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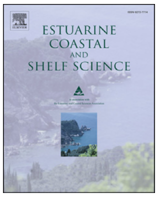

Complex patterns in fish – sediment mercury concentrations in a contaminated estuary: The influence of selenium co-contamination?

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ABSTRACT

Environmental mercury (Hg) loads do not always correspond to Hg concentrations in resident fish and selenium (Se) presence has been reported to play a pivotal role in mitigating Hg bioaccumulation. Total mercury (THg), methylmercury (MeHg) and Se concentrations were measured in sediments and a benthic fish species (*Platycephalus bassensis*) from a contaminated estuary (Derwent Estuary, Tasmania). Elevated sediment concentrations of Se did not result in increased Se concentrations in fish, but low concentrations of Se were associated with increased MeHg bioavailability (% MeHg) from sediments to fish. Where MeHg (≈99% of total Hg) concentration in fish was high Se uptake also increased, indicating that maintaining positive Se:Hg ratios may reduce the toxicity of MeHg. MeHg was detectable in sediments throughout the estuary, and a molar excess of THg over Se suggested that there was insufficient Se to prevent methylation from the sediments. Se:Hg ratios of less than 1.0 in sediments, coupled with high %MeHg fraction and high biotic sediment accumulation factors for MeHg ($\mathrm{BSAF_{MeHg}}$), indicated that the lower region of the Derwent Estuary could be a hotspot for Hg methylation, despite having significantly lower THg concentrations. In contrast, Hg bioavailability to fish from sediments close to the source may be reduced by both inorganic Hg species complexation and lower methylation rates. There was a strong association between THg and Se in estuarine sediments, suggesting that Se plays an important role in sediment Hg cycling and should be a key consideration in any future assessments of Hg methylation, bioavailability and bioaccumulation.

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1. Introduction

The spatial variation of methylmercury (MeHg) production and bioaccumulation in estuarine food webs is broadly understood but poorly characterized (Mason and Lawrence, 1999; Davis et al., 2012). Uptake and transfer of mercury (Hg) between the biotic and abiotic components is not straightforward (Chen et al., 2009), with limited data supporting the concept that elevated Hg concentration in aquatic environments leads directly to high MeHg levels in fish e.g. Brumbaugh et al., 2001; Munthe et al., 2007. Regions with high Hg environmental loads may show low bioaccumulation rates if net methylation rates are low; conversely, low environmental Hg concentrations may result in high fish tissue loadings as a result of raised methylation efficiency (Brumbaugh et al., 2001). Understanding the mechanisms which underpin this variability is especially important to quantifying and managing Hg exposure risks and to developing appropriate management actions (Tom et al., 2010; Davis et al., 2012).

The Derwent Estuary, in southeast Tasmania, exhibits large differences in the THg concentrations (Total Hg = inorganic Hg + organic Hg) in its sediments (Jones et al., 2003). The differences in sediment Hg values are notably not reflected in the THg concentrations of the resident benthic fish sand flathead (*Platycephalus bassensis*) (Jones et al., 2013a). The industrialized middle reaches of the estuary are located ≈20 km from the mouth, and have consistently high THg concentrations in both sediments and sand flathead as a result of historic inputs from a zinc smelter and paper mill (Bloom and Ayling, 1977; Green and Coughanowr, 2003). Conversely, a large and relatively shallow embayment on the lower eastern side of the estuary, called Ralphs Bay, has low sediment Hg levels but high Hg concentrations in fish (Jones et al., 2003, 2013a). Despite 40 years of Hg research in the Derwent Estuary, the reasons for this paradox remain unexplored.

The quantification of biotic exposure to MeHg is complicated by the presence of selenium (Se), a known co-contaminant from metallurgical processing (Yang et al., 2008). Formation and

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excretion of Se-biomolecules by sediment-dwelling organisms results in the production of the mineral selenide (Maher et al., 2010), which is capable of sequestering Hg^{2+} and forming mercuric selenide (HgSe) (Yang et al., 2008). HgSe formed in sediments is relatively inert, and may reduce the concentration of Hg available for methylation (Yang et al., 2008). This process diverts Hg away from biogeochemical cycling into methylated forms; so where Se is absent from sediments, 'hotspots' of Hg bioaccumulation may occur (Ralston and Raymond, 2010). Although it is an essential trace element, Se is toxic at high levels, and can bioaccumulate through food pathways similar to Hg (Cuvin-Aralar and Furness, 1991). In freshwater systems elevated concentrations of Se in fish have been linked to reduced Hg concentrations (Chen et al., 2001; Belzile et al., 2006; Sackett et al., 2010), yet this has never been documented for estuarine systems. Se concentrations in sediments and sand flathead have never been measured in the Derwent Estuary and could explain, at least in part, the spatial disparity between Hg concentrations in sand flathead populations and the sediments.

The objective of this study was to evaluate if the co-occurrence of Se may be mitigating Hg bioavailability from sediments and reducing fish bioaccumulation, and whether this may be the reason why Hg concentrations in sand flathead are lower than might otherwise be expected. We addressed this by: (1) measuring the THg, MeHg and Se concentrations within the sediments and in the muscle tissue of resident populations of sand flathead in the Derwent Estuary; (2) examining the Se:Hg ratios in Derwent Estuary fish and sediment for evidence of spatial variation, and (3) inspecting the relationship between Se and Hg (THg and MeHg) in the sediments to assess evidence of reduced Hg bioavailability.

