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Methylmercury and Isotopic Analysis of Invertebrates from a Lime Treated Tributary of Lake Honnedaga

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by

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Abstract

Recent publications confirm long term gaseous elemental and oxidized mercury deposition trends within the northeastern United States as decreasing. Ascertaining how mercury dynamics in ecosystems respond to acid rain deposition is important for aquatic ecology, and human exposure. Continually, previously low pH, and recovering Honnedaga Lake in the Adirondack Park in New York is only one of seven lakes with heritage New York State Brook Trout (*Salvelinus fontinalis*). In an effort to accelerate the processes of neutralization, and to identify methylmercury effects within the ecosystem, several tributaries within the Lake's Watershed were treated with lime. Macroinvertebrate samples taken from the lime treated tributary (T6) and reference tributaries (T6A and MBBR) of four functional feeding groups (i.e. predators, omnivores, scrappers, and shredders) were analyzed for methylmercury (ng/g dw) and stable isotopes. Only the omnivore functional feeding group had a significant relationship to methylmercury and $\delta\ ^{15}N$ to support the bioaccumulating metal as a trophic position indicator (ANOVA, $p < 0.05$). Trophic positions were found to be 2.81, 2.58, 2.39, and 2.38 for the predator, omnivore, scrapper, and shredder functional feeding groups, respectively. Methylmercury concentrations in macroinvertebrates only had a significant relationship with years (2013 – 2014) of treatment (ANOVA, $p < 0.05$). Furthermore, methylmercury concentrations were shown to have increased at both reference sites from 2013 to 2014 (ANOVA, $p < 0.05$). Further research is needed to clarify what occurs to macroinvertebrates and the associated food web following a liming application.

Abbreviations

ARP – Acid Rain Program

ANC – acid neutralizing capacity

ANOVA – a one-way analysis of variance

C – carbon

Ca – calcium

$\delta^{13}C$ – delta carbon isotopic signature

DOC – dissolved organic carbon

dw – dry weight

EU – Europe

FFG(s) – functional feeding group(s)

GC – gas chromatography column

Hg – mercury

IAEA – International Atomic Energy Association

SO_x – sulfuric [oxide compounds]

MATS – mercury and toxic substances

MBBR – Middle Branch Brook Reservoir

MeHg – methylmercury

N – nitrogen

ng/g – nanograms per gram

$\delta^{15}N$ – delta nitrogen isotopic signature

NO_x – nitric [oxide compounds]

NYS – New York State

NYS DOH – New York State Department of Health

pH – relative acidity

ppb – parts per billion

ppm – parts per million

THg – total mercury

US – the United States of America

USD – United States Dollars

US EPA (EPA) – United States Environmental Protection Agency

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Geoff – thank you for giving me this project, letting me write this as my Honors Thesis, during the last few days before submission, and encouraging me to think for myself.

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Hold yourself to the fire…

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Introduction

Since the late 1960s and early 1970s, acid rain's or acid deposition's deleterious effects on the environment have been observed (Likens, Bormann, & Johnson, 1972; Oden, 1968). Taking any form of precipitation, acid rain contains SO_x and NO_x compounds which affect aquatic and forest ecosystems in North America, Europe, and Asia (Driscoll et al., 2001). The higher levels of the SO_x and NO_x compounds remove exchangeable base cations within the soil; mobilize inorganic aluminum from soil to water; and decrease ANC in surface waters. The result is acute toxic conditions in aquatic and forest ecosystems (Baldigo, et al., 2007, 2009, 2016; Lawrence et al., 2008).

The SO_x and NO_x compounds creating acid rain stem from both natural and anthropogenic sources, but the percentages of which are hotly debated. Many researchers identify the major contributors to the acid rain compounds to the reformation and manufacturing of oil, vehicles and heavy equipment, and the burning of fossil fuels/coal fired power plants to generate electricity The electric generators contribute two thirds of sulfuric and one fourth of nitric compounds into the atmosphere (U.S. EPA 2011, 2013)

Under the Clean Air Act Amendments in 1990, the USEPA created the ARP which sets annual emission requirements on the fossil fuel-fired power plants across the US on the primary precursors of acid rain (U.S. EPA, 2013). Because of the ARP and other legislative efforts, aquatic and forest ecosystems have observed decreases in acid deposition across the US (Burns et al., 2011; Greaver et al., 2012), and have begun to recover (Driscoll et al. 2007a). Decades of acid rain have slowed biological and ecological recovery in lakes and watersheds because of the elimination of Ca and other

cations from the soil communities (Driscoll et al., 2001; Liu et al., 2014; Warby et al., 2005) with initial improvements in surface water chemistry stemming from decreases in acid rain deposition (Chen and Driscoll, 2005; Driscoll et al., 2001, 2003a, 2007a).

