Science of the Total Environment 506–507 (2015) 126–136

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

Bioaccumulation and biomagnification of mercury in African lakes: The importance of trophic status

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HIGHLIGHTS

- We characterized Hg in water and biota from 8 East African study sites.
- Hg concentrations in fish were low and should not pose a risk to human consumers.
- Hg uptake and biomagnification rates were negatively related to trophic status.
- Growth dilution in phytoplankton and consumer trophic levels led to low fish Hg.

ARTICLE INFO

Article history:
Received 21 October 2014
Received in revised form 24 October 2014
Accepted 26 October 2014
Available online 17 November 2014

Editor: Mae Mae Sexauer Gustin

Keywords:
Mercury
Eutrophication
Africa
Biomagnification
Chlorophyll
Trophic status

ABSTRACT

Despite the global prevalence of both mercury (Hg) contamination and anthropogenic eutrophication, relatively little is known about the behavior of Hg in eutrophic and hypereutrophic systems or the effects of lake trophic status on Hg uptake and trophodynamics. In the current study we explore Hg trophodynamics at 8 tropical East African study sites ranging from mesotrophic to hypereutrophic, in order to assess the influence of lake trophic status on Hg uptake and biomagnification. Comprehensive water, plankton and fish samples were collected for analysis of total mercury (THg) and stable carbon and nitrogen isotopic ratios. We found evidence that uptake of THg into phytoplankton tended to be lower in higher productivity systems. THg concentrations in fish were generally low, and THg trophic magnification factors (TMFs; representing the average increase in contaminant concentrations from one trophic level to the next) ranged from 1.9 to 5.6. Furthermore TMFs were significantly lower in hypereutrophic lakes than in meso- and eutrophic lakes, and were negatively related to chlorophyll *a* concentrations both across our study lakes, and across African lakes for which literature data were available. These observations suggest that THg concentrations were strongly influenced by trophic status, with year-round high phytoplankton and fish growth rates reducing the potential for high THg in fish in these productive tropical lakes.

1. Introduction

Several studies over the past decade have greatly increased our understanding of mercury (Hg) concentrations and trophodynamics in arctic, temperate and tropical systems (Campbell et al., 2003a; Swanson and Kidd, 2010; Kidd et al., 2012; Clayden et al., 2013; Lavoie et al., 2013). However, it has been noted that in tropical African lakes, Hg concentrations in fish are 'anomalously' low relative to fish from temperate systems, despite total Hg (THg) concentrations in water that are often comparable (Black et al., 2011). This highlights a need for more detailed analysis of the factors that influence uptake and trophic transfer of Hg in these tropical African systems. Furthermore, the behavior of Hg in eutrophic and hypereutrophic systems (which are common in Africa), and the influence of lake trophic status on Hg trophodynamics remain poorly understood. A recent debate in the literature regarding the relationship between lake trophic status and trophic transfer of Hg (Verburg, 2014; Clayden et al., 2014) points to a clear need for robust field studies addressing this issue. Having a comprehensive understanding of the effects of trophic status on Hg uptake and biomagnification is critical, given the increasing anthropogenic eutrophication of many freshwater and coastal systems around the world, as well as ongoing recovery from eutrophication in many systems

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http://dx.doi.org/10.1016/j.scitotenv.2014.10.094

where nutrient loading has been reduced. Also, eutrophic lakes often support highly productive fisheries, which are a source of daily subsistence upon which riparian populations can be heavily dependent, leading to natural concern about Hg concentrations in these systems.

East Africa is home to lakes that span a wide range in trophic status, from oligotrophic (e.g. Lake Tanganyika, based on its chlorophyll concentrations; Hecky and Kling, 1981) to highly productive lakes, including both naturally hypereutrophic (e.g. Lake George) and anthropogenically eutrophied lakes (e.g. Lake Victoria). Many of these lakes can sustain perennially high standing crops of phytoplankton (Poste et al., 2013), offering a unique opportunity to explore the influence of lake trophic status on Hg uptake and trophic transfer, and in particular, to assess the potential for eutrophication-mediated biodilution (Pickhardt et al., 2002; Herendeen and Hill, 2004; Chen and Folt, 2005; Karimi et al., 2007) of Hg in tropical African lakes. Previous studies of Hg in the food webs of several sub-Saharan African lakes (including several embayments of Lake Victoria, Lake Malawi, Lake Tanganyika, Lake Chad and several smaller lakes; Table 1) have typically reported low Hg concentrations in fish (Black et al., 2011), and Hg biomagnification (Table 1) that falls within the range encountered in freshwater and marine systems worldwide (Lavoie et al., 2013).

The primary objectives of this study were two-fold: 1) to characterize bioaccumulation and biomagnification of Hg in several East African lakes spanning a range of phytoplankton biomass and productivity in the context of food web structure established through stable isotope analysis; and, in particular, 2) to explore Hg trophodynamics in tropical hypereutrophic lakes where primary productivity is high year-round. Specifically, this study will test the hypothesis that eutrophication-mediated biodilution at the base of the food web and at consumer trophic levels will lead to reduced Hg concentrations in biota from higher trophic status lakes.

In this study, we use stable carbon ($\delta^{13}C$) and nitrogen ($\delta^{15}N$) isotope analysis to characterize food web structure and trophic transfer of Hg at the study sites, using $\delta^{15}N$ as an indicator of trophic level (Minagawa and Wada, 1984; Peterson and Fry, 1987), and $\delta^{13}C$ as an indicator of primary carbon source (Cabana and Rasmussen, 1994; Hecky and Hesslein, 1995). We quantify food web biomagnification of Hg using trophic magnification factors (TMFs), defined as 10^b, where b is the slope of the regression of log-transformed Hg concentrations against estimated trophic level (calculated based on $\delta^{15}N$ values; Borgå et al., 2012).

2. Materials and methods

2.1. Study sites and sample collection

Our study included several lakes in Uganda (East Africa), ranging in size from small crater lakes (Saka and Nkuruba) to the African Great Lakes Albert, Edward and Victoria (where two study sites were included); and ranging in trophic status from mesotrophic to hypereutrophic (Table 2, Fig. 1). These sites (excluding Lake Albert) are described in detail by Poste et al. (2013), and results of stable isotope and Hg analysis for Napoleon Gulf and Murchison Bay are reported in Poste et al. (2012).

Water, plankton, and fish samples were collected using trace-metal clean sampling techniques between September 2008 and February 2009 from all study sites except Lake Albert, where samples were collected in April–May 2007. In Lake Edward, water and plankton were collected from two sites (one nearshore site (EdK) near the inflow from Lake George, and one offshore site (EdO)), whereas for fish, Lake Edward was treated as a single site. Sample collection and analytical methodology for determination of chlorophyll *a* concentration are described in detail by Poste et al. (2013). Briefly, integrated euphotic zone water samples were collected on a monthly basis throughout the study period (except for in Murchison Bay and Napoleon Gulf, where samples were collected every two weeks), whole water was filtered through triplicate Whatman GF/F filters (nominal pore size of 0.7 μm), and chlorophyll *a* was determined fluorometrically after acetone extraction (Poste et al., 2013).

Water samples for analysis of total mercury (THg) were collected by lowering certified trace-metal clean glass bottles (VWR EP114-250A) to ~15 cm below surface and opening, filling and re-sealing at depth. Plankton samples were collected through vertical net hauls (20 μm net for phytoplankton; 80 and 153 μm nets for zooplankton) and were concentrated onto pre-combusted quartz fiber filters. Subsamples from net hauls were also preserved and examined microscopically to confirm general sample composition, and zooplankton samples containing appreciable amounts of phytoplankton were excluded from further analysis (this generally occurred where large colonial cyanobacteria were highly abundant). Fish ($n = 509$, representing 28 species from planktivores to top predators) were purchased from local fishermen and catch location was verified where possible. Sub-samples of skinless dorsolateral muscle tissue were removed from fish for stable isotope and THg analyses. When fish were too small to isolate dorsolateral muscle tissue, whole fillets of axial musculature were collected, and where this was not possible (for the cyprinid *Rastrineobola* spp. and two haplochromine cichlids from Lake Saka, which were <5 cm long), they were analyzed whole. Filters and fish samples were kept frozen until further processing.

