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Decline in methylmercury in museum-preserved bivalves from San Francisco Bay, California

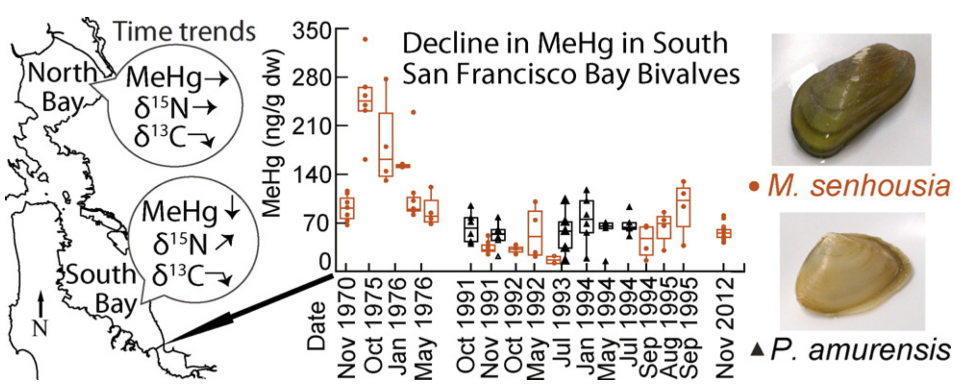
Allison C. Luengen [a,*], Heather M. Foslund [a], Ben K. Greenfield [b,1]

[a] Environmental Sciences Department, University of San Francisco, 2130 Fulton Street, San Francisco, CA 94117, USA
[b] Environmental Health Sciences Division, School of Public Health, University of California, Berkeley, 50 University Hall #7360, Berkeley, CA 94720, USA

HIGHLIGHTS

- First reported long-term decline in methylmercury in San Francisco Bay biota
- Methylmercury concentration decline in preserved bivalves since mine closure
- Stable isotopes indicate methylmercury trends not attributable to food web changes.
- $\delta^{15}N$ and $\delta^{13}C$ trends likely caused by both natural and anthropogenic drivers

GRAPHICAL ABSTRACT



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ABSTRACT

There are ongoing efforts to manage mercury and nutrient pollution in San Francisco Bay (California, USA), but historical data on biological responses are limited. We used bivalves preserved in formalin or ethanol from museum collections to investigate long-term trends in methylmercury (MeHg) concentrations and carbon and nitrogen isotopic signatures. In the southern reach of the estuary, South Bay, MeHg in the Asian date mussel (*Musculista senhousia*) significantly declined over the study duration (1970 to 2012). Mean MeHg concentrations were highest (218 ng/g dry weight, dw) in 1975 and declined 3.8-fold (to 57 ng/g dw) by 2012. This decrease corresponded with closure of the New Almaden Mercury Mines and was consistent with previously observed declines in sediment core mercury concentrations. In contrast, across all sites, MeHg in the overbite clam (*Potamocorbula amurensis*) increased 1.3-fold from 64 ng/g dw before 2000 to 81 ng/g dw during the 2000s and was higher than in *M. senhousia*. Pearson correlation coefficients of the association between MeHg and $\delta^{13}C$ or $\delta^{15}N$ provided no evidence that food web alterations explained changing MeHg concentrations. However, isotopic composition shifted temporally. South Bay bivalve $\delta^{15}N$ increased from 12‰ in the 1970s to 18‰ in 2012. This increase corresponded with increasing nitrogen loadings from wastewater treatment plants until the late 1980s and increasing phytoplankton biomass from the 1990s to 2012. Similarly, a 3‰ decline in $\delta^{13}C$ from 2002 to 2012 may represent greater utilization of planktonic food sources. In a complimentary 90 day laboratory study to validate use of these preserved specimens, preservation had only minor effects (<0.5‰) on $\delta^{13}C$ and $\delta^{15}N$. MeHg increased following preservation but then stabilized. These are the first documented long-term trends in biota MeHg and stable isotopes in this heavily impacted estuary and support the utility of preserved specimens to infer contaminant and biogeochemical trends.