In addition to acid deposition, remote forest ecosystems like the Adirondacks are sensitive to atmospheric Hg deposition (Driscoll et., 2007b; Evers et al., 2007; Yu et., 2011). Global mercury emissions include natural geogenic sources (volcanism and erosion), and anthropogenic sources (fuel combustion, waste incineration, and mining). Anthropogenic sources have increased mercury emissions by a factor of 2 – 15 over natural sources (Driscoll et al., 2013). In the northeastern United States, 60-81% of Hg deposition can be attributed to regional sources by deposition of gaseous or particulate ionic Hg (Pirrone et al., 2010; Seigneur et al., 2004; Selin & Jacob, 2008). Within aquatic ecosystems like in the Adirondacks, DOC has a large role in transporting and making Hg bioavailable (Dittman et al., 2010; Schuster et al., 2008). Eventually, the Hg can be converted to MeHg by iron– or sulfate– reducing bacteria under reducing conditions (Gilmour et al., 1998; Kerin et al., 2006).

In this form, MeHg is more bioavailable too organisms in aquatic web foods, and can be biomagnified up food chains. High concentrations of MeHg have been linked to neurotoxicity in organisms like fish-eating birds, and mammals especially humans. Women under 50 and children under 15 are especially at risk to these negative health impacts of this dangerous toxicant (Bradley et al., 2017; Driscoll et al., 2007b, 2013; Grandjean et al., 2010; Schoch et al., 2011). The gastrointestinal track is the principal route of absorption in organisms. The absorption rate of MeHg ranges from 12 – <95% (Bradley et al., 2017; Cabanero et al., 2004, 2007; Calatayud et al., 2012; He and Wang,

2011; Siedlikowski et al., 2016; and Wang et al., 2013). Once the MeHg enters the lymphatic system, it can cross into the brain, placenta and fetus producing irreversible neurological damage to humans and top–level predators (Hong et al., 2012; Mergler et al., 2007; Schoch et al., 2014). A global health assessment of economic damages of ingesting and inhaling MeHg was hypothesized to be USD 2.9 million annually (Sundseth et al., 2010) while the aggregate US economic benefit of reducing MATS emissions from anthropogenic sources by 2050 could be USD 104 billion (Giang and Selin, 2016).

In NYS, the NYS DOH has issued a statewide consumption advisory to eat no more than one meal (one half pound) of sport fish per week while some regions or individual lakes may have more specific consumption advisory guidelines due to varying concentrations of MeHg like in the Adirondacks and Catskill Parks. These waterways have more strict consumption guidelines on fish because they have even higher Hg levels compared to other lakes (NYS DOH, 2017). A 2008 study of the Adirondack region in NYS showed that Yellow perch's (*Perca flavescens*) median Hg concentrations were more than twice the values observed across the state at 382 ng/g and 162 ng/g, respectively (Simonin, Loukmas, Skinner, and Roy, 2008).

In Northern EU, the northeastern US, and southeastern Canada, aquatic ecosystems are showing signs of biological and ecological recovery from acid deposition (Driscoll et al., 2001; Jeffries et al., 2003; Lovett et al., 2009; US EPA, 2011). According to US EPA (2011), "The evidence is sufficient to infer a causal relationship between sulfur deposition and increased mercury methylation in wetlands and aquatic environments," meaning decreases in acid deposition can lead to decreases in MeHg

concentrations in fish and the prey consumed. MeHg in small lotic ecosystems has been studied before (Bradly et al., 2013; Riva-Murray et al., 2011, 2013), but not within a lime treated tributary.

Despite decreases in US Hg emissions (Drevnick et al., 2012; Zhang et al., 2016; Zhou et al., 2017), Hg concentrations in recovering ecosystems remain elevated (Millard, 2016). This condition presents an ongoing risk of bioaccumulation of this potent neurotoxin. Past studies on direct lake, and watershed lime ($CaCO_3$) application in the Adirondacks; the ongoing addition of wollastonite ($CaSiO_3$) at Hubbard Brook, New Hampshire; and a large-scale liming and monitoring program of streams and lakes within Sweden exemplify the options/evidence available in accelerating the recovery of aquatic ecosystems affected by acid rain (Cho et al., 2010; Driscoll et al., 1996; Eriksson et al., 1983; Holmgren et al., 2016; Millard, 2016).While these studies have shown that lime applications increase pH and ANC, and decrease concentrations of Al^{-}, robust research on post liming effects and MeHg dynamics in small lotic aquatic biota have yet to be addressed especially in lower trophic positions. In this study, I examined the impact of a liming application in a tributary of Honnedaga Lake.

Honnedaga Lake, located in the southwestern Adirondacks, is one of seven Adirondack lakes with a heritage Brook trout (*Salvelinus fontinalis*) population. This genetically unique population has undergone a recovery in recent years in association with increasing lake pH and ANC (Josephson et al. 2014). However, they remain at risk from ongoing effects of acid deposition as well as biomagnifcation, and bioaccumulation of MeHg from lower trophic positions. The addition of lime to Honnedaga Lake tributaries was anticipated to improve and increase spawning habitat for Brook trout. This

experiment provides an opportunity to examine changes in MeHg in food webs after liming a tributary. I hypothesized that higher MeHg concentrations would be associated with higher level tropic positioned organisms. Through observing the MeHg concentrations and associated tropic level positions of macroinvertebrates from liming conditions of this Honnadaga Lake tributary, the picture of the tributaries and the lake's ecological food web could be better understood.