2.2. Sample analysis

Sample processing, analytical methodology and quality assurance/quality control procedures (including SRMs used) for stable isotope and THg analyses for water, plankton and fish are presented in detail in Poste et al. (2012). Briefly, stable carbon ($\delta^{13}C$) and nitrogen ($\delta^{15}N$) isotopic ratios were determined for oven-dried (60 °C for 24 h) and homogenized fish and plankton samples at the Environmental Isotope

Table 1
Review of log(THg) ~ $\delta^{15}N$ regression slopes, trophic magnification factors (TMFs) and mean Chl *a* concentrations for previous THg biomagnification studies in sub-Saharan African lakes.

Site	Site code	Slope log(THg) ~ $\delta^{15}N$	TMF[a]	Chl *a* (μg/L)	Source of data
Bosomtwe, benthic	B_b	0.21*	5.3	10.3	Hg: Poste et al. (2008); Chl *a*: Otu (2010)
Bosomtwe, pelagic	B_p	0.13**	2.8	10.3	""
Nkuruba	Nk_C	0.14*	3.0	6.2	Hg: Campbell et al. (2006); Chl *a*, see Table 1
Saka	S_C	0.14	2.9	90	Hg: Campbell et al. (2006); Chl *a*, see Table 1
Tanganyika	T	0.22**	5.6	0.66	Hg: Campbell et al. (2008); Chl *a*, Pirlot et al. (2005)
Thruston Bay	TB[b]	0.28**	9.0	64.3	Hg, Chl *a*: Campbell et al. (2004)
Napoleon Gulf	NG_C	0.16**	3.6	24.7	Hg: Campbell et al. (2003b); Chl *a*, see Table 1
Winam Gulf	WG	0.17**	3.6	20	Hg, Chl *a*: Campbell et al., 2003b
Malawi, benthic	Ma_b	0.25**	7.1	1	Hg: Kidd et al. (2003); Chl *a*: Guildford et al. (2007)
Malawi, pelagic	Ma_p	0.23**	6.1	1	""
Chad, south basin	C	0.21**	5.2	5.5	Hg: Kidd et al. (2004); Chl *a*: Lemoalle (1979)
Ziway	Z	0.13**	2.8	70.5	Hg: Tadiso et al. (2011); Chl *a*: Hailu (2011)

Statistical significance of log(THg) ~ $\delta^{15}N$ regressions is indicated as: "**" for $P < 0.01$, "*" for $P < 0.05$, and no asterisk where the relationship was not statistically significant.
[a] TMF was calculated from reported log(THg) ~ $\delta^{15}N$ regression slopes based on an assumed ^{15}N trophic enrichment factor of 3.4‰ per trophic level.
[b] This site was not included in the Fig. 2b regression.

Table 2
Physicochemical characteristics of study lakes, including lake trophic status and THg concentrations in water.

Lake	Sampling site	Code	Area (km^2)	z_{max} (m)	Chl *a* (μg/L)	THg (ng/L)
Albert	Butiaba/Kaiso	A	5600	58	19.2 (M/E)	0.46 ± 0.11
Edward	Edward Kazinga	EdK	–	–	66.3 (H)	0.65
Edward	Edward Open	EdO	–	–	21.3 (E)	0.38 ± 0.01
George	George	G	250	7	138 (H)	1.09 ± 0.11
Mburo	Mburo	Mb	13	4	48.6 (H)	0.62 ± 0.04
Victoria	Murchison Bay	Mu	18	7	96.5 (H)	1.30 ± 0.09
Victoria	Napoleon Gulf	NG	26.5	20.5	24.7 (E)	0.53 ± 0.07
Nkuruba	Nkuruba	Nk	0.03	38	6.2 (M)	0.45
Saka	Saka	S	0.15	8.5	90 (H)	0.81 ± 0.07

Morphometric and chlorophyll *a* data for Lake Albert are from Talling (1963) and Poste et al. (2011) respectively. Data for remaining study sites are from Poste et al. (2013), and reported chlorophyll *a* concentrations for these sites represent the mean value over a six-month period from September 2008–February 2009. Trophic status was determined based on Vollenweider and Kerekes (1982) and is reported as M (mesotrophic), E (eutrophic), and H (hypereutrophic) in parentheses alongside chlorophyll *a* concentrations.

Laboratory at the University of Waterloo. Samples were not acidified for removal of carbonates prior to analysis since we did not expect large amounts of carbonates in these biotic samples. Furthermore based on test samples for both fish and plankton, we did not observe any statistically significant difference in $\delta^{13}C$ values between unacidified and acidified samples. To determine variability between runs, standards for carbon (IAEA-CH6 (sugar), EIL-72 (cellulose) and EIL-32 (graphite)) and nitrogen (IAEA-N1 and IAEA-N2, both ammonium sulfate) were analyzed in each sample run (for each group of 100 samples, two sets of three or four replicates of carbon and nitrogen standards are analyzed). Mean standard deviations from expected values for standard reference materials (SRMs) were ±0.2‰ for $\delta^{13}C$ and ±0.3‰ for $\delta^{15}N$. Mean coefficients of variation for samples run in duplicate (one in every ten samples) were ±0.05‰ and ±0.21‰ for $\delta^{13}C$ and $\delta^{15}N$ respectively.

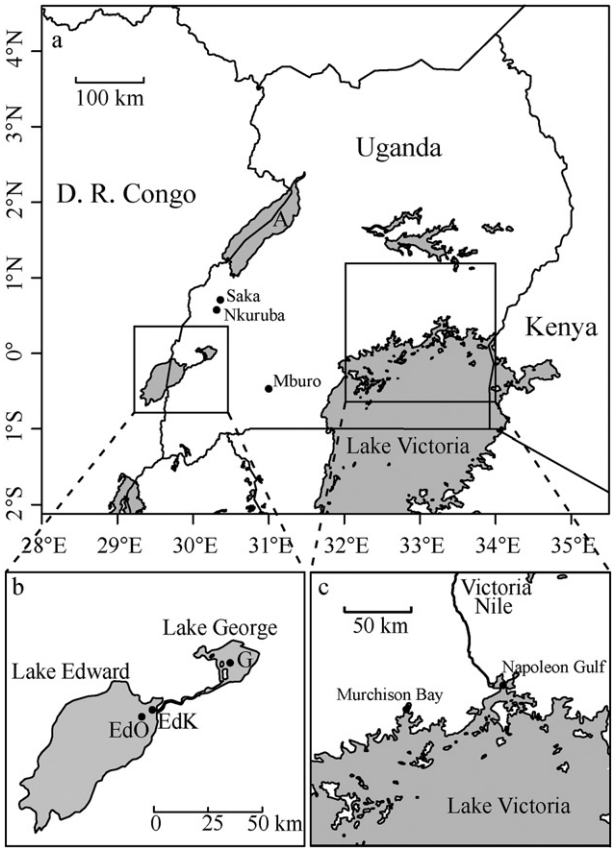

Fig. 1. a. Map of Uganda showing location of study sites; b. detailed map showing the Lake George study site as well as the two Lake Edward study sites; and c. detailed map of northern Lake Victoria showing Napoleon Gulf and Murchison Bay. Site codes used in Fig. 2b are given in Table 2. This figure has been reproduced with slight modifications with permission from Poste et al. (2013). Copyright 2013 by the Association for the Sciences of Limnology and Oceanography, Inc.

THg concentrations were measured in water, freeze dried plankton samples, and frozen (wet) fish at the Canada Centre for Inland Waters (Burlington, Canada) using EPA methods 1631 (for water; US EPA, 1999) and 7473 (for biota; US EPA, 1998). Zooplankton samples (collected from Lake Nkuruba) were gently scraped off of filters, while net phytoplankton (hereafter referred to as simply "phytoplankton") samples were analyzed on filters. A filter blank correction of 0.03 ng THg was applied, based on analysis of blank filters. Dry-weight THg concentrations in zooplankton from lake Nkuruba were converted to wet-weight concentrations by multiplying by 0.1 (Pace and Orcutt, 1981) prior to inclusion in regressions of log(THg) ~ $\delta^{15}N$ and TL (for assessment of biomagnification). Similarly, phytoplankton THg concentrations were converted to wet-weight concentrations for assessment of enrichment of THg from water to phytoplankton (McGeer et al., 2003).

