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Trophic Transfer Efficiency of Methylmercury and Inorganic Mercury to Lake Trout *Salvelinus namaycush* from Its Prey

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Abstract Based on a laboratory experiment, we estimated the net trophic transfer efficiency of methylmercury to lake trout *Salvelinus namaycush* from its prey to be equal to 76.6 %. Under the assumption that gross trophic transfer efficiency of methylmercury to lake trout from its prey was equal to 80 %, we estimated that the rate at which lake trout eliminated methylmercury was 0.000244 day^{-1}. Our laboratory estimate of methylmercury elimination rate was 5.5 times lower than the value predicted by a published regression equation developed from estimates of methylmercury elimination rates for fish available from the literature. Thus, our results, in conjunction with other recent findings, suggested that methylmercury elimination rates for fish have been overestimated in previous studies. In addition, based on our laboratory experiment, we estimated that the net trophic transfer efficiency of inorganic mercury to lake trout from its prey was 63.5 %. The lower net trophic transfer efficiency for inorganic mercury compared with that for methylmercury was partly attributable to the greater elimination rate for inorganic mercury. We also found that the efficiency with which lake trout retained either methylmercury or inorganic mercury from their food did not appear to be significantly affected by the degree of their swimming activity.

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One of the most important factors affecting contaminant accumulation in fish is the efficiency with which fish retain contaminants from their prey (Thomann 1989; Madenjian et al. 1994, 1998a). Although reliable estimates of trophic transfer efficiency are needed for modeling contaminant levels in fish and for providing predictions of exposure risk to people and wildlife eating contaminated fish (Calabrese and Baldwin 1993), trophic transfer efficiencies are not often measured using prey typically eaten by the predator in the predator's native habitat.

The efficiency with which the contaminant in the food ingested by the predator is taken up through the gut wall and incorporated into the body of the predator is referred to as "gross trophic transfer efficiency". After the incorporation of a quantity of contaminant into the body of the predator, a portion of this quantity may eventually be eliminated from the predator's body, and/or a portion of this quantity may be metabolically transformed into another chemical compound. The efficiency with which the contaminant in the food ingested by the predator is retained by the predator, including losses due to elimination and metabolic transformation, is referred to as "net trophic transfer efficiency" (Thomann and Connolly 1984).

Mercury is a persistent contaminant that can impair neurological system structure and function in humans, fish, and wildlife (Bakir et al. 1973; Wren 1986; Scheuhammer et al. 2007; Sandheinrich et al. 2011). Mercury contamination in fish is of particular concern because consumption of contaminated fish represents the primary source of mercury exposure to humans and fish-eating wildlife (Wren 1986). For most mathematical models that have been

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developed for methylmercury accumulation in fishes, gross trophic transfer efficiency is assumed to be 80 % (Norstrom et al. 1976; Rodgers 1994; Trudel and Rasmussen 2001), which corresponds with the mode of laboratory-derived values for fish that are fed natural prey items (Trudel et al. 2000). Although this assumption is important in balancing the fish's mercury "budget", only a few controlled laboratory measurements of trophic transfer efficiency of mercury to fish from its food have been made (Trudel and Rasmussen 2001). In addition, to our knowledge, simultaneous estimation of trophic transfer efficiencies of both methylmercury and inorganic mercury in the laboratory has not been attempted. Furthermore, to our knowledge, the effects of fish activity on trophic transfer efficiency of mercury have yet to be investigated. Although 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. However, other investigations have shown that methylmercury may not always represent nearly all of the mercury within a fish and that the percentage of methylmercury of the total mercury content within a fish can range from 18 to 100 % (Kannan et al. 1998; Weis and Ashley 2007; Raymond and Rossmann 2009). Thus, it is important to consider both fractions of mercury in elimination studies, especially considering that fish eliminate inorganic mercury from their bodies at a rate 2.8 times greater than the rate at which they eliminate methylmercury (Trudel and Rasmussen 1997).