Methods

Study Site

Honnedaga Lake (3.1 km^2) is located in the southwestern Adirondacks (43° 31'06" N and 74°48'31" W). The watershed (13.3 km^2) is completely forested, with 26 tributaries draining into the lake (Figure 1). There were several tributaries studied: T6 as the lime treated tributary, and T6A as well as MBBR as reference tributaries. These were episodically acidic prior to treatment, with pH dropping below 4.8. These watersheds make up a sizeable portion of the entire Honnedaga Lake watershed (T6 – 2.174 km^2, T6A – 1.393 km^2, and MBBR – 2.731 km^2). The treatment watershed received 11 metric tons of limestone ($CaCO_3$), distributed in a pelletized form directly into the stream at a dosage at ~50 kg of Ca/ha which is comparable to application rates for other regional watershed liming studies (Driscoll et al., 1996; Millard, 2016; Peters et al., 2004; and Schmidt & Sharpe, 2002). The lime application will occur in early summer to maximize effects through most of the growing season, and once in late winter to evaluate the

treatment when flows are expected to be highest and stream water the most acidic over several years: October 28th, 2010; July 12th, 2012; June 19th, 2013; March 5th, 2014; June 16th, 2015; and June 21st, 2016. This study focuses predominantly on MeHg, and stable isotope data from 2013, and 2014.

Targeted Taxa

Macroinvertebrates were targeted to collect prey organisms of Brook trout for analysis of MeHg, $\delta^{13}C$, and $\delta^{15}N$. Macroinvertebrate taxa will be collected from the tributary locations. These will include organisms representing four FFGs that are important to the food web of the particular tributary, and will follow Merritt and Cummins (1996), and Thorp and Covich (2001) classifications of FFGs. For macroinvertebrates, the targeted FFGs were shredders (consumers of living and/or decomposing plant material), scrapers (consumers of periphyton and associated biofilm), omnivores (consumers of both plant and animal material), and predators (consumers of other macroinvertebrates). Invertebrates within the previous groups were targeted: shredders–northern case-maker caddisflies, and rolled-winged stoneflies (Trichoptera: Limnephilidae; and Plecoptera: Leuctridae); scrappers–mayflies, fish flies, and hellgrammites (Ephemeroptera: Isonychiidae; and Megaloptrea: Sialidae); omnivores–crayfish and net-spinning caddisflies (Decapoda: Cambaridae; Trichoptera: Hydropsychidae); predators–dragonflies darner, and cordulegaster dragonflies (Odonata: Aeshnidae and Cordulegastrida) (Riva-Murray et al., 2011).

Field Sampling

As described by Riva-Murray (2011), macroinvertebrates were collected by hand-picking, kick-netting, and bank jabbing from all distinct habitat type (rocks, sticks, leaf packs, and depositional areas) with the goal of three samples per taxon per site visit. Field processing of macroinvertebrates was done in accordance with trace–metal clean techniques (detailed in Scudder et al., 2008). Macroinvertebrates were sorted with pre-cleaned plastic forceps, rinsed with de-ionized water, blot dried, weighed, placed in plastic jars, and immediately frozen.

Laboratory Processing and Chemical Analyses

Frozen invertebrate samples were freeze-dried at -80 °C and 0.080 mBar (FreeZone Type 6 plus freeze drier by Labconco) and homogenized either by hand prior or freeze miller (6970D Enclosed Freezer Mill by SPEX Sample Prep LLC). Then, all invertebrate samples were analyzed for MeHg and stable isotopes. MeHg was analyzed by digestion, aqueous ethylation, purge and trap, desorption and cold-vapor atomic fluorescence spectrometry according to U.S. EPA Method 1630 (U.S. EPA 2007). The detection limit for this method is 0.0015ng/L.

Stable isotopes analysis was completed by Cornell University Stable Isotope Laboratory using ThermoFinnigan Delta Plus mass spectrometer. Accuracy and precision of the stable isotope measurements (expressed in the standard per mil notation relative to V-PDB for $\delta^{13}C$ and atmospheric nitrogen for $\delta^{15}N$) were verified by reference materials

provided by the IAEA. Daily precision of the instrument was verified by repeated analyses of internal laboratory standards during the sample runs. Stable isotope composition is expressed in parts per thousand (‰ or 'per mil') as a deviation from a standard material denoted as 'delta' (δ): $\delta^{13}C$ or $\delta^{15}N = ([R_{sample}/ R_{standard}]-1) \cdot 1000$, where $R= {}^{13}C/{}^{12}C$ or ${}^{15}N/{}^{14}N$.