NIST 1641c (diluted to 1.84 ng/L) was used as a standard for analysis of THg in water, and was on average (n = 4) within 3.1% of the expected value. The precision for water samples run in duplicate (n = 11) was on average ±8.9% and the mean standard deviation for replicates within study sites was ±0.06 ng/L (based on duplicate samples from 7 sites). For THg analysis of biotic samples, recovery for the SRMs used (DORM-1 (n = 8), NIST 1556b (n = 20), TORT-2 (n = 20), and NBS SRM-1571 (n = 3)) ranged from 96 to 106% and measured concentrations were always within 10% of the certified value. The mean coefficient of variation for duplicated biotic samples (approximately one in every 15 samples) was 3.1 ± 2.8%.

2.3. Calculations and statistical analyses

Trophic level (TL) was calculated for all biotic samples, assuming a trophic transfer enrichment factor of 3.4‰ (Post, 2002), and based on sample $\delta^{15}N$ values relative to $\delta^{15}N$ of a baseline organism with an assumed well-defined trophic level ($TL_{sample} = (\delta^{15}N_{sample} - \delta^{15}N_{baseline})/3.4‰ + \lambda$; where λ is the assumed trophic level of the baseline organism; Vander Zanden and Rasmussen, 2001). Since we did not have common benthic invertebrate samples across all study sites, for consistency, mean phytoplankton (collected regularly over the six month sampling period) was used as a baseline for TL calculations for all sites but Lake Albert, where plankton samples were not available (and *Brycinus nurse*, an obligate zooplanktivore, was used as a baseline), and Murchison Bay, where phytoplankton $\delta^{15}N$ was highly variable and *Oreochromis niloticus* (a primary consumer) was used as a baseline. It should be noted that calculated TL (and metrics relying on TL values) can be influenced by inaccurate estimates of baseline organism $\delta^{15}N$ as well as by isotopic fractionation rates that differ from the assumed rate of 3.4‰ per trophic level for $\delta^{15}N$ (Borgå et al., 2012). Indeed, several studies have identified differences in fractionation based on primary form of N excretion, diet (e.g. herbivory vs. carnivory), dietary N − content, and starvation (Adams and Sterner, 2000; McCutchan et al., 2003; Vanderklift and Ponsard, 2003; Haubert et al., 2005). However,

despite the limitations associated with these assumptions, calculated TL values based on methods and assumptions that are consistently used in studies from around the world allow for useful within and between system comparisons of trophic position as well as Hg trophodynamics.

In Lake Edward, there was a strong negative relationship between $\delta^{15}N$ and $\delta^{13}C$ of phytoplankton ($r = -0.96$, $P = 0.00005$). This was attributable to spatial differences in phytoplankton C and N isotopic ratios, with significantly lower $\delta^{15}N$ ($P = 0.008$) and higher $\delta^{13}C$ ($P = 0.003$) nearshore (EdK) compared to offshore (EdO), which is likely attributable to increased prevalence of high phytoplankton C demand and fixation of atmospheric N in the more productive nearshore area (as was also reported for Lake Victoria by Hecky et al., 2010). TL values for fish from Lake Edward were corrected for this baseline relationship (Vander Zanden and Rasmussen, 1999; Post, 2002), such that the baseline nitrogen isotopic ratio for all samples was set to the mean $\delta^{15}N$ value for Lake Edward phytoplankton (0.3‰). With the exception of analyses involving TL values, $\delta^{15}N$ values unadjusted for baseline $\delta^{15}N$ were used for all statistical analyses.

Due to the lack of dissolved THg concentrations in water, we were unable to calculate true phytoplankton bioconcentration factors (BCFs; the concentration of a compound in an organism relative to the aqueous concentration of the compound in the environment) for the study sites, however we still sought to assess the relative enrichment of THg in phytoplankton relative to THg in water by dividing measured THg concentrations in phytoplankton (in ng/kg wet weight; THg_{PHYTO}) by THg concentrations in whole water samples (in ng/L; THg_{WATER}). We assume that this ratio (THg_{PHYTO}/THg_{WATER}) is an indicator of the uptake of THg at the base of the food web, and can serve as an estimate (albeit with limitations) of THg bioconcentration into the phytoplankton. We also determined the proportion of THg associated with phytoplankton based on measured THg in phytoplankton, and phytoplankton biomass as determined through microscopy in a related concurrent study (as reported in Poste et al., 2013) and found that for these productive tropical lakes, where net phytoplankton is expected to dominate the particulate matter, only a very small fraction of the THg pool was bound to the THg, suggesting the presence of an appreciable pool of dissolved THg, and further supporting our assumption that THg_{PHYTO}/THg_{WATER} can be considered an approximation of phytoplankton BCF for these study sites.

Mercury trophodynamics were characterized based on fish data (given the lack of MeHg data for plankton samples) through regression of log-transformed THg concentrations against $\delta^{13}C$, $\delta^{15}N$ and TL for fish (zooplankton were included in regressions for lake Nkuruba, because the fish present in this lake did not include any primary consumers based on calculated TL values). Fish THg concentrations were log-transformed to achieve a normal distribution, and were not standardized for length, since there were no consistent relationships across study sites between length and log-transformed THg concentrations for the fish species sampled (as assessed through linear regression on a site-by-site basis). For each site, we used multiple regression models including THg concentrations in fish as the dependent variable and $\delta^{13}C$ and $\delta^{15}N$ as predictors to assess the relative importance of $\delta^{13}C$ and $\delta^{15}N$ in determining THg in fish.

Trophic magnification factors (TMFs) were calculated for each site as 10^b, where "b" is the slope of the log(THg) ~ TL regression (Borgå et al., 2012). TMFs were calculated using only fish data at all sites but Lake Nkuruba, where zooplankton were included in the regression (due to the lack of primary consumer fish in this lake). It is possible that calculation of TMFs for THg using only fish may result in higher TMF values than when calculated including other taxa (e.g. invertebrates) due to lower MeHg/THg ratios and reduced efficiency of trophic transfer in non-fish taxa, however, in a recent global meta-analysis of Hg biomagnification, Lavoie et al. (2013) did not find any significant difference in THg biomagnification for freshwater food webs composed of fish only or fish and other taxa.

Among-lake comparisons of log(THg) ~ TL regression coefficients were carried out using analysis of covariance (ANCOVA). A post hoc multiple comparison test was used to determine whether the regression slopes differed significantly between lakes, and to compare regression intercepts between sites that had homogeneous regression slopes. Linear regressions were carried out between log-transformed chlorophyll *a* concentrations and both THg_{PHYTO}/THg_{WATER} and TMF in order to assess the influence of site trophic status on Hg accumulation and trophic transfer. We used Cook's distance (D_i) to test for statistical outliers when running these regressions, where observations above a specified threshold ($D_i > 4 / (n - k - 1)$; where D_i is Cook's distance, n is the number of observations, and k is the number of independent variables; Fox, 1991) were classified as "too influential" and were removed from the analysis.

Lakes Edward and George provided an opportunity to closely examine the interaction between Hg trophodynamics and trophic status, given their direct hydrological connection and their shared faunal communities. We carried out a comparative case study of Hg concentrations and trophodynamics in these lakes, wherein Welch's two sample *t*-tests were used to compare phytoplankton and fish THg concentrations (log-transformed to achieve normal distribution) between lakes.