Findings from recent studies have suggested that the rate at which fish eliminate mercury from their bodies has been overestimated. Trudel and Rasmussen (1997) used published estimates of methylmercury elimination rates to develop a general model for methylmercury elimination by fish. Using multiple linear regression, these researchers developed two predictive equations for methylmercury elimination rate as a function of water temperature and fish weight: one equation for acute exposure (exposure from a single dose) to mercury and one equation for chronic exposure (exposure from continued dosing over time) to mercury. For modeling accumulation of methylmercury by fish in lakes, rivers, and reservoirs, the chronic exposure equation has typically been applied. Using isotopically enriched mercury, Van Walleghem et al. (2007) estimated the elimination rate of methylmercury by yellow perch *Perca flavescens* in the field. Their field estimate for yellow perch was approximately three times lower than that predicted by the chronic exposure equation, whereas the prediction from the acute exposure equation showed better agreement (overestimation by <50 %) with the actual field estimate of elimination rate of methylmercury by yellow perch. Based on a laboratory experiment in which lake whitefish *Coregonus clupeaformis* were fed rainbow smelt *Osmerus mordax*, and assuming a gross trophic transfer efficiency of 80 %, Madenjian and O'Connor (2008) estimated that lake whitefish eliminated mercury at a rate of 0.000730 day^{-1}. In contrast, the chronic exposure equation yielded an estimate of mercury elimination by the lake whitefish in this laboratory experiment of 0.00174 day^{-1}, a value 2.4 times greater than the estimate by Madenjian and O'Connor (2008). The similarity of the results from the studies by Van Walleghem et al. (2007) and by Madenjian and O'Connor (2008) provided corroborating evidence that methylmercury elimination rates may have been overestimated in previous studies.

Historically, the lake trout *Salvelinus namaycush* has been the top native predator in the Laurentian Great Lakes, and lake trout populations have supported valuable commercial and recreational fisheries in North America (MacCrimmon and Gots 1980; Martin and Olver 1980; Hansen 1999). Mercury contamination of these lake trout fisheries has remained a significant issue (Mohapatra et al. 2007). Despite the ecological and economic importance of lake trout populations, to the best of our knowledge, trophic transfer efficiency of mercury to lake trout from its prey has not been measured in the laboratory. Such a measurement is key in supporting risk assessments for humans consuming lake trout.

Our objectives in this study were four-fold. First, we estimated net trophic transfer efficiencies of both methylmercury and inorganic mercury to lake trout from its prey based on a laboratory experiment. Second, we applied the regression model of Trudel and Rasmussen (1997) for methylmercury elimination to estimate gross trophic transfer efficiency of methylmercury to lake trout from its prey. Third, we determined the value of the methylmercury elimination rate at which gross trophic transfer efficiency of methylmercury to lake trout from its prey was equal to 80 %. We were especially interested in whether our lake trout laboratory experiment results would agree with the findings from Van Walleghem et al. (2007) and Madenjian and O'Connor (2008) for yellow perch and lake whitefish. Fourth, we determined whether the level of fish activity had a significant effect on net trophic transfer efficiency of both methylmercury and inorganic mercury to lake trout from its prey.

Materials and Methods

To evaluate the effect of fish activity on performance of a lake trout bioenergetics model, a laboratory experiment was conducted at the Great Lakes Science Center from February 16 through July 1, 2010 [Madenjian et al. (in press)]. Thus, the total duration of the experiment was 135 days. All tanks were circular and constructed of