Data and Statistical Analyses

Average trophic position was calculated for functional feeding groups to account for potential site-to-site differences in $\delta^{15}N$ of nitrogen sources. This value was calculated by taking the group mean of the difference between $\delta^{15}N$ of the invertebrate feeding group consistently having the lowest $\delta^{15}N$ across sites (Anderson & Cabana, 2007), applying a fractionation constant of 3.4 (Minagawa & Wada, 1984; Post, 2002), and where λ represents the trophic position of the baseline organism (e.g. is 2 for primary consumers):

$$\text{Trophic position} = \delta^{15}N_{\text{consumer}} - \delta^{15}N_{\text{lowest}} / 3.4 + \lambda \quad (1)$$

ANOVA followed by Tukey's HSD post-hoc multiple comparison tests was used to observe differences between group means in MeHg, $\delta^{15}N$, and estimated trophic position. MeHg concentrations were log-transformed to meet assumptions of normal distribution before ANOVA. Statistical analyses were done in SAS.

Results

The predator (N = 56) FFG was the most wildly collected macroinvertebrates: 26 samples from T6, and 15 from both T6A, and MBBR. The omnivore (N=52) FFG had 29 samples taken from T6, 15 from T6A, and 8 from MBBR. The shredder (N=16) FFG had 7 samples taken from T6, and 9 samples from MBBR. The scrappers (N=9) FFG were the least wildly collected macroinvertebrates: 4 samples from T6, 1 from T6A, and 4 from MBBR.

$\delta^{13}C$, $\delta^{15}N$, and trophic position data

The predator FFG had the highest $\delta^{15}N$ range, and the third highest $\delta^{13}C$ range. The FFG's mean trophic position was 2.81, and did not have a significant relationship to $\delta^{15}N$ based on MeHg concentrations. The omnivore FFG had the lowest $\delta^{15}N$, and $\delta^{13}C$ ranges. The FFG's mean trophic position was 2.58, and did not have a significant relationship to $\delta^{15}N$ based on MeHg concentrations (ANOVA, $p < 0.05$). The shredder FFG had the second highest $\delta^{15}N$, and highest $\delta^{13}C$ ranges. The FFG's mean trophic position was 2.38, and did not have a significant relationship to $\delta^{15}N$ based on MeHg concentrations. The scrapper FFG had the third lowest $\delta^{15}N$, and second highest $\delta^{13}C$ ranges. The FFG's mean trophic position was 2.39, and did not have a significant relationship to $\delta^{15}N$ based on MeHg concentrations. Ranges of $\delta^{15}N$, and $\delta^{13}C$; trophic positions; and FFGs associated with $\delta^{15}N$ and Log_{10} MeHg span across all sites, and years (see Tables 1, 4, and 5, and Figure 2).

MeHg data

The simple Year parameter had a significant relationship to changes in MeHg concentrations (ANOVA, $p < 0.05$). Additionally, the intersect between Site and Year had a significant relationship (ANOVA, $p < 0.2$). This intersect was analyzed further to determine the interaction with the environmental conditions among the sites for the length of this study. It was found that the treated tributary (T6) did not have a significant relationship with increased MeHg concentrations across all FFGs from 2013 to 2013. Both reference sites (T6A and MBBR) from 2013 to 2014 yielded a significant relationship for increased MeHg concentrations across all FFGs (ANOVA, T6A & MBBR $p < 0.05$) (see Table 3 and Figure 3).

Discussion

MeHg and food web connections

While no *Salvelinus fontinalis* were analyzed for MeHg or stable isotopes to help understand food chain length, the results found in this study can be utilized in future research. Trophic position is an important predictor of MeHg values of consumer organisms within streams (Chasar et al., 2009; Ward et al., 2010). Similar to this study's result from the omnivore FFG, Chasar et al. (2009) also observed significant relationships to MeHg and $\delta^{15}N$ as a proxy to trophic positions from seven of the eight streams observed in the US for their FFGs. While FFGs are statistically important to MeHg concentrations, there was no interaction with other predictors. It is not entirely clear why

the reference tributaries (T6A, and MBBR) saw an increase in MeHg concentrations across their FFGs while the treated tributary (T6) did not. The application of lime could have steadied the MeHg dynamics within the food web of T6, and because the reference tributaries did not receive lime they are not subjected to the hypothesized beneficial biological effects. Millard (2016) observed MeHg concentrations in the water matrix of the chronically acidic (pH < 4.8) tributaries T16 (lime treatment) and T24 (reference) which are also located in Honnedaga Lake's watershed. He found that MeHg concentrations in the watershed treatment site had been decreasing prior to treatment in 2013, and then both sites saw significant increases of MeHg in 2016. While Millard's research focuses on water chemistry, in both situations the application of lime may have influenced MeHg in reference tributaries. This dynamic should be investigated further. However, these data should be seen only as snap shot from Honnedaga's recovery monitoring program.