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* Corresponding author.
E-mail addresses: aluengen@usfca.edu (A.C. Luengen), hmfoslund@gmail.com (H.M. Foslund), greenfieldben1@gmail.com (B.K. Greenfield).
[1] Current address: Biology Department and Environmental Sciences Program, Southern Illinois University – Edwardsville, IL 62026, USA.

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1. Introduction

Mercury pollution of estuaries poses a global threat to wildlife and human health. Humans are exposed to the organometallic form, methylmercury (MeHg), primarily through consumption of coastal and estuarine seafood (Sunderland, 2007). Wildlife that depend on the productive estuarine habitat are also exposed, and reproductive impairment has been documented in birds (Eagles-Smith et al., 2009; Scheuhammer et al., 2007). Accordingly, there are local, national, and international efforts to reduce mercury. However, our ability to assess the efficacy of regulatory efforts is limited by lack of long-term studies on MeHg concentrations in biota, especially in coastal ecosystems (Lambert et al., 2012).

To evaluate temporal trends, some studies have relied on total mercury (Hg_T) measurements, which include both inorganic and organic forms, in fish (Bhavsar et al., 2010; Gandhi et al., 2014; Kraepiel et al., 2003). Measuring Hg_T works well at the top of the food chain where >95% of the Hg_T is MeHg (Bloom, 1992). However, one disadvantage of this approach is that fish Hg_T concentrations may be decoupled from mercury inputs. For example, Monson et al. (2011) found that Hg_T concentrations in walleye in Ontario declined from 1970 to 1990, reflecting a general decrease in atmospheric inputs in North America and Europe. However, walleye concentrations subsequently increased from the mid-1990s to 2009, possibly due to changes in the food web (Monson et al., 2011).

At our study site in San Francisco Bay, California, USA (Fig. 1), the temporal trends in fish are particularly perplexing. There has been no decline in Hg_T in striped bass from the 1970s to the present (Davis et al., 2016; Greenfield et al., 2005), despite the closure of the New Almaden Mercury Mining District (hereafter New Almaden) in 1975, and ongoing efforts to understand and control MeHg pollution in San Francisco Bay and its watersheds (Davis et al., 2012). New Almaden was once the nation's largest mercury mining district, and it drained into lower San Francisco Bay via the Guadalupe River (Conaway et al., 2008; Davis et al., 2012). Mercury concentrations recorded in sediment cores do show a decline since the mid-20th century (Conaway et al., 2004; Donovan et al., 2013) and since 1970 (Hornberger et al., 1999), so perhaps fish are not the best biomonitor for long-term trends in this estuary.

Another approach is to focus on invertebrates lower in the food chain, such as bivalves, which efficiently accumulate and transfer metals (Pan and Wang, 2011). Resident bivalves have been successfully used as biosentinels to record long-term declines in other metals (e.g., Cu and Ag) in San Francisco Bay (Hornberger et al., 2000), and are widely and successfully employed for contaminant trend biomonitoring (e.g., Lauenstein and Daskalakis, 1998; Luengen et al., 2004; Melwani et al., 2014). Both resident and transplanted mussels have been central to long-term monitoring efforts in California, and nationwide, through the "Mussel Watch" programs (Melwani et al., 2014). Mussel Watch includes three sites in San Francisco Bay, although reliable data are available only from 1986 to 2009 and only for Hg_T (Melwani et al., 2013). Unlike in fish, MeHg is only a fraction of Hg_T in bivalves, and varies considerably, between 12 and 60% (Francesconi and Lenanton, 1992; Pan and Wang, 2011; R. Stewart, USGS, pers. comm.). Studies with transplanted mussels indicate that that measurements of Hg_T in bivalves are not useful for predicting mercury concentrations in higher trophic levels (Gunther et al., 1999), because only the methylated form biomagnifies. Another reason to focus on the methylated form is that it is difficult to contaminate samples (even preserved ones) with MeHg, unlike Hg_T (Vo et al., 2011).