fiberglass. Each of tanks no. 1 through no. 4 contained a volume of 2,380 L, and an average flow rate of 15.8 cm/s was maintained in these four tanks using centrifugal pumps. Each of tanks no. 5 through no. 8 contained a volume of 870 L, and the flow rate in these four tanks averaged only 1.9 cm/s. The difference in flow rates between the two sets of tanks was aimed at forcing lake trout to be relatively active in the larger tanks, while allowing lake trout to be relatively inactive in the smaller tanks [Madenjian et al. (in press)]. Iron-filtered well water was continuously pumped into the 2,380 and 870 L tanks at rates of 15 and 5 L/min, respectively, and water exited each tank through a drain pipe at the same rate it entered each tank. During September 2009, each of the 8 tanks was stocked with 14–19 lake trout. Four to 9 lake trout from each tank were killed on February 16, 2010, so that 10 lake trout remained in each of the tanks at the start of the experiment. Killed fish were weighed and then frozen at −30 °C. In addition, each of the 10 remaining lake trout in each tank was weighed at the start of the experiment. On each day of the experiment, lake trout were fed thawed bloaters *Coregonus hoyi*, which had been caught in Lake Michigan during September 2009 and May 2010. Bloaters were chosen as the food for the lake trout because bloaters are native prey for lake trout from the Laurentian Great Lakes (Madenjian et al. 1998b). After thawing, bloaters were cut into pieces, with each piece weighing between 1 and 5 g. Lake trout were fed as much as they would consume during one feeding each day. The type (September- or May-caught bloaters) and 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 1.2 % of lake trout body weight per day. On average, water temperature was 9.0 °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. The photoperiod duration was controlled with fluorescent lighting, which was adjusted seasonally to mimic daylight duration in the Great Lakes region. All 10 of the remaining lake trout in each tank were weighed at the end of the experiment and then frozen at −30 °C. After the experiment, lake trout were composited by stage (start or end) of experiment and tank. Each lake trout composite was homogenized in a blender, and homogenized fish tissue samples were stored at −30 °C until time of analysis. In addition, 10 3-fish composites of September-caught bloaters and 10 6-fish composites of May-caught bloaters were homogenized in a blender, and these homogenized fish tissue samples were stored at −30 °C.

The fish tissue samples were sent to the United States Geological Survey (USGS) Mercury Research Laboratory in Middleton, WI (http://wi.water.usgs.gov/mercury-laboratory) during 2011 for methylmercury and total mercury determinations. Total mercury determinations were conducted using a modification of method 1631 of the United States Environmental Protection Agency (USEPA 2002). Briefly, homogenized tissue samples were first freeze dried, and the dried material was homogenized using a stainless steel ball mill. Triplicate analysis of individual subsamples were then conducted for each sample by way of solubilization and oxidation in a block digester using a sulfuric-nitric acid mixture, with subsequent addition of bromine monochloride (BrCl), and heating in an oven at 65 °C for 12 h. After digestion, the aqueous-phase of the sample was first prereduced with hydroxylamine hydrochloride (to neutralize the BrCl) and then introduced to the gas-liquid separaton/cold vapor atomic fluorescence spectrometry (CVAFS) instrument. In this process, the oxidized mercury (HgII) in the sample was reduced to gaseous elemental mercury (Hg0) by the automated introduction of $SnCl_2$ and then stripped from the aqueous solution by sparging with ultra-pure argon gas that carries the gaseous mercury to a gold trap, which was then thermally desorbed and carried in the argon stream to the CVAFS detector. Quality-assurance and -control protocols were used throughout sample analysis and included laboratory practices to prevent sample contamination as well as analysis of analytical blanks, sample replicates, and standard reference materials (SRMs). Relative standard deviation (RSD) for each triplicate analysis was calculated and ranged from 0.1 to 11 % (mean 3 %). The limit of detection based on daily detection limit calculations was 1 ng/g. Acceptable levels of recovery from spiked samples ranged from 77 to 123 %. In addition, an SRM (International Atomic Energy Agency Reference Material 407) was used as part of the quality-assurance procedure and included in every sample set.

