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Trophic Transfer Efficiency of Mercury to Lake Whitefish *Coregonus clupeaformis* from its Prey

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Abstract In the laboratory, net trophic transfer efficiency of mercury to lake whitefish *Coregonus clupeaformis* from its prey was estimated to be 63.5%. Assuming that gross trophic transfer efficiency of mercury to lake whitefish from its prey was equal to 80%, we estimated that the rate at which lake whitefish eliminated mercury was 0.000730 day^{-1}. Our laboratory estimate of mercury elimination rate was 2.4 times lower than the value predicted by a published regression equation developed from previous studies on mercury elimination rates for fish. Thus, our results suggested that mercury elimination rates for fish have been overestimated in previous studies.

Keywords Elimination rate · Excretion rate · Mercury · Tracers

The efficiency with which fish retain contaminants from their prey is a critical factor governing contaminant accumulation in fish (Madenjian et al. 1994, 1998). Estimates of trophic transfer efficiency are needed to predict future risk to people and wildlife eating contaminated fish (Calabrese and Baldwin 1993).

Gross trophic transfer efficiency refers to the efficiency with which the contaminant in the food ingested by the predator is taken up through the gut wall of the predator. Once a quantity of contaminant has been taken up through the gut wall, a portion of this quantity may eventually be eliminated from the fish's body, and/or a portion of this quantity may be metabolically transformed into another chemical compound. Net trophic transfer efficiency refers to the efficiency with which the contaminant in the food ingested by the predator is retained by the predator, including the losses due to elimination and metabolic transformation (Thomann and Connolly 1984).

Mercury is a persistent contaminant that can damage the neurological systems in humans and wildlife (Bakir et al. 1973; Wren 1986). Mercury contamination in fish is of special concern because fish in the diet represents the primary source of mercury contamination in humans and in fish-eating wildlife (Wren 1986). Several mathematical models have been developed for mercury accumulation in fishes (Norstrum et al. 1976; Rodgers 1994; Trudel and Rasmussen 2001). For most of these models, gross trophic transfer efficiency of mercury to the fish from its food is assumed to be 80%. Despite the importance of this assumption in predicting mercury concentration in fish, only a few laboratory measurements of these trophic transfer efficiencies have been made (Trudel and Rasmussen 2001).

Trudel and Rasmussen (1997) used published estimates of mercury elimination rates to develop a general model for methylmercury elimination by fish. Using multiple linear regression, these researchers developed a predictive equation for methylmercury elimination rate as a function of water temperature and fish weight.

The most important commercial fishery in the upper Laurentian Great Lakes is the lake whitefish *Coregonus clupeaformis* fishery (Madenjian et al. 2006). In spite of this commercial importance, to the best of our knowledge, trophic transfer efficiency of mercury to lake whitefish from its prey has not been measured in the laboratory. Such measurements would be key in assessing risk to humans consuming lake whitefish from the Great Lakes.

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Our objectives in this study were threefold. First, we estimated net trophic transfer of mercury to lake whitefish from its prey, based on a laboratory experiment. Second, we applied the Trudel and Rasmussen (1997) general model for mercury elimination to estimate gross trophic transfer efficiency of mercury to lake whitefish from its prey. Third, we determined the mercury elimination rate value at which gross trophic transfer efficiency of mercury to lake whitefish from its prey was equal to 80%. Fish tissue samples were available from a previous laboratory study on polychlorinated biphenyl (PCB) trophic transfer to lake whitefish (Madenjian et al. 2006). Thus, an opportunity existed to determine trophic transfer efficiency of mercury to lake whitefish from its food.

Materials and Methods

A laboratory experiment to evaluate a bioenergetics model for lake whitefish was conducted at the Great Lakes Science Center (GLSC) during 23 July through 5 December 2003 (Madenjian et al. 2006). Thus, the experiment was 133 days in duration. During July 2003, tanks 1, 2, 3, and 4 received lots of 25, 29, 25, and 25 lake whitefish. The tanks were circular and constructed of fiberglass, and each tank contained a volume of 2,380 L. Iron-filtered well water was continuously pumped into the tanks at a rate of 15 L/min, and water exited each tank through a drain pipe at the same rate. Each lake whitefish was weighed at the start of the experiment, when 10, 14, 10, and 10 lake whitefish were sacrificed from tanks 1, 2, 3, and 4, respectively. Sacrificed fish were frozen at −30°C. Lake whitefish were fed rainbow smelt *Osmerus mordax* each day of the experiment. The amount of food introduced into each tank, the amount of uneaten food in each tank, and the water temperature in each tank were monitored daily. Uneaten food was removed each day. Feeding rate averaged 0.76% of lake whitefish body weight per day. On average, water temperature was 12.4°C during the course of the experiment. Total water hardness ranged from 450 to 550 mg/L, and pH ranged from 8.4 to 8.8. Photoperiod duration was controlled with fluorescent lighting, which was adjusted seasonally to mimic daylight duration in the Great Lakes region. All 15 of the remaining lake whitefish in each tank were weighed at the end of the experiment, and then frozen at −30°C. After the experiment, lake whitefish were composited by stage (start or end) of experiment and tank. Each lake whitefish composite was homogenized in a blender, and homogenized fish tissue samples were stored at −30°C until time of analysis. In addition, ten 50-fish composites of rainbow smelt were homogenized in a blender, and these homogenized fish tissue samples were stored at −30°C.