Unfortunately, there are no historical measurements of MeHg in invertebrates because methods to measure MeHg were not established until 1988 (Bloom and Fitzgerald, 1988) and not widely applied until much later. This lack of historical data leaves the analysis of archived specimens as the only potential avenue for determination of past concentrations. Along those lines, Vo et al. (2011) recently used bird

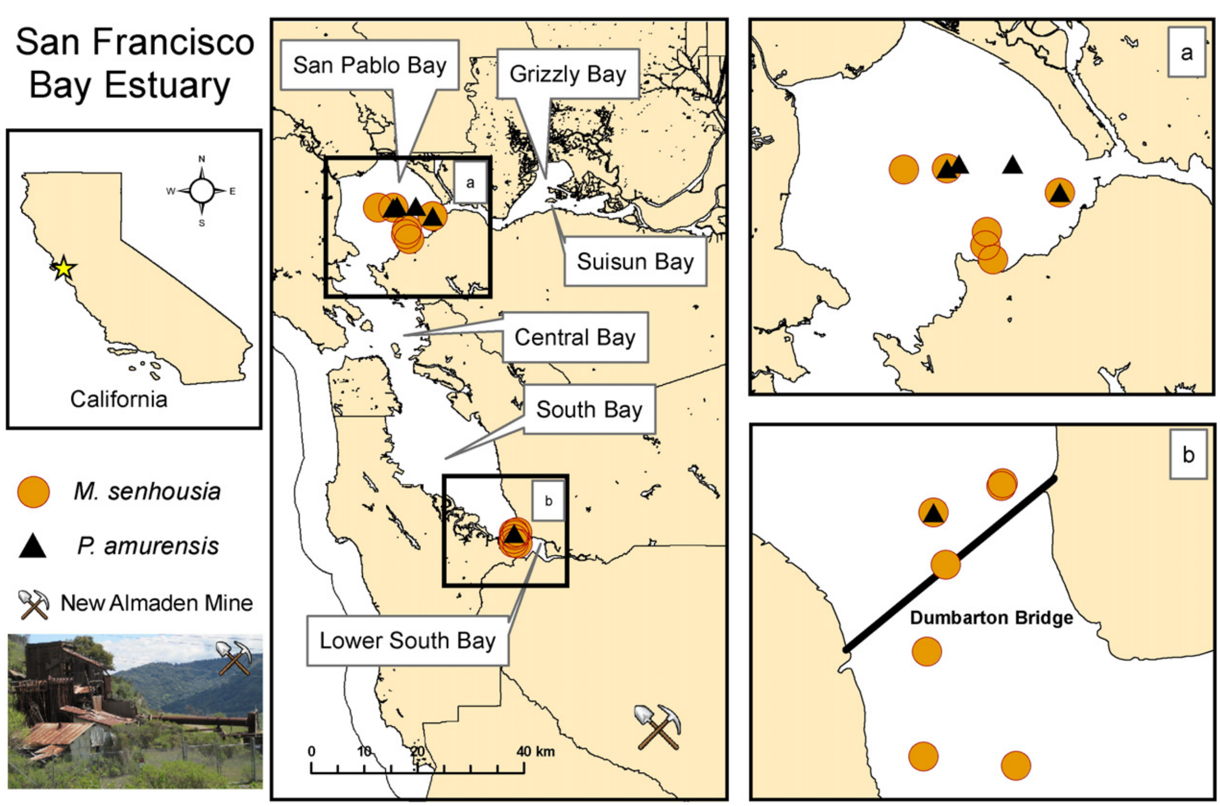

Fig. 1. San Francisco Bay Estuary is divided into distinct hydrological sub-embayments. Bivalves were available in natural history collections from both the well-flushed northern reach of the estuary and the more stagnant southern reach, which also has the former New Almaden mercury mining district in its watershed. Specimens of the Asian date mussel, *Musculista senhousia*, were available from 1970 to 2012. Specimens of the overbite clam, *Potamocorbula amurensis*, were available from 1988 through 2002.

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