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Response to Comment on "Mercury Biomagnification through Food Webs Is Affected by Physical and Chemical Characteristics of Lakes"

In his response to our paper on how physical and chemical characteristics of lakes affect mercury (Hg) biomagnification through food webs,[1] Piet Verburg states that "no evidence has been presented, by the authors or elsewhere, for biomagnification decreasing with increasing biomass in lakes as an effect of biomass dilution." Biomass and growth dilution have been cited in other studies as possible mechanisms for reducing concentrations of Hg at the base of aquatic food webs or in upper-trophic-level biota, respectively.[2,3] Our paper on 11 lake food webs in Kejimkujik National Park and National Historic Site (KNPNHS), Nova Scotia, Canada, used principal component analysis (PCA) of chemical and physical variables for these lakes and then regressions of biomagnification slopes (log Hg versus δ^{15}N in a food web) against these components to understand whether ecosystem characteristics were related to Hg biomagnification. We found that a component with higher aqueous nutrient (calcium, Ca; total organic carbon, TOC; total nitrogen, TN; and total phosphorus, TP), methyl mercury (MeHg), and chloride (Cl) loadings (hereafter, the "nutrients/MeHg/chloride" factor) was inversely related to Hg biomagnification slopes in these lakes. As described in the paper, we used a PCA and regression approach to examine variability in Hg biomagnification slopes because many lake characteristics are inherently highly correlated (Table S4[1]), and we wanted to avoid arbitrarily isolating one or a few variables as predictors.[4]

Indeed biomass dilution is mentioned in two places in the Results and Discussion section of our paper.[1] First, we state that "This negative relationship (between slopes and the nutrients/MeHg/Cl factor) suggests higher Hg biomagnification in lakes with lower trophic status, and supports other studies showing that biotic mercury concentrations can undergo biomass and growth dilution in more nutrient-rich systems." Second we conclude with "Although the biomagnification slopes herein were similar to those of other studies, we found that lakes in KNPNHS with lower nutrients/MeHg/Cl scores had higher slopes, possibly because of biomass and growth dilution or effects of nutrients on Hg bioavailability." We feel that the results of this PCA approach do support these statements. In particular, Table S8[1] shows that Hg biomagnification slopes were best described by the "nutrients/MeHg/chloride" factor according to our model selection criteria. Furthermore, Table S3[1] shows that Ca and TP (which, as Verburg states, is "usually a better correlate of plankton biomass than TN") are the most influential variables in this factor, followed by TN, Cl, MeHg and TOC. We never link Hg biomagnification slopes (or biotic concentrations) *solely* to measures of lake biomass or productivity. The nature of our statistical analyses precludes us from doing that and, as Verburg mentions, we mainly use indirect measures (proxies) of biomass such as aqueous TP. In the "Hg Biomagnification through Food Webs" section of the paper, we present biomass dilution as one possible mechanism to explain the results of regression analyses using principal components. In the next sentence of this section, we then discuss the high loading of calcium (a micronutrient) in this component and how that cation may affect Hg biomagnification, and go on to say that "Most of the individual variables with strong loadings in the nutrients/MeHg/chloride factor were also highly correlated with TOC".

Most of Verburg's criticisms stem from a correlation matrix that shows how biomagnification slopes are related to chemical and physical characteristics of the lakes (Table S4[1]). On the basis of discussions with other researchers, feedback from reviewers, and consideration of the ecological statistics literature (e.g., refs 4 and 5), we felt it was inappropriate to rely solely on correlations using individual lake variables to interpret our results because we would, by necessity, have to exclude interactions between variables and arbitrarily assume that one was more important than another. Other studies have shown that abiotic cycling and speciation of Hg in lakes depends on complex and poorly understood interactions of many factors, including redox conditions, quality and content of organic matter, concentrations and speciation of other metals (e.g., iron, manganese), photochemical reactions, etc.[6,7] As such, we feel Verburg's criticisms about the relative strength of correlations between biomagnification slopes and TOC, TN, or TP are moot because it is incorrect to suppose that they can be used to infer the relative importance of variables affecting Hg biomagnification slopes or Hg concentrations in various organisms, and we avoided making any suppositions to this effect in our paper. Nonetheless, we included Table S4[1] in the Supporting Information to facilitate comparisons to other studies and future research in this area.

We appreciate the mechanisms Verburg presents as possible explanations for how Hg biomagnification slopes may vary among food webs. It is a challenge to understand among-system differences in slopes given the different mechanisms that may be at play, and we welcome suggestions on how to identify the ones that are involved in this variability. Interestingly, similar questions are being posed in food web studies of other biomagnifying compounds, such as the persistent organic pollutants (POPs[8,9]), because ecosystem characteristics are also related to the slopes of POPs versus δ^{15}N regressions.[10] A recent global meta-analysis of all marine and freshwater studies that have quantified Hg biomagnification using δ^{15}N found significantly higher regression slopes in freshwater systems at higher than lower latitudes and some influences of water chemistry (pH, TP, dissolved organic carbon) on slopes from a subset of studies.[11] However, the variability in the Hg biomagnification slopes that is explained is low (R^2 = 0.10–0.21 for slopes of log MeHg versus δ^{15}N), and this clearly shows that there are other factors to consider both within and among regions. As we state in our paper, comparisons of systems across a broader range of conditions, such as this one,[11] require similar experimental designs and more supporting

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information on ecosystem characteristics to improve our understanding of Hg biomagnification through aquatic food webs. We hope that our paper has presented some "food for thought" on why some systems biomagnify Hg more than others and look forward to seeing other advances in this area.

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