The fish tissue samples were sent to the State Laboratory of Hygiene at the University of Wisconsin during 2005 for mercury determinations. Fish tissue samples were digested in a block digester using a sulfuric–nitric acid mixture according to the procedure outlined by Pellizzari et al. (1999). Mercury concentrations in the digested samples were determined via atomic fluorescence, following the procedure by Pellizzari et al. (1999). Appropriate quality control measures were taken, including blanks, duplicates, and spikes. The limit of detection was 1 ng/g. Acceptable levels of recovery from spiked samples ranged from 77% to 123%. Duplicate samples were considered acceptable if their relative percent difference was less than 24%. In addition, a standard reference material (tuna fish, Reference Material No. 463, Institute for Reference Materials and Measurements, Retieseweg, B-2440 Geel, Belgium) was used as part of the quality assurance procedure. Total mercury concentration in each of the fish tissue samples was determined. Although the mercury contained within a fish can exist in either the inorganic form or the methylated form, Bloom (1989, 1992) reported that nearly all (>95%) of the mercury in a fish is methylated. Other investigations have shown that methylmercury may not always represent nearly all of the mercury within a fish (Kannan et al. 1998; Weis and Ashley 2007).

To calculate the net trophic transfer efficiency, γ, of mercury to lake whitefish from their food, we used the estimator presented by Madenjian et al. (2000):

$$\gamma = (\Delta\text{Hg body burden})/(\text{amount of Hg ingested})$$

where ΔHg body burden is the increase in the mercury body burden of lake whitefish in the tank during the experiment (in nanograms of mercury), and the amount of Hg ingested is the weight of mercury in the food eaten by the lake whitefish in the tank during the experiment (in nanograms of mercury). Increase in the mercury body burden was calculated as:

$$\Delta\text{Hg body burden} = ([\text{Hg}_f]\text{W}_f) - ([\text{Hg}_i]\text{W}_i)$$

where $[\text{Hg}_f]$ is the average mercury concentration of lake whitefish in the tank at the end of the experiment (in nanograms per gram wet weight), W_f is the average weight of lake whitefish in the tank at the end of the experiment (in grams wet weight), $[\text{Hg}_i]$ is the average mercury concentration of lake whitefish in the tank at the start of the experiment (in nanograms per gram wet weight), and W_i is the average weight of lake whitefish in the tank at the start of the experiment (in grams wet weight). The amount of mercury ingested was calculated by multiplying the cumulative amount of food eaten by the average lake whitefish in the tank by the mercury concentration in the food. We calculated a net trophic transfer efficiency for each tank. We calculated the mean value of net trophic

transfer efficiency by averaging the estimates of γ across the four tanks, and we also calculated the standard error about the mean. Because direct uptake of mercury by fish from the water can be regarded as negligible (Trudel and Rasmussen 2001), we did not account for direct uptake of mercury from the water in estimating trophic transfer efficiencies.

To estimate the gross trophic transfer efficiency, θ, of mercury to lake whitefish from its prey, we used the Trudel and Rasmussen (1997) general model for mercury elimination by fish:

$$\ln(K) = 0.066T - 0.20(\ln(W)) - 5.83$$

where K is the mercury elimination rate coefficient (in day^{-1}), T is water temperature (in °C), and W is fish weight (in grams wet weight). Once K was calculated, then θ was calculated using the following equation developed by Trudel and Rasmussen (1997):

$$\theta = (([Hg_f] - [Hg_i]e^{-(G+K)\Delta t})(G+K)) \Big/ ([Hg_p]I(1 - e^{-(G+K)\Delta t}))$$

where G is growth rate (in grams of growth per gram of fish per day), $[Hg_p]$ is mercury concentration in the prey (in nanograms per gram wet weight), I is the feeding rate (in grams of food per gram of fish per day), and Δt is the duration of the experiment (in days). We calculated a gross trophic transfer efficiency for each tank. We calculated the mean value of gross trophic transfer efficiency by averaging the estimate of θ across the four tanks, and we also calculated the standard error about the mean.

To determine the value of K at which gross trophic transfer efficiency was equal to 0.80, we assumed K was constant across all four tanks. Preliminary calculations indicated that a value of K between 0.000700 and 0.000800 day^{-1} would yield a gross trophic transfer efficiency of about 0.80. Therefore, we considered all values of K between 0.000700 and 0.000800 day^{-1}, in increments of 0.000001 day^{-1}. For each value of K, we calculated θ for each of the four tanks using the Trudel and Rasmussen (1997) estimator given above. We then averaged θ across all four tanks. We selected the value of K that yielded an average value of θ that was within 0.0001 of 0.80.

