



# Mercury cycling in aquatic ecosystems and trophic state-related variables – Implications from structural equation modeling

Curtis D. Pollman \*

Aqua Lux Lucis, Inc., 8411 NW 55th Place, Gainesville, FL USA



## HIGHLIGHTS

- Structural equation modeling was used to evaluate variable interactions with fish Hg.
- Modeling was conducted using data from Florida lakes, streams, and the Everglades.
- The benefits of SEM as a tool for quantifying complex interrelationships are shown.
- MeHg signal is the most important determinant of fish Hg across all three systems.
- DOC effects on fish Hg are complex with competing antagonistic pathways.

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## ABSTRACT

Structural equation modeling (SEM) provides a framework that can more properly handle complex variable interactions inherent in mercury cycling and its bioaccumulation compared to more traditional regression-based methods. SEM was applied to regional data sets for three different types of aquatic ecosystems within Florida, USA – lakes, streams, and the Everglades – to evaluate the underlying nature (*i.e.*, indirect and direct) of the relationships between fish mercury concentrations and trophic state related variables such as nutrients, dissolved organic carbon (DOC), sulfate, and alkalinity. The modeling results indicated some differences in key variable relationships – for example, the effect of nutrients on fish mercury in lakes and streams was uniformly negative through direct and indirect pathways consistent with biodilution or eutrophication-associated effects on food web structure. Somewhat surprisingly, however, was that total phosphorus did not serve as a meaningful variable in the Everglades model, apparently because its effects were masked or secondary to the effects of DOC. What is perhaps a more important result were two key similarities across the three systems. First, the modeling clearly indicates that the dominant influence on fish tissue mercury concentrations in all three systems is related to variations in the methylmercury signal. Second, the modeling demonstrated that the effect of DOC on fish mercury concentrations was exerted through multiple and antagonistic pathways, including facilitated transport of total mercury and methylmercury, enhanced rates of methylation, and limitations imposed on bioavailability. Indeed, while the individual DOC pathways in the models were all highly significant (generally  $p < 0.001$ ), the *net* effect of DOC in each model was greatly reduced or insignificant. These results can help explain contradictory results obtained previously by other researchers in other systems, and illustrate the importance of SEM as a modeling tool when studying systems with complex interactions such as the aquatic mercury cycle.

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## 1. Introduction

Mercury (Hg) contamination in aquatic ecosystems has been an issue of considerable concern for over fifty years (McCurry, 2006) because highly toxic methylmercury (MeHg) readily bioaccumulates through aquatic food webs (*e.g.*, Watras and Bloom, 1992; Thera and Rumbold, 2014; Seixas et al., 2014), often to concentrations that pose risk to not only aquatic organisms, but also to terrestrial wildlife and

humans that consume higher trophic level organisms such as predatory fish obtained from these ecosystems. Risk from consumption and exposure to MeHg comes in several forms, including effects as a powerful neurotoxin (Clarkson, 1997; Clarkson et al., 2003; Grandjean et al., 2010) and reproductive impairment in fish and birds (Crump and Trudeau, 2009; Scheuhammer et al., 2007; Evers et al., 2008; Frederick and Jayasena, 2011).

Hg cycling in aquatic ecosystems and accumulation in aquatic foodwebs is quite complex and, although atmospheric inputs are the primary source for most aquatic ecosystems with elevated biota Hg concentrations, variations in biota Hg concentrations across aquatic

\* Tel.: +1 352 222 4039.

E-mail address: [cpollman@aqualuxlucis.org](mailto:cpollman@aqualuxlucis.org).

ecosystems within a geographic region characteristically reflect variations in hydrological and biogeochemical characteristics to a greater extent than variations in atmospheric inputs. For example, the coefficient of variation (CV) of modeled total Hg deposition fluxes (wet plus dry) to lakes distributed across Florida is 25% compared to a CV of 66% for total Hg concentrations measured in largemouth bass from the same lakes (normalized to 15 inches length) (data from Pollman, 2012a). Thus, if the relationship between atmospheric inputs of Hg and biota response for a given aquatic ecosystem is linear as indeed several studies suggest (Krabbenhoft et al., 2004; Orihel et al., 2006, 2007), variations in atmospheric inputs can only account for a minor fraction of the observed variations in biota concentrations. In contrast, the CV for a suite of key biogeochemical variables (alkalinity, DOC, chlorophyll *a*, sulfate, and total phosphorus) generally considered to be important in influencing aquatic Hg cycling all exceed (up to nearly 3×) that of the CV for largemouth bass Hg concentrations in the same lakes, and thus can easily account for most of the variance in biota concentrations.

The geographic scale of emission sources contributing to the delivery of atmospheric Hg to most aquatic resources with elevated biota Hg concentrations is regional to global (Seigneur et al., 2004; Gbor et al., 2007). Moreover, the contribution of current anthropogenic sources to global Hg deposition is only about one-quarter to one-third, with legacy anthropogenic sources contributing approximately one-third to three-fifths the current flux, and natural sources contributing the remaining fraction (Pirrone et al., 2010; Amos et al., 2013). While the legacy pool of Hg is likely recent in origin, and thus would expectedly have a fairly rapid turnover time in response to reductions in global emissions (Engstrom et al., 2013), the ability to mitigate the vast majority of impaired aquatic ecosystems over the immediate near term (years) by controlling local emissions is marginal at best. For example, based on CMAQ model estimates (UMAQL, 2012), emission sources within Florida contribute on average only ~2% of the total atmospheric Hg directly deposited to Florida's lakes and rivers/streams, and for ~96% of the lakes and streams the local contribution was 10% or less (Pollman, 2012a).