Methylmercury determination was conducted using the dilute nitric acid–extraction procedures of Hammerschmidt and Fitzgerald (2006). This method is an adaptation of USEPA (1998) method 1630, whereby dilute nitric acid is used in the extract step instead of the distillation procedure called for in method 1630. Briefly, approximately 25–150 mg freeze-dried and homogenized tissue was weighed into Teflon digestion tubes and extracted in 10 mL 5 M HNO_3 at 60 °C for 8 h. Next, approximately 150 µL sample extract and an equivalent volume of 4.5 M KOH was added to 42 mL glass vials, and 50 µL sodium tetraethylboarate (NaTEB) was added as a derivitization agent, resulting in the conversion of methylmercury to methylethylmercury (MeEeHg). Due to its volatility, the MeEeHg is stripped from the liquid phase with argon gas, retained on Tenex traps, desorbed back into the sample stream, and separated from the inorganic forms of mercury in the sample with a gas chromatography column. Next, the individual mercury species were reduced to elemental

mercury and quantified using a CVAFS detector. The limit of detection based on daily detection limit calculations was 2 ng/g. All mercury concentrations were expressed on a wet-weight basis. Subtracting methylmercury concentration from total mercury concentration yielded the inorganic mercury concentration.

To calculate the net trophic transfer efficiency, γ, of both methylmercury and inorganic mercury to lake trout from their food, we used the estimator presented by Madenjian et al. (2000) as folllows:

$$\gamma = (\Delta \text{ Hg body burden})/(\text{amount of Hg ingested}) \quad (1)$$

where Δ Hg body burden is the increase in the mercury (either methylmercury or inorganic mercury) body burden of lake trout in the tank during the experiment (in nanograms of mercury), and the amount of Hg (either methylmercury or inorganic mercury) ingested is the weight of mercury in the food eaten by the lake trout in the tank during the experiment (in nanograms of mercury). Increase in the mercury body burden was calculated as follows:

$$\Delta \text{ Hg body burden} = ([\text{Hg}_\text{f}]W_\text{f}) - ([\text{Hg}_\text{i}]W_\text{i}) \quad (2)$$

where $[Hg_f]$ is the average mercury concentration of lake trout in the tank at the end of the experiment (in nanograms of mercury per gram wet weight), W_f is the average weight of lake trout in the tank at the end of the experiment (in grams wet weight), $[Hg_i]$ is the average mercury concentration of lake trout in the tank at the start of the experiment (in nanograms of mercury per gram wet weight), and W_i is the average weight of lake trout 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 trout in the tank by the mercury concentration in the food. We calculated a net trophic transfer efficiency for both methylmercury and inorganic mercury for each tank. For both methylmercury and inorganic mercury, we calculated the mean value of net trophic transfer efficiency by averaging the estimates of γ across the eight tanks, and we also calculated the standard error (SE) about the mean. We did not account for direct uptake of mercury from the water in estimating trophic transfer efficiencies. Some researchers have characterized the direct uptake of mercury from the water as a negligible component of the fish's mercury budget (Trudel and Rasmussen 2001); however, recent research using stable mercury isotopes has showed that direct uptake of mercury from the water may account for $\geq$10 % of the mercury accumulated by fish in certain lakes (Hrenchuk et al. 2012). To determine whether net trophic transfer efficiency of both methylmercury and inorganic mercury to lake trout from its prey differed significantly between active and inactive lake trout, we applied a two-sample Student t test to compare mean γ for the larger tanks with mean γ for the smaller tanks.

To estimate gross trophic transfer efficiency, θ, of methylmercury to lake trout from its prey, we used both of the regression equations developed by Trudel and Rasmussen (1997) for methylmercury elimination by fish. One set of θ estimates was generated using the chronic exposure equation, and a second set of θ estimates was generated using the acute exposure equation. The chronic exposure equation was as follows:

$$\ln(K) = 0.066T - 0.20(\ln(W)) - 5.83 \quad (3)$$

where K is the methylmercury elimination rate coefficient (in day^{-1}), T is water temperature (in °C), and W is fish weight (in grams wet weight). The acute exposure equation was as follows:

$$\ln(K) = 0.066T - 0.20(\ln(W)) - 6.56 \quad (4)$$

Once K was calculated, then θ was calculated using the following equation developed by Trudel and Rasmussen (1997) as follows:

$$\theta = \left(\left([\text{Hg}_\text{f}] - [\text{Hg}_\text{i}]\text{e}^{-(G+K)\Delta t}\right)(G + K)\right) / \left(\left[\text{Hg}_\text{p}\right]\text{I}\left(1 - \text{e}^{-(G+K)\Delta t}\right)\right) \quad (5)$$

where G is growth rate (in grams of growth per gram of fish per day), $[Hg_p]$ is methylmercury 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 eight tanks, and we also calculated the SE about the mean.

To determine the value of K at which gross trophic transfer efficiency equaled 0.80, we assumed that K was constant across all eight tanks. Preliminary calculations showed that a value of K between 0.000200 and 0.000300 day^{-1} would yield a gross trophic transfer efficiency of approximately 0.80. Therefore, we considered all values of K between 0.000200 and 0.000300 day^{-1} in increments of 0.000001 day^{-1}. We calculated θ for each of the eight tanks, using the Trudel and Rasmussen (1997) estimator given previously, for each value of K. We then averaged θ across all eight tanks. The value of K that yielded an average value of θ within 0.0001 of 0.80 was chosen as the estimate of K.

To estimate the gross trophic transfer efficiency of inorganic mercury to lake trout from its prey, we used the following procedure. We estimated K for inorganic mercury by multiplying the previously mentioned estimate of K for methylmercury by 2.8 because Trudel and Rasmussen (1997) estimated that fish eliminate inorganic mercury

at a rate 2.8 times greater than that for methylmercury. Then, we used the estimator of Trudel and Rasmussen (1997) for θ presented previously to estimate gross trophic transfer of inorganic mercury to lake trout from its prey for each of the eight tanks. We then averaged θ across the eight tanks to yield an estimate of gross trophic transfer efficiency of inorganic mercury to lake trout from its prey.

Results and Discussion

Methylmercury concentrations in the September-caught bloater composites ranged from 72 to 93 ng/g (mean 80.0) and an SE about the mean of 2.3 ng/g. Methylmercury concentrations in the May-caught bloater composites ranged from 46 to 95 ng/g (mean 76.2) and an SE about the mean of 5.5 ng/g. Initial methylmercury concentrations in the lake trout ranged from 16 to 45 ng/g (Table 1). Final methylmercury concentrations in the lake trout ranged from 64 to 131 ng/g. On average, 84.8 and 86.7 % of the total mercury found in September- and May-caught bloater composites, respectively, was methylmercury. On average, 81.7 and 86.0 % of the total mercury found in the lake trout at the start of the experiment and at the end of the experiment, respectively, was methylmercury.

Estimates of net trophic transfer efficiency of methylmercury to lake trout from their food ranged from 0.623 to 0.943 (Table 1). Net trophic transfer efficiency averaged 0.775 (SE = 0.051) and 0.758 (SE = 0.067) for active and inactive lake trout, respectively. Furthermore, net trophic transfer efficiency did not differ significantly between active and inactive lake trout ($t = -0.20$; $df = 6$; $P = 0.8464$), demonstrating that activity did not have a significant effect on the efficiency with which lake trout retain methylmercury from their food. Averaging across all eight tanks, mean net trophic transfer efficiency of methylmercury to lake trout from their food was 0.766, and the SE about this mean was 0.039.

Inorganic mercury concentrations in the September-caught bloater composites ranged from 12 to 17 ng/g (mean 14) and an SE about the mean of 0.47 ng/g. Inorganic mercury concentrations in the May-caught bloater composites ranged from 8 to 14 ng/g (mean 12) and an SE about the mean of 0.75 ng/g. Initial inorganic mercury concentrations in the lake trout ranged from 4 to 9 ng/g (Table 2). Final inorganic mercury concentrations in lake trout ranged from 14 to 23 ng/g.