3. Results and discussion

3.1. THg in water and uptake by phytoplankton

Mean THg concentrations in water (Table 2) ranged from 0.38 ng/L (in offshore Lake Edward; EdO) to 1.30 (in Murchison Bay), suggesting low levels of mercury contamination across all of our sites. THg concentrations in the current study were generally below concentrations previously reported for Lake Victoria as well as the smaller African lakes Naivasha and Mariut (Campbell et al., 2003a) and freshwater reservoirs in Burkina Faso (Ouédraogo and Amyot, 2013). However, our observed concentrations were similar to observations from the Laurentian Great Lakes (offshore: 0.3–0.8 ng/L, and generally under 1 ng/L with the exception of some polluted nearshore stations; Dove et al., 2012) and the majority of the study sites (Table 2) had THg concentrations that fell within the range reported for several remote lakes in the Canadian arctic (0.29–0.72 ng/L; Gantner et al., 2010a).

We observed a positive relationship between total mercury concentrations in water and chlorophyll *a* concentrations ($P = 0.005$, $r = 0.87$); however, we did not observe a correlation between phytoplankton bound THg (in ng/L, calculated from measured THg in phytoplankton and phytoplankton biomass) and THg in whole water samples, suggesting that this relationship is not a reflection of differences in the amount of phytoplankton-bound mercury included in these samples. One possible explanation is that when anthropogenic input of nutrients and resultant algal biomass are high, there may also be increased anthropogenic input of mercury. There are several other factors that may also act to determine mercury concentrations in water, including the extent of wetlands, stratification, mixing, and site depth. In particular, shallow non-stratified sites (including Murchison Bay, Lake Mburo, Lake George and Lake Saka; all highly productive lakes) are more likely to experience frequent re-suspension of sediment-borne mercury.

Phytoplankton uptake of inorganic mercury is generally a passive process, while there is evidence to suggest active uptake of MeHg (Pickhardt and Fisher, 2007). There are many factors that can influence Hg uptake and concentrations in phytoplankton, including: Hg concentrations in water, the proportion of aqueous Hg present as methyl mercury (MeHg), water chemistry parameters that moderate MeHg production and bioavailability of MeHg to the aquatic food web (e.g. dissolved organic carbon (DOC) concentrations and pH; Pickhardt and Fisher, 2007; Luengen et al., 2012), phytoplankton community composition, and phytoplankton growth rate. In lakes with high primary productivity, the rapid rate of phytoplankton biomass production may outstrip Hg uptake and result in "growth dilution" (Pickhardt et al., 2002; Herendeen and Hill, 2004; Karimi et al., 2007). Under these

conditions, phytoplankton growth rates are high, and rapidly diving cells may not fully equilibrate with the available dissolved Hg concentrations (Herendeen and Hill, 2004; Karimi et al., 2007). Effectively, high growth rates can serve to dilute Hg concentrations at the base of the food web, resulting in high phytoplankton biomass with low cellular Hg concentrations, a phenomenon which has been observed in both experimental (Pickhardt et al., 2002; Karimi et al., 2007) and observational (Chen and Folt, 2005; Kehrig et al., 2009; Luengen and Flegal, 2009; Verburg et al., 2014) studies, and which can translate to reduced Hg concentrations in consumer organisms (Pickhardt et al., 2005).

Mean THg concentrations in phytoplankton ranged from 13 to 31 ng/g dry weight for the study sites. THg_{PHYTO}/THg_{WATER} values (as an estimate of BCF; range: 1465–8026 L/kg w.w.) were generally an order of magnitude lower than phytoplankton BCFs reported in the literature for THg in temperate lakes (~28000–60000 L/kg w.w., Watras and Bloom, 1992) calculated based on whole water THg concentrations (as done in the current study and reported as THg_{PHYTO}/THg_{WATER}). However, they were higher than BCFs observed by Wang et al. (2012) for inorganic Hg (THg less MeHg, calculated using dissolved concentrations; 100 L/kg w.w. for "small seston") in hypereutrophic Lake Taihu. The low values observed could reflect reduced Hg in phytoplankton due to growth dilution in these productive tropical lakes, or that MeHg only makes up a small proportion of the THg pool. These comparisons are, however, limited by the fact that we did not determine true BCF values in the current study, and because we estimated bioconcentration as phytoplankton THg relative to whole water THg, if an appreciable amount of the THg pool in the whole water samples was bound to particulate matter (both phytoplankton and non-phytoplankton), this could lead to reduced availability of THg for uptake and subsequently THg_{PHYTO}/THg_{WATER} values.

Based on phytoplankton Hg concentrations and phytoplankton biomass (as determined through microscopy and reported in Poste et al., 2013), we found that the proportion of THg in whole water samples that was associated with phytoplankton was generally low (site means ranged from 0.8 to 8.6%). Furthermore, most of our study sites are eutrophic or hypereutrophic and have abundant phytoplankton populations, and so phytoplankton is likely to dominate the suspended particulate matter. This, taken with our observations that only a small proportion of the THg was associated with phytoplankton, suggests that there may be a substantial pool of dissolved Hg available for uptake in these lakes. Although it would have been preferable to calculate true BCF values using the dissolved THg fraction, for these lakes, calculations using whole water THg should yield reasonable estimates of bioconcentration. Furthermore, when we calculated THg_{PHYTO}/THg_{WATER} based on whole water THg less phytoplankton-associated THg resulted in only a very small effect on the resultant ratios.

The current study includes several hypereutrophic lakes where phytoplankton biomass is extremely high (for example, in Lake George, mean phytoplankton biomass over the study period was 35.7 mg/L; Poste et al., 2013). We observed a negative relationship between THg_{PHYTO}/THg_{WATER} and log-transformed chlorophyll *a* (Fig. 2; $r^2 = 0.66$, $P = 0.015$, suggesting lower accumulation of THg at the base of the food web at sites with higher trophic status, which is consistent with observations from temperate and subtropical lakes (Chen and Folt, 2005; Kidd et al., 2012; Wang et al., 2012; Verburg et al., 2014), and tropical estuaries (Kehrig et al., 2009). This is of critical importance as reduced Hg concentrations at the base of the food web can translate into lower concentrations throughout the food web as a whole (Pickhardt et al., 2002). The evidence that there may be appreciable dissolved THg concentrations in these lakes (as discussed above) suggests that the reduced Hg concentrations in phytoplankton from more productive lakes are more likely to be attributable to growth dilution, rather than biomass dilution; where, in theory, high biomass of phytoplankton draws down the pool of Hg available for uptake, leading to reduced phytoplankton Hg concentrations (Pickhardt et al., 2002).

However, interpretation of the THg_{PHYTO}/THg_{WATER} results of the current study is limited by the fact that dissolved THg concentrations were not measured and MeHg concentrations were not determined. Between-site differences in THg_{PHYTO}/THg_{WATER} could also reflect differences in MeHg availability between the study sites, whereby sites with higher relative contribution of MeHg to the THg pool may have higher subsequent THg_{PHYTO}/THg_{WATER} values (due to more efficient uptake of MeHg relative to inorganic Hg). Information regarding controls on MeHg production and degradation in African lakes is scarce (Black et al., 2011), although the high temperatures and availability of organic matter in many of our study lakes might be expected to promote methylation of Hg (Ullrich et al., 2001). On the other hand, there is also evidence that the high temperatures in tropical lakes may enhance divalent gaseous Hg formation, thus limiting the Hg pool available for methylation (Peretyazhko et al., 2006).