These two key aspects of aquatic Hg cycling (the controlling effect of biogeochemistry and the limited ability to mitigate Hg sources) suggest that mitigation approaches that focus on biogeochemistry may be more effective than controlling local Hg emissions. In a recent synthesis of studies on Hg bioaccumulation in the Great Lakes region, Evers et al. (2011) concluded “concentrations of mercury in biota are related to, and can be predicted from, environmental conditions in aquatic systems that are known to influence ecosystem sensitivity to mercury.” Understanding these conditions can potentially lead to management strategies for mitigating the Hg problem. For example, a number of studies have shown that biota Hg concentrations are inversely related to alkalinity or acid neutralizing capacity (ANC) (Håkanson et al., 1988; Spry and Wiener, 1991; Lange et al., 1993; Kamman et al., 2004; and Dittmann and Driscoll, 2009), and Yu et al. (2011) suggest that atmospheric emission controls of SO<sub>2</sub> (thus reducing acidic deposition) will likely result in decreasing biota Hg concentrations as well. Similarly, biota Hg concentrations have been shown to decrease in response to declining rates of sulfate deposition or loading, thus suggesting a management strategy of reducing sulfur emissions from sources impacting aquatic ecosystems where the availability of sulfate limits sulfate reduction and attendant Hg methylation rates (Hrabik and Watras, 2002; Drevnick et al., 2007; and Coleman Wasik et al., 2012). In addition, there has been considerable debate about whether reductions in total phosphorus (TP) and coupled changes in aquatic community structure resulting from the restoration of the Florida Everglades will exacerbate an already profound biota Hg problem (Fink et al., 1999; Green and Perko, 2001; Krabbenhoft et al., 2001). For example, restoration is expected to induce shifts in highly eutrophied areas of the northern Everglades characterized by simplified, less tightly coupled food webs, to more complex, tightly coupled food webs more conducive to promoting higher aquatic biota Hg concentrations (Stober et al., 2001).

One approach towards developing a better understanding of the biogeochemical variables that govern Hg cycling and bioaccumulation in aquatic ecosystems has been through either correlation analysis or constructing multivariate – in particular, multiple linear regression (MLR) – models that relate, for example, variations in fish tissue Hg concentrations to a set of independent biogeochemical variables. Such models characteristically are constructed across a broad set of lakes or streams and examples include studies conducted in Sweden (Håkanson et al., 1988), the Upper Midwest and Minnesota (Grieb et al., 1990; Wiener et al., 2006), the northeastern US, including the Adirondacks (Chen et al., 2005; Kamman et al., 2005; Simonin et al., 2008; Dittmann and Driscoll, 2009), and Florida (Lange et al., 1993; Pollman, 2012a).

An important assumption in the potential use of MLR models as management or assessment tools is that the independent variables are indeed truly independent. For example, a number of studies have shown that fish tissue Hg concentrations are inversely related to ANC and positively related to MeHg concentrations. At the same time, ANC typically declines and MeHg concentrations may increase in response to the acidifying effects of protons accompanying anthropogenic or excess sulfate concentrations. Thus the question arises – with changes in fish tissue Hg resulting from changes in sulfate loadings and resultant changes in ANC – how much of the fish tissue Hg response is due to direct changes in ANC and how much of the response is due to the effects of changing sulfate on methylation? Traditional MLR models only control for direct effects, and provide no information on indirect, and thus the *total* effect on response when the independent variable (e.g., sulfate) also is mediated by the effect of other variables in the model.

One approach that allows and more properly accounts for the effects of mediating variables is structural equation modeling (SEM) which, through the development of a hypothesized model, seeks to reproduce the covariance structure of the observed data (Ullman, 2007). SEM can include linking confirmatory factor analysis (which can be used to construct latent variables representing unmeasured processes) with path analysis and is often referred to as *causal analysis* (Ullman, 2007) because the approach often is used to test or confirm an *a priori*, hypothesized model (Austin, 2007). While SEM does not actually prove causality, it does provide a framework for selecting or rejecting hypotheses based on the empirical data (Iriondo et al., 2003). Although the roots of SEM extend back in time over 90 years with Wright's (1921, 1934) development of path analysis, application of SEM to aquatic systems is relatively novel and rare. Examples include using SEM to model macroinvertebrate distribution in riverine ecosystems (Bizzi et al., 2013) and estuaries (Malaeb et al., 2000), submersed macrophyte biomass (Hung et al., 2007), lacustrine phytoplankton dynamics (Arhonditsis et al., 2006; Liu et al., 2010; Korhonen et al., 2011), and MeHg cycling and bioaccumulation in the Florida Everglades (Stober et al., 2001). In addition, SEM has been used to construct models with a clear environmental management focus (La Peyre et al., 2001; Stober et al., 2001; and Reckhow et al., 2005). For example, La Peyre et al. (2001) used SEM to model national wetland protection efforts as a function of five latent variables (defined as economic capital, social capital, government characteristics, environmental characteristics, and land-use pressure). Their model suggested that variations in social capital had the greatest influence on wetland protection efforts, and concluded that continued focus on social development was necessary to further wetland protection. Readers interested in learning more about SEM, including the underlying mathematics, basic elements of constructing an SEM, issues and limitations, and detailed examples of its implementation, are directed to the papers by Malaeb et al. (2000) and Ullman (2007).

## 2. Materials and methods

### 2.1. Data sets

The objective of this paper is to use SEM to model fish Hg bioaccumulation in response to key biogeochemical variables in three different

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