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The mismatch between bioaccumulation in field and laboratory environments: Interpreting the differences for metals in benthic bivalves

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ABSTRACT

Laboratory-based bioaccumulation and toxicity bioassays are frequently used to predict the ecological risk of contaminated sediments in the field. This study investigates the bioassay conditions most relevant to achieving environmentally relevant field exposures. An identical series of metal-contaminated marine sediments were deployed in the field and laboratory over 31 days. Changes in metal concentrations and partitioning in both sediments and waters were used to interpret differences in metal exposure and bioaccumulation to the benthic bivalve *Tellina deltoidalis*. Loss of resuspended sediments and deposition of suspended particulate matter from the overlying water resulted in the concentrations of Cu, Pb and Zn (major contaminants) becoming lower in the 1-cm surface layer of field-deployed sediments. Lower exchange rates of overlying waters in the laboratory resulted in higher dissolved metal exposures. The prediction of metal bioaccumulation by the bivalves in field and laboratory was improved by considering the metal partitioning within the surface sediments.

1. Introduction

Laboratory-based bioassays are routinely used during the assessment of risk posed by contaminated sediments (ASTM, 2010; Casado-Martinez et al., 2006; Kennedy et al., 2009; Simpson and Spadaro, 2011). However, the conditions by which organisms are exposed to sediments in the laboratory can be quite different from those that occur from the same sediments in the field (Burton et al., 2005a,b; Liber et al., 2007; Hill et al., 2011). Static laboratory-based bioassays frequently result in much higher dissolved contaminant concentrations in overlying waters than occurs for the same sediment in the field. This is due to the higher sediment to overlying water ratios that occur in laboratory vessels in comparison to field sites, and continuous renewal of overlying water at field locations, further diluting contaminant fluxes from the sediments to the overlying water (Mann et al., 2010; Rosen et al., 2012). Organisms within field-based bioassays have to contend with greater variation in conditions than laboratory assays, including dissolved oxygen, light, temperature, turbidity and resuspension of sediments, tidal flows and competition from other organisms, including predation (Ringwood and Keppler, 2002; Anderson et al., 2004). Uncaged organisms in field locations may also reduce their exposure to contaminants by avoidance behaviours (Ward et al., 2013a,b). As such, the relationship between laboratory and field exposures has been the topic of ongoing debate.

Organism behaviour is influenced by the surrounding environment and, in particular, food availability and quality. Aquatic organisms obtain nutrition from both the dissolved and particulate phases of the environment, utilising both passive uptake and active ingestion (Rainbow, 2007). For benthic organisms, ingestion processes, establishing/maintaining habitats (e.g., burrowing) and irrigation of such habitats often cause sediment bioturbation, a process which alters sediment substrata by perturbing redox stratification, porewater equilibria and increases oxidation of metal-binding phases such as acid-volatile sulfide (AVS) (Riedel

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et al., 1997; Simpson and Batley, 2003; Atkinson et al., 2007; Simpson et al., 2012). These activities influence contaminant bioavailability from the perspective of both chemical (contaminant partitioning and speciation) and biological exposures (organisms burrowing and feeding behaviour) (Ciutat and Boudou, 2003; Atkinson et al., 2007; Simpson et al., 2012).

Here we compare the changes in the dissolved and particulate exposure and bioaccumulation of metals to the estuarine benthic deposit-feeding bivalve *Tellina deltoidalis* for the same series of metal-contaminated sediments in the laboratory and field over 31 days. The deployed sediments allowed the influence of contaminant concentrations and properties on the metal exposure and bioaccumulation to be assessed. Changes in metal concentrations and sediment properties influencing metal bioavailability (e.g. particle size, AVS, TOC) in the surface and deeper sediments, metal concentrations in overlying waters, and metal bioaccumulation and bivalve health using a Condition Index were used in the interpretation of the results.

2. Material and methods

2.1. General methods

All glass and plastic-ware for analyses were new and cleaned by soaking in 10% (v/v) HNO_3 (BDH, Analytical Reagent grade) for $\geq$ 24 h, followed by thorough rinsing with deionized water (Milli-Q, 18 MΩ cm). Test vessels used for bioassays were washed in a dishwasher (Gallay Scientific) sequentially with phosphate-free detergent, 1% HNO_3 and Milli-Q water. All chemicals were analytical reagent grade or equivalent analytical purity. Water pH, salinity, temperature and dissolved oxygen measurements were made with probes from WTW (Wissenschaftlich-Technische Werstätten) calibrated according to manufacturer instructions. Dissolved ammonia was analysed colorimetrically using a Merck Spectroquant Kit (14752, Merck). The fraction of fine sediment (< 63 μm) was determined by wet sieving through 63 μm nylon mesh followed by gravimetry, and total organic carbon (TOC) determined by high temperature TOC analyser (OC) after the removal of inorganic carbon with 1 M HCl until effervescence was complete.