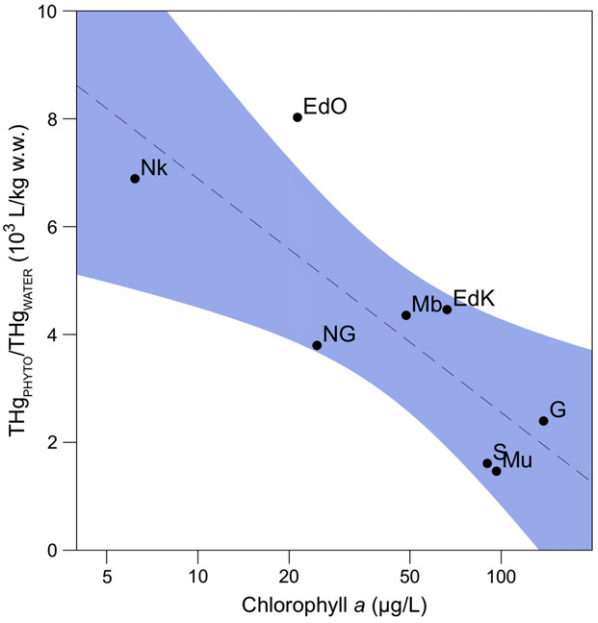

Fig. 2. Linear regression between THg_{PHYTO}/THg_{WATER} (as an estimate of bioconcentration) and log-transformed Chl *a* concentrations for study lakes ($n = 8$, $THg_{PHYTO}/THg_{WATER} = (11221 \pm 2374) - (4334 \pm 1406) * (\log_{10}\text{Chl } a)$, $r^2 = 0.66$, $P = 0.015$). The shaded band represents the 95% confidence interval for the regression and site codes used are defined in Table 2.

3.2. Further evidence of low Hg in fish from African lakes

THg concentrations in fish (Table 3, with detailed species-level information in Table S1) ranged from 0.8 ng/g (in an individual *O. niloticus* from Lake George) to a maximum of 855 ng/g (in an individual *Haplochromis squamipinnis* from Lake Edward). For all fish analyzed, THg concentrations were below 20 ng/g in >90% of fish from Lakes George and Mburo, >75% of fish from Lakes Edward, Nkuruba and Saka, >60% of fish from Napoleon Gulf and Murchison Bay, and 8% of fish from Lake Albert. THg concentrations exceeding 200 ng/g were found only occasionally in individual higher trophic level fish from Lake Albert, Lake Edward and Napoleon Gulf (Lake Victoria).

The food webs observed for these Ugandan lakes (Figures S1, S2) had several general characteristics, many of which have been described in detail in other studies (Campbell et al., 2003b, 2005, 2006; Mbabazi et al., 2004; Poste et al., 2012). With the exception of Lakes George, Edward and Albert, strongly piscivorous fish appeared to be rare, meanwhile omnivory was common, with few apparent obligate feeding relationships. The key species present in these lakes included tilapiine and haplochromine cichlids (often detritivorous and phytoplanktivorous, with some incorporation of primary consumers including benthic invertebrates (Greenwood, 1958)). Nile perch (*Lates niloticus*) were also present in several of

Table 3
Summary of results of stable isotope and mercury analyses (ranges in $\delta^{13}C$, $\delta^{15}N$, TL, and THg) for fish included in regressions, log(THg) ~ $\delta^{15}N$ regression results (r^2_{adj} and P values), and calculated TMFs. Species-level results for $\delta^{15}N$, $\delta^{13}C$ and THg analyses are found in Table S1.

Site	n	n fish spp.	$\delta^{13}C$ (‰)	$\delta^{15}N$ (‰)	TL	THg median	THg range	log(THg) ~ $\delta^{15}N$ and log(THg) ~ TL regression equations	r^2	*P*	TMF
Albert	48	14	−20.6 to −12.6	5.5–11.1	2.0–3.6	39.4	9.6–609	$-0.14 \pm 0.32 + (0.22 \pm 0.04)\delta^{15}N$ $-0.39 \pm 0.36 + (0.75 \pm 0.12)TL^{a,b}$	0.43	<0.001	5.6 ± 1.6
Edward	61	7	−18.7 to −9.6	3.6–13.6	2.0–4.9	10.2	3.5–855	$0.01 \pm 0.17 + (0.15 \pm 0.02)\delta^{15}N$ $-0.47 \pm 0.24 + (0.51 \pm 0.07)TL^{b,c}$	0.47	<0.001	3.3 ± 0.5
George	67	9	−26.6 to −1.5	1.8–11.0	1.9–4.6	2.3	0.8–67.7	$-0.01 \pm 0.10 + (0.08 \pm 0.02)\delta^{15}N$ $-0.37 \pm 0.18 + (0.28 \pm 0.06)TL^{e}$	0.25	<0.001	1.9 ± 0.2
Mburo	62	6	−20.4 to −8.7	3.1–12.3	2.1–4.8	3.7	1.5–32.2	$0.20 \pm 0.12 + (0.09 \pm 0.02)\delta^{15}N$ $-0.15 \pm 0.21 + (0.30 \pm 0.08)TL^{d,e}$	0.20	<0.001	2.0 ± 0.3
Murchison Bay	79	9	−20.7 to −11.0	4.9–12.4	1.2–3.7	13.1	3.5–101	$0.13 \pm 0.19 + (0.13 \pm 0.02)\delta^{15}N$ $0.25 \pm 0.17 + (0.43 \pm 0.07)TL^{c,d}$	0.32	<0.001	2.7 ± 0.4
Napoleon Gulf	95	14	−18.4 to −10.2	2.9–9.3	1.5–3.4	9.8	2.0–340	$-0.15 \pm 0.12 + (0.20 \pm 0.02)\delta^{15}N$ $-0.56 \pm 0.16 + (0.68 \pm 0.06)TL^{a}$	0.55	<0.001	4.8 ± 0.7
Nkuruba	22	3	−30.3 to −24.8	2.4–9.3	1.5–3.6	6.4	1.5–23.6	$-0.61 \pm 0.19 + (0.22 \pm 0.03)\delta^{15}N$ $-1.24 \pm 0.28 + (0.75 \pm 0.10)TL^{a,b}$	0.71	<0.001	5.6 ± 1.3
Saka	42	6	−23.8 to −17.4	2.3–7.7	2.1–3.7	5.1	1.9–49.2	$0.41 \pm 0.16 + (0.08 \pm 0.03)\delta^{15}N$ $-0.003 \pm 0.33 + (0.28 \pm 0.11)TL^{d,e}$	0.11	<0.05	1.9 ± 0.5

Regressions for all sites were carried out using only fish data, except for Lake Nkuruba, where zooplankton were included due to the lack of primary consumer fish in this lake. Addition of zooplankton to the Lake Nkuruba regression analysis had a substantial effect on the regression results (regression equation for fish only regression: $-0.13 \pm 0.21 + (0.13 \pm 0.03)\delta^{15}N$). Superscript letters next to log(THg) ~ TL regression equations describe the results of an ANCOVA followed by a post hoc multiple comparison test; sites labeled with a common letter had no significant difference in regression slope ($P < 0.1$).

the study lakes. The highest trophic levels in these lakes were typically occupied by *Clarias gariepinus*, *Bagrus docmac* and *Protopterus aethiopicus*, all known to be omnivores that feed primarily on a mix of benthic invertebrates and fish (Greenwood, 1958).

For all study sites, THg concentrations in fish were consistently lowest in primary consumers (including tilapiine cichlids such as *Oreochromis* spp.) and highest in secondary and tertiary consumers (such as *Lates* spp., *B. docmac*, *C. gariepinus*, and piscivorous haplochromine cichlids). In Lake Edward, as well as in Napoleon Gulf and Murchison Bay (as previously discussed in Poste et al. (2012)), *P. aethiopicus* (lungfish, a benthivore/piscivore typically found in wetlands; Goudswaard et al., 2002) had THg concentrations similar to those observed for primary consumers despite high calculated TL values, suggesting that differences in habitat or Hg accumulation kinetics may lead to divergent THg concentrations in lungfish from these sites. As such, *P. aethiopicus* was excluded from log(THg) ~ TL regressions for these three sites.