Estimates of net trophic transfer efficiency of inorganic mercury to the lake trout from their food ranged from 0.423 to 0.828 (Table 2). Net trophic transfer efficiency averaged 0.603 (SE = 0.093) and 0.668 (SE = 0.079) for active and inactive lake trout, respectively. Moreover, net trophic transfer efficiency did not differ significantly between active and inactive lake trout ($t = 0.53$; $df = 6$; $P = 0.6156$); therefore, activity did not appear to have a significant effect on the efficiency with which lake trout retain

Table 2 Estimates of net trophic transfer efficiency (γ) of inorganic mercury to lake trout from their food

Tank no.	Average initial [InorgHg] of lake trout (ng/g)	Average final [InorgHg] of lake trout (ng/g)	Amount of [InorgHg] ingested (μg)	γ
1	9	18	24	0.683
2	9	16	28	0.481
3	8	23	32	0.826
4	9	15	37	0.423
5	9	15	26	0.452
6	8	14	9	0.724
7	7	16	17	0.667
8	4	17	18	0.828

InorgHg inorganic mercury

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

Tank no.	Average initial weight of lake trout (g)	Average final weight of lake trout (g)	Average initial [MeHg] of lake trout (ng/g)	Average final [MeHg] of lake trout (ng/g)	Consumption (g)	Amount of MeHg ingested (μg)	γ	θ (chronic)[a]	θ (acute)[b]
1	907	1,345	41	91	1,734	138	0.623	0.733	0.680
2	860	1,339	40	125	1,999	159	0.836	0.963	0.904
3	890	1,518	35	123	2,344	186	0.831	0.949	0.898
4	817	1,566	43	131	2,649	210	0.809	0.934	0.884
5	694	1,242	45	116	1,870	149	0.763	0.886	0.833
6	729	853	31	64	641	51	0.635	0.777	0.704
7	754	1,050	35	88	1,203	95	0.690	0.811	0.751
8	729	1,092	16	103	1,336	106	0.943	1.060	1.006

[a] Based on chronic exposure equation by Trudel and Rasmussen (1997) for methylmercury elimination rate

[b] Based on acute exposure equation by Trudel and Rasmussen (1997) for methylmercury elimination rate

inorganic mercury from their food. Averaging across all eight tanks, mean net trophic transfer efficiency of inorganic mercury to lake trout from their food was 0.635, and the SE about this mean was 0.058.

Using the chronic exposure equation, estimates of K for methylmercury ranged from 0.001282 to 0.001372 day^{-1} (Table 3) (mean 0.001332). Using the acute exposure equation, estimates of K for methylmercury ranged from 0.000618 to 0.000661 day^{-1} (Table 3) (mean 0.000642).

Based on the chronic exposure equation for methylmercury elimination of Trudel and Rasmussen (1997), estimates of gross trophic transfer efficiency of methylmercury to the lake trout from their food ranged from 0.733 to 1.060 (Table 1) (mean 0.889) and an SE about the mean of 0.039. Based on the acute exposure equation for mercury elimination, estimates of gross trophic transfer efficiency of methylmercury to lake trout from their food ranged from 0.680 to 1.006 (Table 1) (mean 0.832) and an SE about the mean of 0.040. Thus, both values of mean θ were >0.80, although mean θ based on the acute exposure equation was substantially closer to 0.80 than mean θ based on the chronic exposure equation.

When methylmercury elimination rate, K, was assigned a value of 0.000244 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 no. 1 through no. 8 were 0.650, 0.870, 0.869, 0.855, 0.803, 0.661, 0.717, and 0.974, respectively.

