located along the stream reach. The areal mass fluxes of Hg_T and MeHg at Onondaga Creek, Ley Creek and Harbor Brook, which drain through the city of Syracuse, less the background values obtained for Ninemile Creek at Amboy were assumed to be due to Hg inputs from urban runoff.

The accumulation of Hg_T and MeHg in the hypolimnion of Onondaga Lake during the ice free period was estimated from the slopes of the regression analysis of Hg_T and MeHg pools for 1992 as a function of time on a lake surface area basis, using data obtained from Jacobs *et al.* (1995). These values were compared to the corresponding external fluxes estimated from the loadings from the tributaries.

The Regional Mercury Cycling Model (RMCM) was used to simulate the mass balance of Hg_T and MeHg in the lake. This is a steady-state mechanistic model, designed to simulate the biogeochemical cycling of Hg in lakes. Input parameters include lake/watershed physical characteristics, water chemistry, atmospheric deposition and aquatic biomass. Most of these parameters were obtained from Effler (1996). Atmospheric deposition was estimated based on values of 20 ng Hg_T L^{-1} (Glass *et al.,* 1991) and 0.016 ng MeHg L^{-1} (Fitzgerald *et al.,* 1991) in wet deposition, applied to the lake surface area. A dry deposition flux of Hg_T of 3.5 μg^{-2} yr^{-1} (Fitzgerald *et al.,* 1991) was assumed in the model. Groundwater flux was estimated from concentrations of Hg in groundwater (Jacobs *et al.,* 1995) applied to the expected groundwater flow into the lake (Effler, 1996). Mercury concentrations at Ley Creek were used to estimate mass fluxes from minor tributaries. A detailed description of the RMCM is provided by Tetra Tech Inc. (1996).

The model was calibrated using Hg^o concentrations in the water column observed by Bloom and Effler (1990). Because the model tends to predict high Hg^o concentrations, rates of demethylation (1×10^{-4} to 1×10^{-5} ng MeHg $^{-3}$ da^{-1}) and $Hg^{2+}{}_T$ reduction (3×10^{-2} to 3×10^{-5} da^{-1}) were adjusted from default values so that model simulated values matched measured water column Hg^o concentrations. Default values for the model were used for all other parameters (Tetra Tech, 1996). The model was used to predict Hg concentrations in the water column, sediments and smallmouth bass in the lake. These values were compared to measured values observed in recent studies. Juvenile yellow perch were assumed to be prey species for smallmouth bass. To assess the possible Hg remediation scenarios, we considered hypothetical simulations with the calibrated RMCM. These scenarios included: 1) business as usual (current Hg loadings), 2) elimination of the inputs of Hg from METRO, 3) elimination of inputs of Hg from the former chlor-alkali facility, 4) elimination of inputs of Hg from both the former chlor-alkali facility and METRO, 5) elimination of inputs of Hg from the former chlor-alkali facility, METRO and urban runoff, and 6) inputs of Hg from only atmospheric deposition to the watershed.

3. Results and Discussion

3.1. Mass Loads from Tributaries to Onondaga Lake

Mass loadings of Hg_T and MeHg calculated using FLUX showed that the total loads to the lake from the major tributaries were 9,498 g yr^{-1} and 310 g yr^{-1}, respectively. METRO effluent and drainage from the former chlor-alkali facility were major contributors of Hg_T to the lake. Localized inputs from groundwater sources also were responsible for Hg_T in Onondaga Creek. The inputs from leachates and groundwater sources to both Ninemile Creek and Onondaga Creek were diluted during the spring snow melt period when flow rates were high. METRO effluent accounted for about half the external MeHg load (49 %; Table I) to the lake. In Harbor Brook and Ley Creek, watershed characteristics and sulfate reduction played important roles in the dynamics of MeHg.

TABLE I
Mass loadings and percent contribution of Hg_T and MeHg to Onondaga Lake (g yr^{-1}).