Our results are consistent with previous reports of low mercury concentrations in fish from African lakes as reviewed by Ouédraogo and Amyot (2013) and Black et al. (2011). In the current study, top predators typically had lower THg concentrations than have been reported for fish at similar trophic levels in uncontaminated arctic (e.g. Gantner et al., 2010a), temperate (e.g. Kidd et al., 2012), and some tropical sites (e.g. Bowles et al., 2001). Several possible explanations have been proposed for the consistently low THg concentrations observed in fish from tropical African lakes, including: shorter lifespans of tropical relative to temperate fish, higher growth rates in these warm and productive tropical lakes, differences in community composition and Hg uptake in the lower food web, and differences in methylation potential or MeHg degradation through photodemethylation (Black et al., 2011; Ouédraogo and Amyot, 2013). There is evidence from temperate and tropical systems that selenium (Se) availability can moderate Hg bioaccumulation and toxicity, with higher Se availability leading to lower Hg concentrations in biota and reduced toxicity to consumers (Bjerregaard et al., 2011; Sørmo et al., 2011). However, published Se data for African freshwaters and biota are limited to one study (of several reservoirs in Burkina Faso), where reported Se concentrations were low (36–170 ng/L in water; Ouédraogo and Amyot, 2013).

Studies in a wide range of aquatic systems have revealed that fish from systems with high primary productivity tend to have lower concentrations of Hg and other contaminants (e.g. organochlorines: Taylor et al., 1991; Larsson et al., 1992; Kidd et al., 1999). In an extensive study of environmental predictors of fish Hg concentrations in 161 Wisconsin lakes, Rypel (2010) found significant negative relationships between fish Hg concentrations and lake trophic status for largemouth bass, pike and walleye. Similarly, negative relationships were observed between chlorophyll *a* and Hg concentrations in largemouth bass from Florida lakes (Lange et al., 1993) and smallmouth and largemouth bass from lakes in New York state (Simonin et al., 2008). For several lakes in the northeastern United States, Chen and Folt (2005) report reduced Hg concentrations in fish from lakes with higher plankton densities. Meanwhile, in a comparative study of Hg accumulation in fish from one eutrophic and one oligotrophic lake, substantially lower Hg concentrations were observed in fish from the more productive study site (Kidd et al., 1999). Furthermore, Hg in fish has also been found to be low in highly productive Chinese reservoirs and lakes (Yan et al., 2010; Wang et al., 2012; Razavi et al., 2014). Given the high trophic status of many of the lakes included in the current study, it is not surprising that such low Hg concentrations are observed in fish from these lakes, however, causal mechanisms for these patterns remain largely undefined.

3.3. Negative relationships between THg in fish and $\delta^{13}C$

At several of the study sites we observed significant ($P < 0.05$) negative relationships between log-transformed THg concentrations in fish and $\delta^{13}C$. However, given that there were also significant ($P < 0.05$) relationships between $\delta^{15}N$ and $\delta^{13}C$ at several sites, we used multiple regression (including both $\delta^{15}N$ and $\delta^{13}C$ as predictors) to assess whether $\delta^{13}C$ had a significant effect on THg concentrations (independent of $\delta^{15}N$). Based on partial regression coefficients (Table S2), $\delta^{13}C$ was negatively related to, and a significant ($P < 0.05$) predictor of, THg concentrations in fish independent of $\delta^{15}N$ in Lake Edward, Lake George, Lake Mburo and Napoleon Gulf, but not at the remaining study sites (Table S2).

In temperate lakes with clear water and low primary productivity, the range of $\delta^{13}C$ observed in consumer trophic levels often reflects the relative importance of benthic (generally more ^{13}C-enriched) and pelagic (^{13}C-depleted) carbon sources (Hecky and Hesslein, 1995). However, at the sites included in the current study, phytoplankton biomass is high and transparency is low; and benthic primary productivity is likely to be strongly limited by the low-light conditions (Vadeboncoeur et al., 2003). Based on light attenuation coefficients and site depths as reported by Poste et al. (2013), the mean percentage of light reaching the bottom at our study sites never exceeded (and was often much lower than) 1% of surface irradiance. As such, pelagic carbon sources should dominate the

energy flow in these lakes and the contribution of benthic algal carbon should be low, with even benthic organisms likely to be strongly reliant on pelagic detritus. Even in our most transparent lake, Nkuruba, the contribution of benthic algal carbon may be limited by the steep morphometry of this crater lake, which reduces the extent of the littoral zone.

The ranges in $\delta^{13}C$ observed in the consumer food webs of our low transparency study lakes were aligned with the observed ranges in phytoplankton $\delta^{13}C$ (Table S1, Figures S2, S3). The variability in consumer $\delta^{13}C$ was likely driven by variability in phytoplankton $\delta^{13}C$ driven by temporal and spatial differences in growth rates and the resultant reduction in degree of isotopic fractionation (Deuser et al., 1968; Hecky and Hesslein, 1995; Poste et al., 2013) rather than by differences in the incorporation of benthic vs. pelagic carbon sources (Poste et al., 2012). Previous research on these study lakes (Poste et al., 2012, 2013) has shown that variability in phytoplankton $\delta^{13}C$ is primarily determined by primary productivity, as evidenced by strong positive relationships between phytoplankton $\delta^{13}C$ and chlorophyll *a*. Similar positive relationships between $\delta^{13}C$ of POM and primary productivity have also been observed for lakes (Gu et al., 1996; France et al., 1997) and marine systems (Fry and Wainright, 1991). As such, the negative relationships that we observed between THg concentrations and $\delta^{13}C$ in fish from several study sites may suggest that at these sites, fish that are either directly or indirectly reliant to a higher degree on ^{13}C-enriched phytoplankton with high primary production and biomass tend to have lower THg concentrations. This study demonstrated a dependence of Hg in fish on phytoplankton productivity as parameterized by $\delta^{13}C$. These observations highlight the importance of understanding the site-specific factors that determine the range of $\delta^{13}C$ values encountered in fish (i.e. this metric does not always reflect the relative importance of benthic vs. pelagic primary carbon sources). These results also provide further evidence for the importance of primary productivity and phytoplankton growth rates in determining eventual THg concentrations at higher trophic levels.

3.4. Trophic status influences THg biomagnification

The regression slope of log-transformed THg concentrations against $\delta^{15}N$ (as a measure of trophic level) has been widely used to quantify and compare the biomagnification of contaminants among aquatic systems (Kidd et al., 1995; Borgå et al., 2012). More recently, regression of log(THg) against calculated TL (based on $\delta^{15}N$ values) and TMF values derived from these regressions have become common for characterizing the increase in contaminant concentrations per TL (Borgå et al., 2012). However, despite their regular use, relatively few studies have explored the ecological factors that influence these metrics (as discussed by Borgå et al., 2012).

Recent studies have sought to make use of the intercepts of log(THg) ~ TL regressions in order to gain insight into contamination, bioavailability and uptake at the base of the food web (Borgå et al., 2012). For example, Kidd et al. (2012) observed a negative relationship between regression intercepts and total phosphorus concentrations (as an indicator of trophic status) across several temperate lakes, indicating lower contaminant uptake where trophic status is higher. In the current study, we did not observe any significant relationships between log(THg) ~ TL regression intercepts and measured water quality parameters. However, when we compared the intercepts of lines without significantly different slopes (Table 3) we found that the only significant difference that emerged was that Murchison Bay had a higher intercept than Lake Edward ($P < 0.001$). This likely reflects the difference in degree of contamination between this urban embayment (mean THg in water of 1.3 ng/L) and the relatively unimpacted Lake Edward (where THg in water ranged from 0.37 to 0.65 ng/L).