Under the assumption of the gross trophic transfer efficiency of methylmercury to the lake trout from its food was equal to 0.80, the chronic exposure equation overestimated mercury elimination rate for lake trout in our laboratory tanks by a factor of 5.46. Again under the assumption that θ equals 0.80, the acute exposure equation overestimated mercury elimination rate for lake trout in our laboratory tanks by a factor of 2.63. Thus, the acute exposure equation appeared to be substantially more accurate than the chronic exposure equation in predicting mercury elimination rate. Nevertheless, our analysis results suggested that even the acute exposure equation may be overestimating methylmercury elimination rate.

Table 3 Estimates of methylmercury elimination rate (K) for lake trout based on regression model by Trudel and Rasmussen (1997)

Tank no.	K (day^{-1}) Chronic[a]	K (day^{-1}) Acute[b]
1	0.001317	0.000635
2	0.001346	0.000649
3	0.001282	0.000618
4	0.001300	0.000627
5	0.001311	0.000632
6	0.001369	0.000660
7	0.001372	0.000661
8	0.001357	0.000654

[a] Based on chronic exposure equation

[b] Based on acute exposure equation

Based on the assumption that the elimination rate for inorganic mercury was 2.8 times greater than the elimination rate for methylmercury (Trudel and Rasmussen 1997), we estimated the gross trophic transfer efficiency of inorganic mercury to lake trout from its prey to be equal to 0.702. We have shown that the net trophic transfer efficiency to lake trout from its prey for inorganic mercury was considerably lower than that for methylmercury. This lower net trophic transfer efficiency was partly attributable to the elimination rate for inorganic mercury being substantially greater than the elimination rate for methylmercury. In addition, this lower net trophic transfer efficiency may have been partly due to a lower gross trophic transfer efficiency for inorganic mercury compared with that for methylmercury.

Our results were in good agreement with the results of Van Walleghem et al. (2007) and Madenjian and O'Connor (2008), with all three studies indicating that methylmercury elimination rates have been overestimated in the past. Using isotopically enriched mercury, Van Walleghem et al. (2007) estimated that juvenile yellow perch in the field eliminated methylmercury at a rate of 0.00142 day^{-1}, which was 3.09 times lower than the elimination rate predicted by the chronic exposure equation but only 1.49 times lower than the elimination rate predicted by the acute exposure equation. Van Walleghem et al. (2007) concluded that the chronic exposure equation significantly overestimated the mercury elimination rate by the yellow perch and that the acute exposure equation predicted elimination rate with considerably greater accuracy than the chronic exposure equation. Yet, the analysis results by Van Walleghem et al. (2007) provided evidence that the acute exposure equation was slightly overestimating methylmercury elimination rate. In a laboratory study, Madenjian and O'Connor (2008) estimated that lake whitefish eliminate mercury at a rate of 0.000730 day^{-1}, which was 2.39 times lower than the elimination rate predicted by the chronic exposure equation but only 1.15 times lower than the elimination rate predicted by the acute exposure equation. Madenjian and O'Connor (2008) concluded that the chronic exposure equation was substantially overestimating elimination rate, but again there was some evidence that the acute exposure equation was slightly overestimating methylmercury elimination rate. Our results indicated that both the chronic and the acute exposure equations overestimate elimination rate. Computing an average based on the results of our study and the studies by Van Walleghem et al. (2007) and Madenjian and O'Connor (2008),

the chronic exposure equation overestimated methylmercury elimination rate by a factor of 3.6.

Until a set of more accurate estimates of methylmercury elimination rate over wide ranges of both fish size and water temperature becomes available, we suggest that the predictions of the chronic exposure equation by Trudel and Rasmussen (1997) be divided by 3.6 to provide reasonably accurate estimates of methylmercury elimination rate. Modeling of methylmercury accumulation in fish has been used both in assessing risk to humans and wildlife that eat contaminated fish and in estimating food consumption by fish (Trudel et al. 2000; Pastorok et al. 2002). To provide a more realistic representation of methylmercury accumulation by fish with these modeling applications, we recommend this adjustment to the predictions of methylmercury elimination rate from the chronic exposure equation.