Input	Hg_T	MeHg
Background	2039 (21 %)	59.0 (19 %)
Urban Runoff	867 (9 %)	59.9 (19 %)
METRO	3473 (37 %)	151.2 (49 %)
Former Chlor-alkali Facility	3119 (33 %)	39.8 (13 %)
Total	9498	310.0

3.2. Rates of Hypolimnetic Accumulation of Mercury

The internal rates of Hg_T and MeHg accumulation in the hypolimnion of Onondaga Lake (Table II) were estimated for the summer stratification period (May - September 1992). These values were obtained from the slopes of the regression of the hypolimnetic Hg pool expressed in g m^{-2} on a lake surface area basis as a function of time. The internal rate of Hg_T release in the hypolimnion was 0.45 μg m^{-2} da^{-1}. This value is higher than the rate estimated by Driscoll and Wang (1996; 0.28 μg m^{-2} da^{-1}). The rate of $Hg^{2+}{}_T$ release from hypolimnetic sediments was 0.22 μg m^{-2} da^{-1}. The rate of MeHg release (0.23 μg m^{-2} da^{-1}) was about half the estimated rate of Hg_T release. The source of $Hg^{2+}{}_T$ and MeHg supplied to the hypolimnetic waters is unclear. The release of $Hg^{2+}{}_T$ (Hg_T-MeHg) to the hypolimnion may be due to desorption of Hg from recently deposited particles, mineralization of Hg in recently deposited organic matter, or from sediments. The accumulation of sulfide in the hypolimnion may facilitate release of $Hg^{2+}{}_T$ from particles through the formation of aqueous complexes (Wang and Driscoll, 1995). The supply of MeHg to the hypolimnion also may be due to release from recently deposited particulate matter or internal production of MeHg.

The external input of $Hg^{2+}{}_T$ from the tributaries (2.08 μg m^{-2} da^{-1}) was about an order of magnitude higher than release of $Hg^{2+}{}_T$ to the hypolimnion (0.22 μg m^{-2} da^{-1}). In contrast the external inputs of MeHg (0.07 μg m^{-2} da^{-1}), was considerably less than the rate of

TABLE II

Comparison of rates of external Hg input to internal Hg release/production from hypolimnetic sediments on a lake surface area basis for Onondaga Lake (in μg m^{-2} da^{-1}).

	Hg_T	Hg^{2+}_T	MeHg
External	2.15	2.08	0.07
Internal	0.45	0.22	0.23

Note: $[Hg^{2+}_T] = [Hg_T] - [MeHg]$

hypolimnetic accumulation (0.23 μg m^{-2} da^{-1}). The magnitude of these values suggests that external inputs of Hg^{2+}_T are sorbed to particles, deposited to sediments, and subsequently, released to the water column in the hypolimnion. Moreover, a portion of the Hg^{2+}_T supplied by external inputs appears to be methylated in the anoxic zone during the summer stratification period. The rate of MeHg accumulation in the hypolimnion of Onondaga Lake compares to the rate of MeHg production reported for the ice-free conditions in the Wabigoon/English River Lake System (0.037-0.55 μg m^{-2} da^{-1}; Parks *et al.,* 1989).

3.3. Model Evaluation and Mass Balances of Hg_T and MeHg for Onondaga Lake

Results from the calibration of the RMCM to current inputs of Hg_T and MeHg were compared to measurements made in previous studies of Hg in Onondaga Lake (Table III). This was done in order to appraise the performance of the model in predicting concentrations of Hg_T and MeHg. Concentrations of Hg_T and MeHg simulated by the model in the epilimnion, hypolimnion, and in fish lie within the range of observed values from previous studies. Concentrations of Hg_T in the sediments were significantly lower than values observed by Klein and Jacobs (1995). The RMCM simulates only labile (reactive) Hg (R. Harris, personal communication, June 1997). Therefore, if sediments contain a high fraction of non-labile Hg (Hg that does not readily react with the aqueous phase), the discrepancy between measured and model-calculated values could be explained.

TABLE III

Comparison of model-predicted concentrations of Hg in the water column, sediments and fish with measurements made in previous studies of Onondaga Lake.