Rebuttal of MeHg macroinvertebrate tributary study

Clayden et al., 2014 preformed canonical RDA testing to increase clarity in identifying specific indicators of MeHg concentrations regarding trophic position for macroinvertebrates in similar FFGs. The researchers found that water pH was the best environmental indicator to MeHg concentrations for both predatory and non-predatory macroinvertebrates ($R^2_{adj} = 0.32$, $p<0.001$). Additionally, location (latitude and longitude) of sites was associated with 15% of the MeHg variation for their predator and primary consumer macroinvertebrate groupings. While it is significant to characterize and

proportion individual factors that can influence MeHg concentrations and trophic position within lake and lotic food webs, the statistical backing behind Clayden et al. results is weak when compared to the articles they argue against. Each ecosystem is different and it is the culmination of a variety of factors including pH that influences MeHg concentrations and a macroinvertebrate's trophic position (Driscoll et al., 2007b; Foster and Sprules, 2010; Riva-Murray et al., 2011; Ward et al., 2010, 2012).

Limitations to this experiment

There are a variety of limitations to this study. Chasar et al., 2009, and Riva-Murry et al., 2011 found that THg, MeHg, DOC, ANC, and pH each play a significant role in MeHg concentrations found in aquatic organisms within forest ecosystems. The association between MeHg bioaccumulation and acid deposition increases the probability of observing these relationships over time. Pairing, and correlating water chemistry results from the tributaries in this study could strengthen the MeHg macroinvertebrate results. While there is lack of water chemistry data specifically pertaining to episodically acidic tributaries, Millard (2016) has examined chronically acidic tributaries within the same watershed, but not MeHg concentrations of macroinvertebrates. The contributions of shallow (surface and shallow-subsurface flow systems) geochemical exchange between terrestrial compartments and aquatic ecosystems in streams makes them particularly sensitive to watershed characteristics that develop MeHg production and transport (Ward et al., 2010). Hg bioaccumulation and biomagnification in lotic systems without point source emissions has been strongly correlated to forest cover, wetlands

extent and connectivity, and hydrologic alteration (Rypel et al., 2008; Scudder et al., 2009; St. Louis et al., 1994; Ward et al., 2010). Not only could water chemistry improve the understanding of MeHg food web dynamics for the tributaries of this study, but it could paint a clearer picture of spatial distribution of MeHg for the watershed. Small sample sizes impacted the statistical significance for MeHg concentrations associated with sources and trophic position within FFGs (see Tables 2, 3 & 4). The scrappers FFG had one sample collected from MBBR for 2013 and T6A for 2014, and no samples collected from T6 and T6A in 2013. However, Honnedaga Lake remains a fragile ecosystem and care has been taken to preserve it by collecting only the necessary amount of sample. Finally, the macroinvertebrate samples from 2015–2016 years were analyzed, but were not added to this study due to time constraints, and should be taken into consideration in future research. Future research conducted on the episodically acidic tributary should observe the Hg dynamics between the tributary's water chemistry; proximal spatial location; incorporate larger sample sizes; other levels of trophic positioned organisms; and archived datasets to ascertain a clearer picture of Honnedaga's food web.

Conclusion

While I hypothesized that organisms with a higher trophic position would have higher methylmercury concentrations, only one of the FFGs from this study had statistical significance. Methylmercury concentrations showed significant changes for only the years of this study. Finally, concentrations of the neurotoxin showed significant

increases for both reference tributaries. These results compliment the larger Honnedaga Lake research project to characterize and develop a better understanding of the transport, transformations, and bioaccumulation of mercury in forest-surface water ecosystems in response to tributary and watershed liming. This research will assist in ascertaining solutions to restoring ecosystems impacted by acidification, and mercury deposition in New York State.