Our findings, in conjunction with results from the Van Walleghem et al. (2007) and Madenjian and O'Connor (2008), suggested that re-evaluation of the techniques for estimating mercury elimination rates in fish is needed. One of the pitfalls of estimating mercury elimination rates has been not allowing a sufficiently long duration of the experiment to accurately measure the rate. Trudel and Rasmussen (1997) concluded that analysis of results from short-term experiments (<90 day duration) overestimated mercury elimination rate in fish by a factor of more than 7, on average, compared with results based on long-term experiments (>90 day duration). These researchers attributed this overestimation to not having a sufficiently long-term series of observations to adequately distinguish between the fast and slow components of elimination. Another pitfall of estimating mercury elimination rates has been using mercury concentration in muscle tissue as a surrogate for the whole-fish mercury concentration. Although there is a general one-to-one correspondence between mercury concentration in muscle tissue of fish and whole-fish mercury concentration (Trudel and Rasmussen 2001), departures from this general rule have been noted (Becker and Bigham 1995). Certainly, changes in muscle mercury body burden over time did not always closely track changes in whole-fish mercury body burden for yellow perch during the field experiment conducted by Van Walleghem et al. (2007). Consequently, using muscle mercury concentrations as a surrogate for whole-fish mercury concentrations could potentially lead to error in estimating the rate at which yellow perch eliminated mercury from their bodies. Given the slow mercury elimination rates for fish, estimation of elimination rate based on muscle tissue determinations of mercury concentration may not be a sufficiently accurate technique. One additional problem with estimating mercury elimination rates in fish has been with the method chosen to spike a fish with isotopically enriched mercury. This may have led to elimination of the spiked mercury in such a manner that it did not accurately mimic the natural process of mercury elimination in fish. For example, Ruohtula and Miettinen (1975) spiked rainbow trout *Oncorhynchus mykiss* in the laboratory by injecting the isotopically enriched mercury either directly into the stomach by way of a plastic catheter or into the dorsal muscle. This injection method of administering the isotopically enriched mercury may have introduced artifacts that did not allow for accurate characterization of the rate at which mercury is eliminated by fish in a natural setting. In contrast, Van Walleghem et al. (2007) spiked the yellow perch in their study by adding the isotopically enriched mercury, in solution, to an entire ecosystem, thus allowing the mercury to be incorporated into the food web during a period of several years through natural processes and pathways (Harris et al. 2007).

Conclusion

To our knowledge, our study represented the first attempt to: (1) simultaneously estimate trophic transfer efficiencies to fish from their food for both methylmercury and inorganic mercury in the laboratory, (2) estimate trophic transfer efficiencies of mercury to lake trout from its prey, and (3) determine the effects of swimming activity of the fish on trophic transfer efficiencies for mercury. Net trophic transfer efficiency of methylmercury to lake trout from its prey was estimated to be equal to 0.766, and the net trophic transfer efficiency for methlymercury did not appear to be significantly influenced by the degree of swimming activity by the lake trout. Net trophic transfer efficiency of inorganic mercury to lake trout from its prey was estimated to be equal to 0.635, and the net trophic transfer efficiency for inorganic mercury also did not appear to be significantly affected by the degree of swimming activity by the lake trout. The lower net trophic transfer efficiency for inorganic mercury compared with that for methylmercury was partly due to the greater elimination rate for inorganic mercury, but it also may have been partly due to a lower gross trophic transfer efficiency for inorganic mercury compared with that for methylmercury. Assuming that gross trophic transfer efficiency for methylmercury was equal to 0.80, we estimated that the rate of methylmercury elimination by the lake trout was equal to 0.000244 day^{-1}. Our results, in conjunction with results from two other recent studies, suggested that mercury elimination rates for fish have been overestimated in previous studies. We recommended that techniques for estimating the rate of mercury elimination by fish be reevaluated and that certain pitfalls be avoided in measuring these mercury elimination rates for fish.

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