Media	Species	Model Value	Jacobs *et al.* (1995)	Wang and Driscoll (1995)	Bloom and Effler (1990)	Klein and Jacobs (1995)	NYS DEC (1987)
Epilimnion (ng L^{-1})	Hg^o	0.014			0.01 - 0.24		
	MeHg	0.5	0.2 - 1.0		0.4 - 2.0		
	Hg_T	8.5	3.0 - 9.4	4.0 - 15.7	7.1 - 18.8		
Hypolimnion (ng L^{-1})	Hg^o	0.5			0.02 - 0.07		
	MeHg	3.1	0.4 - 12		1.61 - 6.68		
	Hg_T	29.5	5.3 - 21.7	1.4 - 34.9	10.1 - 25.7		
Sediments (μg g^{-1})	MeHg	0.14					
	Hg_T	0.29				1 - 1.8	
Smallmouth bass (μg g^{-1})	Hg	1.10					0.95 - 1.4

The RMCM was used to simulate mass fluxes of Hg_T and MeHg in the lake (Table IV). Fluxes of Hg_T and MeHg obtained from previous studies are also included in Table IV for comparison. The total input of Hg_T to the lake during the study period was 815 $\mu g\ m^{-2}\ yr^{-1}$ with the major tributaries, including METRO effluent, contributing about 96 %. Atmospheric deposition directly to the lake surface, which includes both dry and wet deposition, accounted for about 2.6 % of the input fluxes. Other inputs from minor tributaries not sampled and groundwater were estimated to contribute less than 1 % of the input fluxes of Hg_T to the lake. The sinks of Hg_T include the lake outflow, volatilization and sediment burial. Outflow of Hg_T from the lake was estimated to be 524.5 $\mu g\ m^{-2}\ yr^{-1}$ (77 % of the Hg_T sinks), while 154.9 $\mu g\ m^{-2}\ yr^{-1}$ (22 % of the Hg_T sinks) was retained through net sedimentation or burial.

TABLE IV
Comparison of mass balances of Hg_T and MeHg ($\mu g\ m^{-2}\ yr^{-1}$) for Onondaga Lake for October 1995-September 1996, with results reported in previous studies.

	This study		Driscoll and Wang (1996)	Henry et al. (1995)	
Sources/Sinks	Hg_T	MeHg	Hg_T	Hg_T	MeHg
Major Tributaries	789.3	25.8	450.8	1,133.3[1]	21.7[1]
Atmospheric Input	21.0	0.2	10.8	36.7	0.5
Other Inflows	4.9[2]	0.2[2]	3.3[2]	1.7[3]	0.1[3]
Outflow	514.5	32.3	466.7	233.3	20.0
Volatilization	0.2	0	12.5	1.3	0
Net Methylation	-	146.7	-	-	52.5
Burial[4]	152.9	132.6	9.2	936.7	50.3

[1] Includes minor tributaries
[2] Input other minor tributaries and groundwater
[3] Groundwater inputs only
[4] Net of settling, resuspension and diffusion from sediments

The major tributaries including METRO supplied 25.8 $\mu g\ m^{-2}\ yr^{-1}$ MeHg to Onondaga Lake, about 14.9 % of the total inputs (172.8 $\mu g\ m^{-2}\ yr^{-1}$). The major source of MeHg in the lake was net internal production (146.7 $\mu g\ m^{-2}\ yr^{-1}$; 84.8 % of the total MeHg inputs). Atmospheric deposition and inputs from minor tributaries and groundwater were minor sources contributing less than 1 % to the MeHg inputs of the lake. Comparing the rate of hypolimnetic accumulation of MeHg estimated above (0.23 $\mu g\ m^{-2}\ da^{-1}$) with the net MeHg production estimated using the RMCM (172.8 $\mu g\ m^{-2}\ yr^{-1}$; i.e. 0.47 $\mu g\ m^{-2}\ da^{-1}$), suggests that the model may provide a reasonable estimate of in-lake MeHg production. Our value of the rate of MeHg accumulation in the hypolimnion undoubtedly underestimates the true value as methylation also occurs in the epilimnetic sediments and/or in the hypolimnetic sediments during the nonsummer months (October to April). The major sinks for MeHg in the lake were net sedimentation or burial (132.6 $\mu g\ m^{-2}\ yr^{-1}$; 80 % of the total MeHg sink) and outflow (32.3 $\mu g\ m^{-2}\ yr^{-1}$; 20 % of the total MeHg sink).