2. Methods

2.1. Site selection

The Derwent Estuary (42° 54′S, 147° 18′E; Fig. 1) is a micro-tidal (~1 m) drowned river valley, 52 km in length, with a maximum depth of 30 m, and is located in southern Tasmania (Whitehead et al., 2010). The waterway has been extensively studied through monitoring and management programs and has been the focus for hydrodynamic modelling and previous studies of metal contamination (Margvelashvili et al., 2005; Jones et al., 2013a) Three regions were sampled: two of those regions (Middle Estuary (ME) and Ralphs Bay (RB)) were within the Derwent Estuary, whilst the third region, Mickey's Bay (MB), was located 48 km south of the estuary (Fig. 1.). The Middle Estuary (ME), the industrialised region, and Ralphs Bay (RB) in the Derwent Estuary are both well-mixed (dominated by wind-driven and tidal mixing) water bodies, but vary significantly in their sediment composition (Thomson and Godfrey, 1985; Margvelashvili et al., 2005). Mickeys Bay (MB), the reference region, is an embayment similar to Ralphs Bay and was included to provide comparative data from a region that has not been contaminated with either Hg or Se (Jones et al., 2013b).

2.2. Flathead collection

Fish ($n = 120$) were sampled by line fishing during November and December (2010 and 2011). Each fish was sealed in a plastic bag and stored on ice until transfer to the laboratory, where they were frozen (−40 °C). Fish were measured (fork length, FL) and then dissected. One fillet of muscle tissue, posterior to the pectoral fin, was removed from each fish and refrozen in acid-cleaned polypropylene tubes. Muscle samples were lyophilized to constant mass (±0.01 g) and homogenized, with a subsample of tissue from each region taken for separate THg, Se and MeHg analyses. Sagittal otoliths were extracted for age determination, following a validated method (Jordan et al., 1998), where resin-mounted, sectioned sagittal otoliths were read by two independent readers, with between- and within-reader precision examined by an index of average percent error (Beamish and Fournier, 1981).

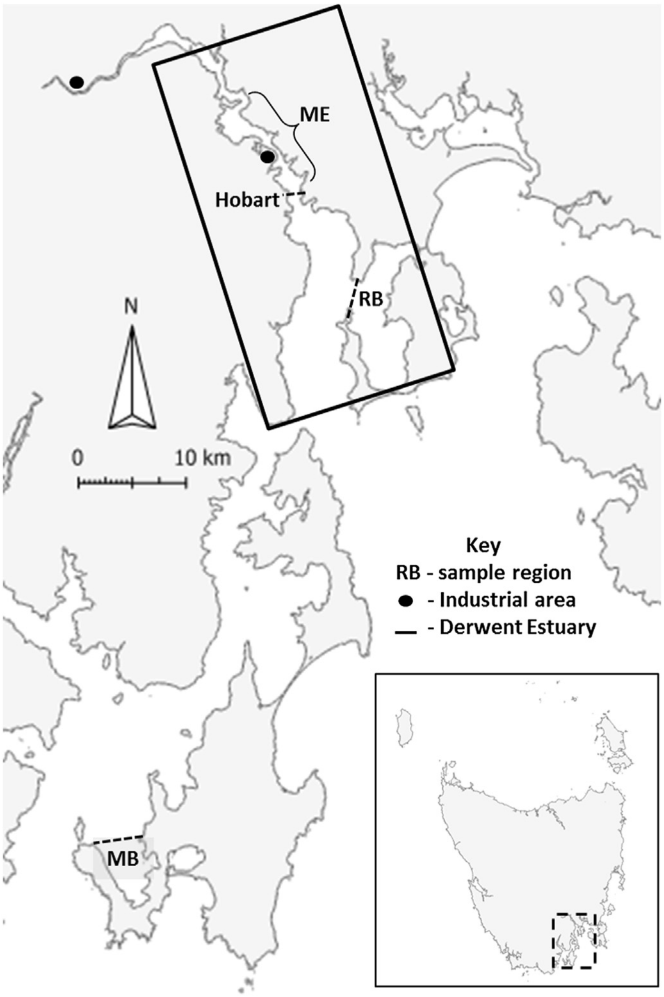

Fig. 1. Southern Tasmania and the Derwent Estuary, with locations of the two estuary regions Middle Estuary (ME), Ralphs Bay (RB) and the reference region Mickeys Bay (MB) 48 km south of the Derwent Estuary.

2.3. Sediment collection and analyses

Sediment sites ranged in depth from 7 to 15 m. Water measurements were taken 1 m above the surface of the sediment using a multi-parameter probe (6600 v2 Sonde, YSI, Australia). Sediment cores ($n = 39$) were collected using a purpose-built tri-corer consisting of three polycarbonate pipes (250 mm length × 45 mm internal diameter) that could be pushed into the sediment to a depth of 4 cm (±0.5 cm). The sediment collected was transferred to glass jars and frozen (−40 °C). The three samples collected by the tri-corer were pooled, freeze-dried and sieved through 500 μm mesh to remove large shell fragments. Samples were homogenized before subsamples were taken for analysis of iron (Fe), % Sulfur (%S), % Carbon (%C), % Nitrogen (%N), acid volatile sulfide (AVS), grain size and total organic carbon (TOC), as well as THg, MeHg and Se. Quantification of %S, %C and %N was achieved by elemental analysis (Thermo Finnigan EA 1112). AVS was determined by a rapid fluorescence method (Simpson, 2001), and grain size composition was measured by laser diffraction (ATA Scientific). Total organic carbon

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