Figures

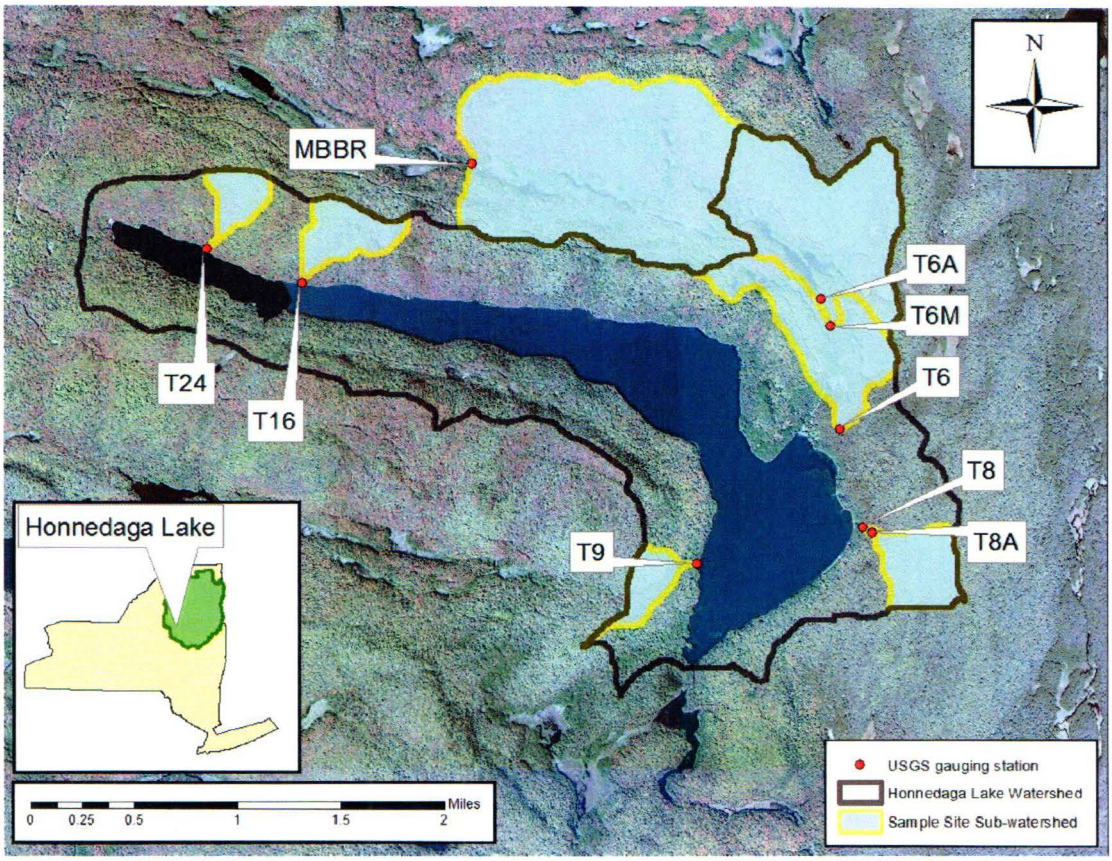

Figure 1: Map of Honnedaga Lake and associated treatment (T6), observed (T6M & T6P) and reference watersheds (MBBR & T6A) located in the northeast portion of the Adirondack Park (43° 31'06"N and 74°48'31"W).

Table 1: FFGs' $\delta^{13}C$ or $\delta^{15}N$ signature values across all tributaries from 2013 – 2014 in 10th, 25th, 50th, 75th, and 90th percentiles, their respective ranges, and order of the ranges. Red, and blue highlights dignify N, and C, respectively.

FFG	Isotope signature	Quantile value					Range	Order of range
		0.1	0.25	0.5	0.75	0.9		
Scrapper	$\delta^{15}N$	3.3	4.4	4.9	5.7	6.8	(1.37) – (5.61)	3rd
	$\delta^{13}C$	-34.4	-32.5	-30.3	-28.7	-28.2	(-34.40) – (-28.18)	2nd
Predator	$\delta^{15}N$	3.3	4.4	4.9	5.7	6.8	(3.30) – (6.77)	1st
	$\delta^{13}C$	-33.3	-31.8	-30.3	-28.7	-24.8	(-33.34) – (-24.80)	3rd
Omnivore	$\delta^{15}N$	2.2	3.5	4.3	4.8	6.1	(2.18) – (7.50)	4th
	$\delta^{13}C$	-33.1	-29.5	-27.3	-25.2	-23.6	(-33.08) – (-23.57)	4th
Shredder	$\delta^{15}N$	1.1	2.7	3.4	4.3	5.0	(1.10) – (4.96)	2nd
	$\delta^{13}C$	-31.9	-31.0	-29.3	-27.0	-26.3	(-31.87) – (-26.30)	1st

Table 2: Calculated p-values based on site (i.e. T6, T6A, and MBBR), FFGs (predator, omnivore, shredder, and scrapper), and paired parameters for dependent variable $\log_{10}$ MeHg (ng/g dw) concentrations. MeHg had a statistically significant relationship with each year (2013 and 2014). This means from year to year, there was a difference in MeHg concentrations. Site*Year was tested further for its statistical significance. The green highlight indicates statistical significance. $\alpha = 0.05$

Source	p-value
Site	0.757
FFG	0.026
Site*FFG	0.22
Year	<0.001
Site*Year	0.062
FFG*Year	0.999
Site*FFG*Year	0.201

Table 3: Calculated p-values based on site (i.e. T6, T6A, and MBBR) against 2013 – 2014 for dependent variable $\log_{10}$ MeHg (ng/g dw) concentrations. MeHg concentrations had a statistically significant relationship with T6A, and MBBR 2013 vs. 2014. This means MeHg concentrations have increased in T6A and MBBR from 2013 to 2014.The green highlights indicate statistical significance. $\alpha = 0.05$

Parameter	Estimate	p-value
T6 2013 vs. 2014	-0.116	0.535
T6A 2013 vs. 2014	-0.675	0.014
MBBR 2013 vs. 2014	-0.935	<0.001