These fluxes were compared with previous studies conducted in Onondaga Lake. The Hg_T fluxes were intermediate between the values reported in the 1992 study by Driscoll and Wang (1996) and Henry *et al.* (1995), while MeHg fluxes were higher than those reported by Henry *et al.* (1995). Differences in fluxes could be due to the method of flux estimation, the model used and the water quality parameters used in calibration. In all mass balance studies, inputs from the tributaries, including METRO, were the major source of Hg_T to the lake. While this study and that of Driscoll and Wang (1996) indicate that outflow was the major sink of Hg_T, Henry *et al.* (1995) reported that net sedimentation was the major sink. For MeHg, net methylation was the major source and sediment burial the major sink. These observations are consistent with Henry *et al.* (1995).

3.4. Impact of Reduced External Inputs of Mercury to Onondaga Lake

With significant inputs of Hg originating from the former chlor-alkali facility, METRO and urban runoff, the effects of reducing these inputs on Hg concentrations in the epilimnion, hypolimnion, sediments and fish in the lake were investigated using the RMCM. The fish species used in the simulations was smallmouth bass and Hg concentrations for the five-year age class were reported. The various scenarios investigated were: 1) current inputs, 2) removal of the METRO inputs, 3) removal of the inputs from the former chlor-alkali facility, 4) combined removal of METRO inputs and inputs from the former chlor-alkali facility, 5) combined removal of METRO, inputs from the former chlor-alkali facility and urban runoff, and 6) only atmospheric deposition of Hg to the lake watershed. Results of the concentration and fluxes for these scenarios are given in Tables V and VI, respectively.

Model simulations showed that reductions in tributary loads for the various scenarios resulted in decreases in Hg concentrations in the lake water, sediments and fish. Because the model calculates steady-state values, and because water quality parameters are assumed to remain constant, percentage reduction in concentrations and fluxes predicted closely followed the percentage load reduction. When inputs from both METRO and the former chlor-alkali facility were eliminated (Scenario 4), decreasing input fluxes of Hg_T and MeHg by 69 % and 61 %, respectively, the predicted concentrations of Hg_T and MeHg in the water column, sediments and fish were reduced by about 66 % and there was a 67 % reduction in net methylation for the lake. The model calculations suggest that atmospheric deposition contributes little Hg to Onondaga Lake. Results of this analysis suggest that reduction of major inputs to Onondaga Lake could substantially reduce fish Hg concentrations. Note that different sources of Hg_T to the lake undoubtedly exhibit different bioavailability (e.g., wastewater effluent vs. leachate from the former chlor-alkali facility). While these differences are undoubtedly important, they are not reflected in the model calculations.

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TABLE V
Simulated concentrations of Hg in water (ng L^{-1}), sediments (μg g^{-1}) and fish (μg g^{-1}) in Onondaga Lake for the various scenarios considered.

	Epilimnion			Hypolimnium			Sediments		Fish
Scenario	Hg^o	MeHg	Hg_T	Hg^o	MeHg	Hg_T	MeHg	Hg_T	MeHg
1	0.014	0.51	8.54	0.45	3.09	29.45	0.140	0.288	1.101
2	0.014	0.31	5.54	0.42	1.94	19.11	0.088	0.184	0.686
3	0.014	0.37	5.88	0.43	2.19	20.31	0.099	0.200	0.789
4	0.014	0.17	2.83	0.39	1.04	9.94	0.047	0.096	0.375
5	0.013	0.12	2.06	0.38	0.72	7.32	0.033	0.068	0.257
6	0.01	0.03	0.44	0.37	0.14	1.89	0.001	0.014	0.050

TABLE VI
Major fluxes of Hg_T and MeHg (μg m^{-2} yr^{-1}) in Onondaga Lake for the various scenarios considered.

	Inflow streams		Outflows		Burial		Net Methylation
Scenario	Hg_T	MeHg	Hg_T	MeHg	Hg_T	MeHg	MeHg
1	789.3	25.8	514.5	32.3	152.9	132.6	146.7
2	500.7	13.2	332.6	20.0	98.9	88.5	94.9
3	530.1	22.5	351.2	23.3	104.4	99.7	100.1
4	241.5	9.9	168.9	11.0	50.2	47.5	48.2
5	169.5	4.9	123.4	7.5	36.7	33.0	35.2
6	25.7	1.4	25.7	1.4	7.1	6.4	7.4

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