In the current study, log(THg) ~ $\delta^{15}N$ regression slopes for fish ranged from 0.08 in Lakes George and Saka to 0.22 in Lakes Albert and Nkuruba while TMF values (based on log(THg) ~ TL regressions; Fig. 3a) for THg ranged from 1.9 to 5.6 (Table 3). Previous studies in African lakes have reported log(THg) ~ $\delta^{15}N$ regression slopes ranging from 0.12 to 0.28 (Table 1), which correspond to TMF values (assuming a ^{15}N trophic enrichment factor of 3.4‰ per TL; Post, 2002) between 2.8 and 9.0. Log(THg) ~ $\delta^{15}N$ regression slopes have been reported from tropical, temperate and arctic freshwater systems around the world (Lavoie et al., 2013); however, to our knowledge, the regression slopes from hypereutrophic Lakes George (0.08 ± 0.02), Mburo (0.09 ± 0.02) and Saka (0.08 ± 0.03) represent some of the lowest reported slopes (for regressions based on fish data) in lake food webs to date. However, if the trophic enrichment of ^{15}N in these productive tropical lakes was lower than the assumed value of +3.4‰ per trophic level, then TL values and the number of trophic transfers would be underestimated, leading, in

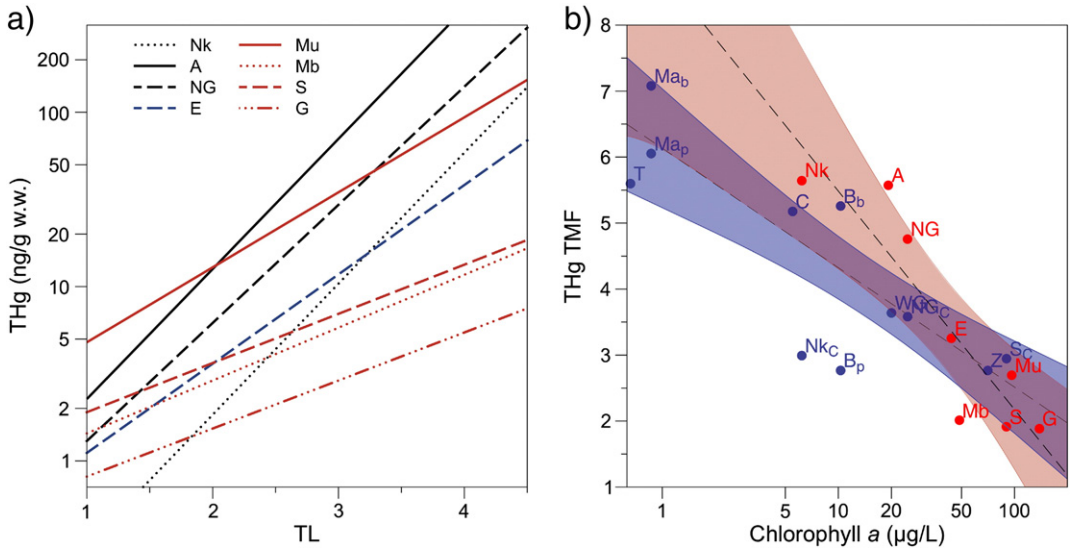

Fig. 3. a. Linear regressions between log-transformed THg concentrations in fish and calculated trophic level for all study sites (regression coefficients and ANCOVA post-hoc multiple comparison results are shown in Table 2); sites shown in red are hypereutrophic and in black are mesotrophic or eutrophic, Lake Edward is shown in blue since the there is a strong gradient from hypereutrophic to eutrophic conditions in the area where food web samples were collected; and b. linear regressions between THg TMF and log-transformed Chl *a* for sites in the current study alone (red site labels and 95% confidence interval shaded red; $n = 8$, TMF = 8.8 ± 1.2 − (3.3 ± 0.7) * ($\log_{10}$Chl *a*), $r^2 = 0.81$, $P < 0.01$) and in combination with literature data for other sub-Saharan African lakes (blue site labels and 95% confidence interval shaded blue; $n = 19$, TMF = 6.1 ± 0.4 − (1.8 ± 0.3) * ($\log_{10}$Chl *a*), $r^2 = 0.67$, $P < 0.0001$). Site codes are defined in Tables 1 and 2.

fact, to an overestimation of the already low biomagnification (or vice versa if enrichment of ^{15}N was higher than 3.4‰ per trophic level). It should also be noted that given the lack of data available regarding the proportion of Hg present as MeHg in tropical African fish, calculating TMFs based on THg rather than MeHg may carry a higher degree of uncertainty than in temperate systems, where MeHg generally makes up most of the Hg present in fish (Bloom, 1992).

We found that log(THg) ~ TL regression slopes were significantly higher in Lakes Albert, Nkuruba and Napoleon Gulf than in Lakes George, Saka, Mburo and Murchison Bay (Table 3, Fig. 3a). The regression slope for Lake Edward was intermediate, and did not differ significantly from that of Lake Nkuruba, Lake Albert or Murchison Bay (Table 3). These two main groups of sites can also be divided based on trophic status, with the first group of sites consisting of mesotrophic and eutrophic lakes and the second group of sites consisting entirely of hypereutrophic lakes (Table 2).

We also observed a strong negative relationship between TMF and log-transformed chlorophyll *a* (as an indicator of trophic status) across all study sites ($r^2 = 0.81$, $P = 0.002$; Fig. 3b), indicating that in lakes with higher primary productivity and phytoplankton biomass, biomagnification is occurring at a lower rate. These differences are unlikely to be due to differences in food web structure given that, despite nearly identical fish assemblages and food web structure (based on stable isotope analysis, Table S1), Lakes George and Edward (which receives its major inflow from Lake George) had highly divergent regression slopes (Fig. 3a). Similarly, as previously reported by Poste et al. (2012, Fig. 3a), Napoleon Gulf and Murchison Bay, two embayments in northern Lake Victoria, also had significantly different regression slopes despite strong similarities in species composition and food web structure, with a lower TMF in hypereutrophic Murchison Bay than in eutrophic Napoleon Gulf. These differences indicate that growth dilution through the consumer trophic levels is occurring, whereby fish are growing more quickly in higher productivity systems, resulting in reduced Hg concentrations in higher trophic level fish (Stafford and Haines, 2001; Simoneau, 2005; Lavigne et al., 2010).

We also sought to explore whether the negative relationship observed between THg TMF and trophic status (based on chlorophyll *a*) was applicable for other sub-Saharan African study sites as well. TMFs for THg were calculated from previously reported log(THg) ~ $\delta^{15}N$ regression slopes and mean chlorophyll *a* concentrations for each site were taken from the literature (Table 1). Thruston Bay (Napoleon Gulf, Lake Victoria; Campbell et al., 2004) had a much higher calculated TMF (9.0) than at any other site (including very nearby sites), potentially due to the small number of fish sampled ($n = 23$). Based on Cook's distance test, Thruston Bay was identified as being too influential on the regression results ($D_i = 0.58$, where cut-off was 0.33 based on Fox (1991)), and was therefore removed from the analysis (removal of this site increased the r^2 value from 0.29 to 0.67 and decreased the P value from 0.014 to 1.88×10^{-5}); no other sites were identified as being outliers based on this test. We found that much of the variability in TMF across all sites (current study along with literature data) could be explained by the negative relationship between TMF and chlorophyll *a* ($r^2 = 0.67$, $P = 1.88 \times 10^{-5}$; Fig. 3b), and as such, in tropical African lakes, trophic status appears to be an important driver not only of THg uptake at the base of the food web, but of TMFs as well. These processes may also explain the low Hg biomagnification (and subsequent fish Hg concentrations) observed in hypereutrophic Lake Taihu (Yan et al., 2010) as well as in (presumably highly productive) Chinese aquaculture ponds (Cheng et al., 2011).

Few studies to date have explored the influence of lake trophic status on contaminant biomagnification, and recent studies have yielded contradictory results, leading to a debate in the literature (Verburg et al., 2014; Verburg, 2014; Clayden et al., 2014), pointing to a strong need for additional field data. In a meta-analysis of literature data Lavoie et al. (2013) found a positive relationship between biomagnification of THg and chlorophyll *a*, while this relationship was found to be negative for MeHg. Previous field based studies have identified positive relationships between biomagnification of THg and total phosphorus (Kidd et al., 2012) and chlorophyll *a* (Verburg et al., 2014), and positive correlations between total phosphorus and TMF of some polychlorinated biphenyls (PCBs) and organochlorine pesticides (Houde et al., 2008). Meanwhile, Gantner et al. (2010b) found no relationship between TMF and chlorophyll *a* for arctic char food webs across 18 ultraoligotrophic arctic lakes, and Clayden et al. (2013) report a negative relationship between trophic status and biomagnification of THg in 11 Canadian lakes, based on principal components analysis. A recent study in New Zealand comparing Hg trophodynamics along a three-lake gradient in trophic status (Verburg et al., 2014) revealed that higher trophic status was related to lower THg in phytoplankton (as observed in the current study), but higher THg biomagnification (in contrast to the current study). Verburg et al. (2014) suggest that this may reflect a link between the prevalence of eutrophication-driven anoxia and MeHg availability. The contrast between the observations of Verburg et al. (2014) and the results of the current study may reflect the mixing dynamics of the Ugandan study sites, where diurnal stratification and daily circulation of the whole water column are common (Poste et al., 2013), allowing for replenishment of oxygen.