Table 4: Calculated p-values for FFGs (predator, omnivore, shredder, and scrapper) across all site (i.e. T6, T6A, and MBBR), and years (2013 – 2014) with $\log_{10}$ MeHg (ng/g dw) concentrations, for dependent variable $\delta^{15}N$. MeHg concentrations have a significant relationship to $\delta^{15}N$ for predators and omnivore FFGs. The green highlight indicates statistical significance. $\alpha = 0.05$

FFG	p-value
Scrapper	0.440
Predator	0.561
Omnivore	0.009
Shredder	0.629

Table 5: FFGs (predator, omnivore, shredder, and scrapper) trophic positions', across all sites (T6, T6A, and MBBR), and years (2013 – 2014) in 10^{th}, 25^{th}, 50^{th}, 75^{th}, and 90^{th} percentiles, and mean trophic position.

FFG	Quantile value					Mean trophic position
	0.1	0.25	0.5	0.75	0.9	
Predator	1.97	2.65	2.85	2.99	3.42	2.81
Omnivore	1.88	2.22	2.66	2.85	3.63	2.58
Scrapper	1.66	2.09	2.47	2.81	2.92	2.39
Shredder	2.00	2.22	2.27	2.51	3.12	2.38

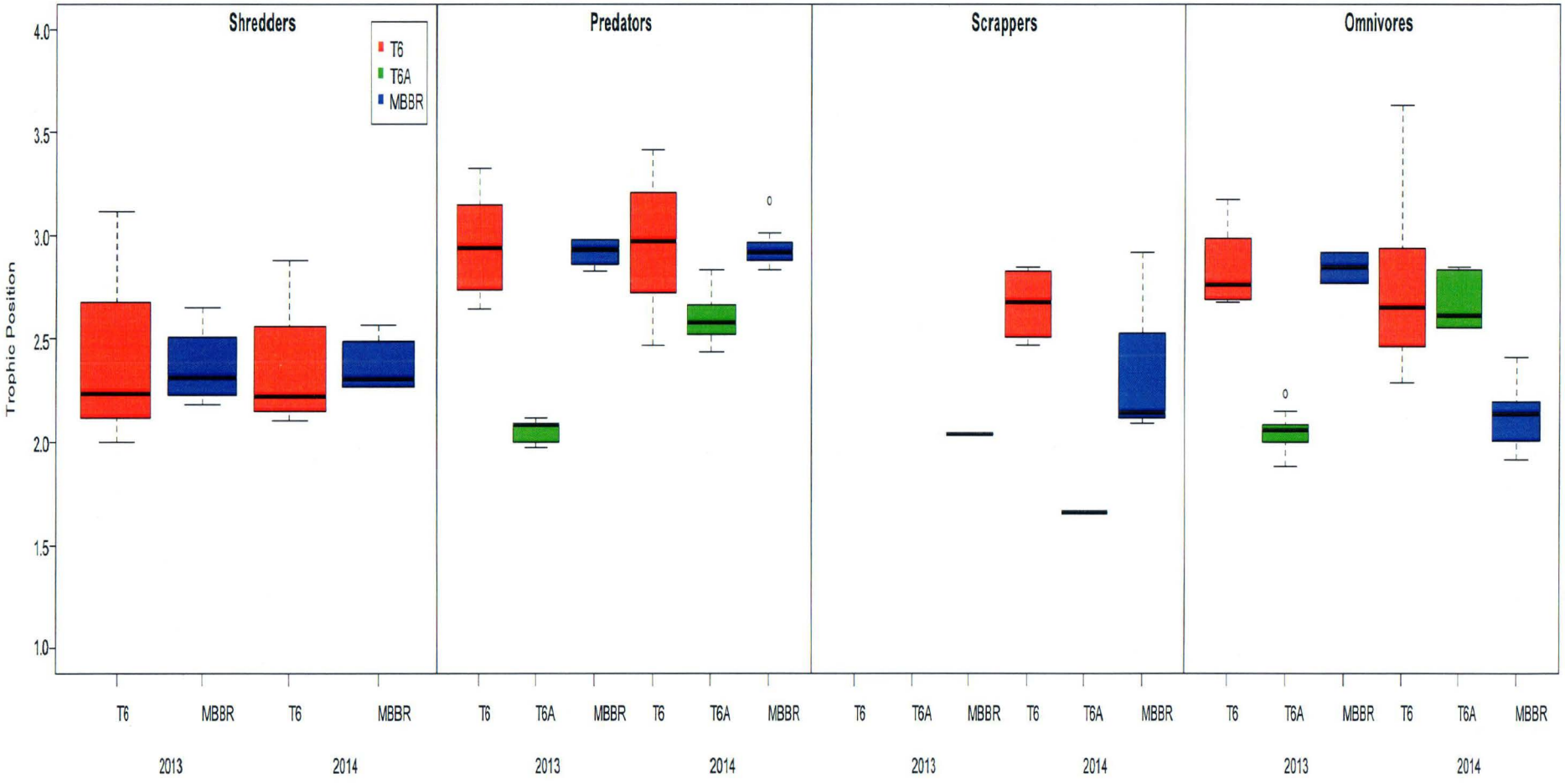

Figure 2: Trophic position of macroinvertebrate of FFGs (shredders, predators, scrappers, omnivores) taken from T6 (red), T6A (green), and MBBR (blue) from 2013 – 2014. *Lower* and *upper* boundaries of each *box* represent 25th and 75th percentiles; *lower*, and *upper whiskers* represent 10th and 90th percentiles; and the *line within the box* represents the median. *Clear circles* are outliers.