Results and Discussion

Mercury concentrations in the rainbow smelt composites ranged from 11 to 17 ng/g, with a mean value of 12.9 ng/g. The standard error about this mean was 0.6 ng/g. Initial mercury concentrations in the lake whitefish ranged from 18 to 20 ng/g (Table 1). Final mercury concentrations in the lake whitefish ranged from 22 to 25 ng/g.

Estimates of net trophic transfer efficiency of mercury to lake whitefish from their food ranged from 0.476 to 0.816 (Table 1). Mean net trophic transfer efficiency of mercury to lake whitefish from their food was 0.635, and the standard error about this mean was 0.081.

Based on the Trudel and Rasmussen (1997) general model for mercury elimination, estimates of gross trophic transfer efficiency of mercury to lake whitefish from their food ranged from 0.776 to 1.197 (Table 1). Mean gross trophic transfer efficiency of mercury to lake whitefish from their food was 1.029, and the standard error about this mean was 0.090. Because the estimated mean gross trophic transfer efficiency exceeded 100%, it appeared that the Trudel and Rasmussen (1997) model was overestimating mercury elimination rate.

When mercury elimination rate, K, was assigned a value of 0.000730 day^{-1}, average gross trophic transfer efficiency was equal to 0.800. At this value of K, estimates of gross trophic transfer efficiency for tanks 1, 2, 3, and 4 were 0.602, 0.872, 0.980, and 0.745, respectively.

Taking into account water temperature and fish size, our estimate of mercury elimination rate was substantially lower than previous estimates of mercury elimination rates for fish. The Trudel and Rasmussen (1997) model, which was developed from a set of published values for mercury elimination rates for fish, predicted a mean K value of 0.00174 day^{-1} for lake whitefish in our four laboratory tanks. Thus, our estimate of K was 2.39 times lower than that predicted by the Trudel and Rasmussen (1997) model.

Our estimate of K corroborated the recent findings by Van Walleghem et al. (2007). Using isotopically enriched methylmercury, Van Walleghem et al. (2007) estimated the mercury elimination rate for yellow perch in the field to be 0.00142 day^{-1}. Based on the average yellow perch weight

Table 1 Estimates of net (γ) and gross (θ) trophic transfer efficiencies of mercury to lake whitefish from their food

Tank number	Average initial weight of lake whitefish (g)	Average final weight of lake whitefish (g)	Average initial [Hg] of lake whitefish (ng/g)	Average final [Hg] of lake whitefish (ng/g)	Consumption (g)	Amount of mercury ingested (ng)	γ	θ
1	713	897	19	22	1,010	13,009	0.476	0.776
2	736	886	18	24	865	11,136	0.724	1.086
3	841	996	18	25	928	11,948	0.816	1.197
4	726	796	20	23	559	7,202	0.524	1.057

(6 g) and average water temperature experienced by the yellow perch (11.4°C), the Trudel and Rasmussen (1997) model predicted a value of K equal to 0.00439 day^{-1}. Thus, the Van Walleghem et al. (2007) estimate of K was 3.09 times lower than that predicted by the Trudel and Rasmussen (1997) model. Further, when standardized by fish size and water temperature, our estimate of K was within 23% of the Van Walleghem et al. (2007) estimate of K. Van Walleghem et al. (2007) attributed the overestimation of K in previous studies to the duration of the experiments being insufficiently long. Most of the experiments upon which the Trudel and Rasmussen (1997) general model was based were less than 70 days in duration. In contrast, our experiment was 133 days in duration.

For modelers assigning a value of 0.80 for gross growth efficiency of mercury to fish from its prey, we recommend that the predictions from the Trudel and Rasmussen (1997) model be adjusted according to our results and the results of Van Walleghem et al. (2007). The ratio of our estimate of K to the predicted K from the Trudel and Rasmussen (1997) model was 0.419. The ratio of the Van Walleghem et al. (2007) estimate of K to the predicted K from the Trudel and Rasmussen (1997) model was 0.323. The average of these two ratios was 0.371. Until a set of realistic estimates of K over a range of fish sizes and water temperatures becomes available, we propose that the predictions of the Trudel and Rasmussen (1997) model be multiplied by 0.371 to estimate mercury elimination rate for fish.

Our findings, coupled with the findings of Van Walleghem et al. (2007), had implications for the application of the mercury tracer technique to estimate food consumption by fish. Trudel and Rasmussen (2001, 2006) have advocated using mercury as a tracer to estimate the amount of food eaten by fish in lakes and rivers, and they have incorporated the Trudel and Rasmussen (1997) general model for mercury elimination into their mercury tracer methodology. Based on the estimates of K from our study and the Van Walleghem et al. (2007) study, the Trudel and Rasmussen (1997) model overestimates mercury elimination rate by a factor of about 2.7. We have calculated that this overestimation of mercury elimination rate would lead to a 30% overestimation of food consumption by the lake whitefish during our laboratory experiment.

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