Although several studies have observed a negative relationship between lake primary productivity and fish Hg concentrations (Lange et al., 1993; Kidd et al., 1999; Rypel, 2010), the current study identifies a strong negative relationship between productivity and TMF, indicating that in these tropical African lakes, biomagnification (and not only uptake) of Hg may be moderated by lake trophic status. These processes, as well as trophic status-mediated differences in uptake at the base of the food web (i.e. phytoplankton bioconcentration), have important implications with respect to how Hg concentrations in water translate to Hg concentrations in top predators and subsequent risk to human consumers, as evidenced in the case study of Lakes Edward and George below.

3.5. Confirmatory case study — Lakes Edward and George

Lake George is a shallow hypereutrophic lake with an outlet that drains (via the Kazinga Channel) into the deeper and less productive Lake Edward (Poste et al., 2013), one of the least studied large lakes of the world. These lakes are of interest for a comparative study of THg trophodynamics given that they have very different limnological characteristics (especially with respect to trophic status) and yet they are connected, and have very similar species assemblages and food web structure, with common species occupying similar trophic levels (Table S1).

THg concentrations in water from Lake George were elevated relative to the remaining study sites (aside from Murchison Bay, a highly polluted urban embayment), while offshore Lake Edward (EdO) had the lowest observed THg concentrations in water for our study sites (Table 2, Fig. 4a). Mercury concentrations at the nearshore Lake Edward study site (EdK), near the inflow from Lake George, were intermediate between those observed in Lake George and those in offshore Lake Edward (EdO; Table 2, Fig. 4a), likely reflecting the strong hydrological connection between Lake George and nearshore Lake Edward (EdK). Meanwhile, the spatial pattern in phytoplankton THg concentrations across these study sites was reversed relative to THg in water (Fig. 4a), with the highest concentrations observed in phytoplankton from offshore Lake Edward (EdO), and the lowest concentrations observed in phytoplankton from Lake George (although these differences were not statistically significant). This reflects the difference in THg_{PHYTO}/THg_{WATER} between the three sites, given that THg accumulation was relatively lower in lakes with higher trophic status (Fig. 2). In Lakes Edward and George, biological dilution at the base of the food web appears to overshadow the spatial differences in THg concentrations in water with respect to eventual THg concentrations in phytoplankton. These differences are also unlikely to be due to between-site differences in Hg

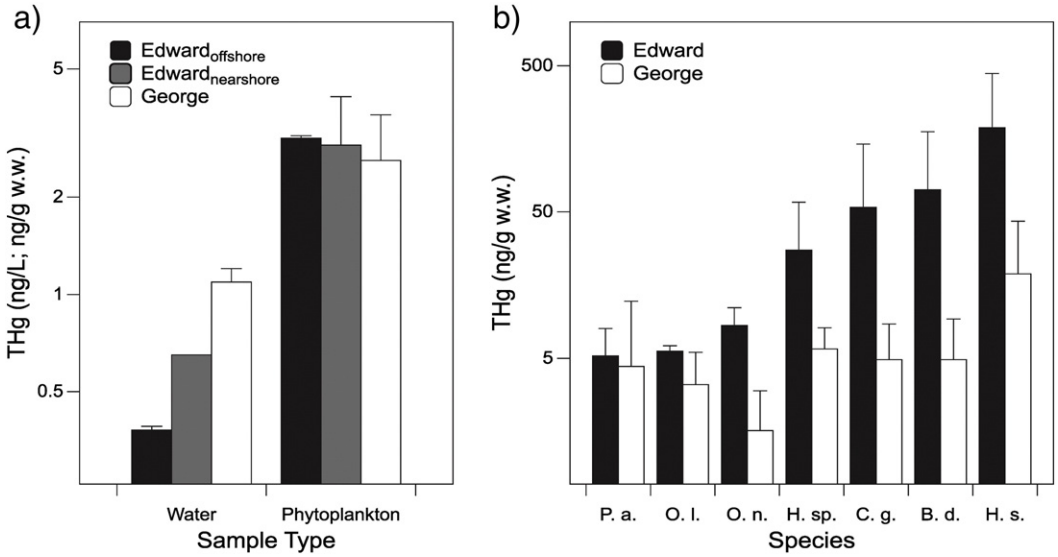

Fig. 4. a. THg concentrations in water (ng/L) and phytoplankton (ng/g w.w.) from offshore Lake Edward (EdO), nearshore Lake Edward (EdK) and Lake George; and b. THg concentrations in common fish species for Lakes Edward and George. Species abbreviations are as follows: P. a. = *Protopterus aethiopicus*, O. l. = *Oreochromis leucostictus*, O. n. = *Oreochromis niloticus*, H. sp. = *Haplochromis* sp., C. g. = *Clarias gariepinus*, B. d. = *Bagrus docmac*, and H. s. = *Haplochromis squamipinnis*.

methylation since if anything, methylation should be higher in productive Lake George, where high concentrations of organic matter can support increased methylation (Ullrich et al., 2001), and where mean water column light intensity is generally lower (Poste et al., 2013), suggesting reduced potential for MeHg loss through photodemethylation.

At consumer trophic levels (where Lake Edward was treated as a single site), the lower Hg biomagnification in Lake George (TMF of 1.9 ± 0.2) compared to Lake Edward (TMF of 3.3 ± 0.5; Fig. 3a, Table 3) appears to lead to reduced THg concentrations in fish from Lake George relative to Lake Edward (Fig. 4b). For all fish species common to both lakes, except for *P. aethiopicus*, THg concentrations were significantly ($P < 0.05$) higher in Lake Edward than in Lake George (Fig. 4b) despite the fact that these species occupy similar trophic positions in both lakes (Table S1). These results demonstrate how differences in bioconcentration at the base of the food web and biomagnification through consumer trophic levels, driven by trophic status moderated differences in phytoplankton and fish growth rates, can lead to widely divergent THg concentrations in fish. In the current study, the highest THg concentrations in fish (some of the highest ever reported for uncontaminated sites in Africa) were encountered in Lakes Albert and Edward, which had some of the lowest THg concentrations in water. Meanwhile, consistently low concentrations (THg below 16 ng/g in 66 of the 68 fish sampled) were observed in Lake George, where THg concentrations in water were three-fold higher (Table 2).

3.6. Conclusions

Mercury concentrations in fish from these African lakes appeared to be more strongly determined by processes at the base of the food web than by mercury concentrations in water. Year-round high phytoplankton biomass and growth rates in productive tropical lakes may reduce the potential for high THg in fish. Growth dilution of Hg in phytoplankton may reduce Hg concentrations at the food web base, while growth dilution at consumer trophic levels may reduce Hg biomagnification, both of which contribute to reduced Hg in aquatic food webs, thereby reducing risk of dietary exposure of Hg through fish consumption. In particular, trophic status-mediated differences in biomagnification can have progressively stronger effects on top predator Hg concentrations with increasing aquatic food chain length.

These results highlight the need to consider the potential effects of changes in lake nutrient status (e.g. through cultural eutrophication, or nutrient reduction measures) on past, present and future patterns in Hg contamination of aquatic food webs.

Acknowledgments

We thank Uganda's National Fisheries Resources Research Institute and Dr. Lauren Chapman's research team in Uganda for providing workspace and assistance in the field. Greg Lawson (Environment Canada, Aquatic Contaminants Research Division) carried out total mercury analysis in water samples. This work was funded through an International Development Research Council Doctoral Research Award to AEP.

Appendix A. Supplementary data

Supplementary data to this article can be found online at http://dx.doi.org/10.1016/j.scitotenv.2014.10.094.

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