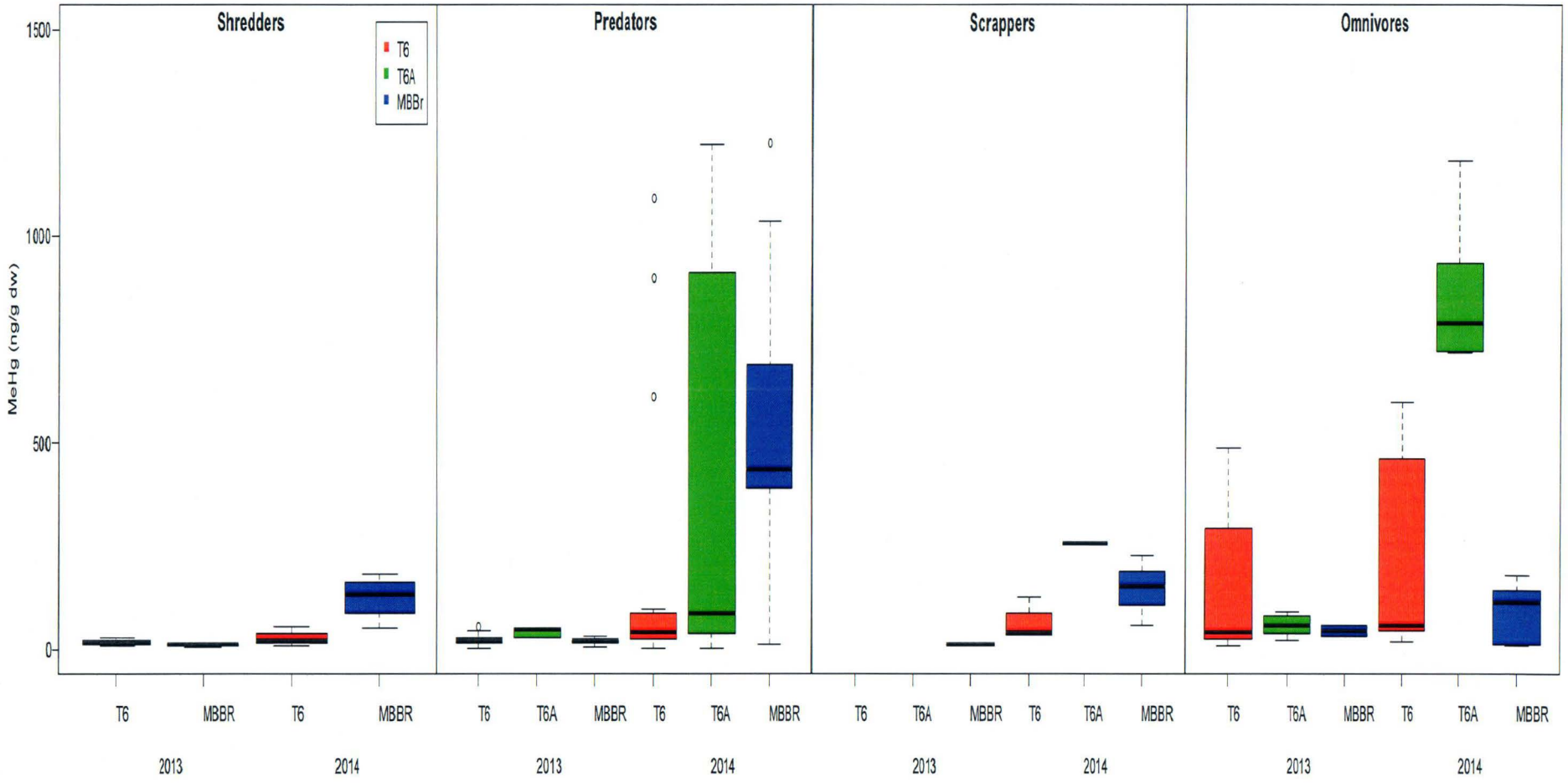

Figure 3: MeHg (ng/g dw) concentrations of macroinvertebrate of FFGs (shredders, predators, scrappers, omnivores) taken from T6 (red), T6A (green), and MBBR (blue) from 2013 – 2014. *Lower* and *upper* boundaries of each *box* represent 25th and 75th percentiles; *lower*, and *upper whiskers* represent 10th and 90th percentiles; and the *line within the box* represents the median. *Clear circles* are outliers.

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