Overlying water samples were membrane filtered (0.45 μm cellulose acetate, Sartorius Ministart) immediately after collection and acidified with concentrated HNO_3 (2% (v/v), Tracepur, Merck). Methods for analyses of total recoverable metals (TRM, by microwave assisted aqua regia) were performed as per Strom et al. (2011), and dilute acid-extractable metals (AEM, 1 M HCl) and acid-volatile sulfide (AVS) (all determined on sub-samples of the same homogenised sediment) as per Simpson (2001). Biota tissues (pool of seven individuals per sample) were freeze-dried before microwave-assisted (MARS Express) nitric acid extraction (HNO_3 at 200 °C for 30 min) as described in King et al. (2010). Dissolved metal concentrations in acid-digests of waters, sediment, and biological tissue samples were determined by a combination of inductively coupled plasma - optical emission spectrometry (ICP-OES, Varian 730-ES) and inductively coupled plasma-mass spectrometry (ICP-MS, Agilent 7500ce).

For quality assurance, filter and acid-digest blanks, duplicate analyses for 20% of all samples, sample spike recovery and certified reference material (CRMs) analyses were performed. Duplicates were within 20% and recoveries for spikes and CRMs, PACS-2 for sediment (National Research Council Canada, NRCC, Ottowa, ON, Canada) and DORM-3 for biota (*Mytilus galloprovincialis*, NRCC), were within 85–115% of certified values. The limits of reporting for the various methods were less than 10% of the lowest measured values.

2.2. Test media and organisms

Clean seawater was collected from Port Hacking, Sydney (New South Wales, Australia), membrane filtered (0.45 μm) and stored in the dark at 4 °C. Where necessary, the salinity of the filtered seawater was adjusted to the test salinity of 30 PSU using deionised water. Prior to use in experiments all waters were acclimated in a temperature-controlled room (21 ± 1 °C).

Control sediments were collected from an estuarine location in Bonnet Bay, Sydney, following the procedure previously described by Strom et al. (2011). This silty sediment (95% < 63 μm) had been previously characterised and found to have relatively low concentrations of metal and organic contaminants (Spadaro et al., 2008), and supported the survival and growth of the bivalve *T. deltoidalis* (King et al., 2010; Campana et al., 2013). The silty control sediment was used either unmodified (Sediment 1) or as a 30:70 mixture with clean sand (98% > 180 μm) (Sediment 5). Contaminated sediments were collected from Port Kembla (Sediment 4) and three sites in Sydney Harbour (Sediments 6, 7 and 8). Previous studies found these sediments contained concentrations of Cu, Pb and Zn which greatly exceeded the sediment quality guideline values, and caused toxicity to amphipod reproduction (Amato et al., 2014), but relatively low concentrations of organic contaminants (Chariton et al., 2010; Dafforn et al., 2012). All sediments were sieved (<1 mm, plastic mesh), homogenized and stored at 4 °C in the dark until use. A dilution series was created by mixing Sediment 4 (contaminated) with Sediment 1 (control) (both 95% < 63 μm) at ratios of 1:1 (Sediment 3) and 1:3 (Sediment 2). Mixing occurred in a nitrogen atmosphere and the sediments were equilibrated for four weeks before use (Simpson et al., 2004). Test sediments were grouped into two series (silty S1–S4, or sandy S5–S8) according to their physical properties.

The *T. deltoidalis* (shell lengths of 5–12 mm) were collected from Lane Cove River (NSW, Australia) and maintained as described previously (Atkinson et al., 2007; King et al., 2010; Campana et al., 2013). Bivalves were analysed for both metal content (soft tissues after 24-h depuration) and Condition Index (CI). Condition Index calculations were made on seven bivalves per sediment sample according to Freeman (1974), in which Condition Index = (dry soft body mass/dry shell mass) × 100. Further details about the sediments and bivalve are provided in the Supporting Information.

2.3. Field bioaccumulation bioassays

The field-deployed bioassays were performed in chambers (1-L Nalgene bottles with three 4.5 cm × 8 cm openings cut in the sides to allow water circulation) held within cages deployed in an uncontaminated section of the Woronora River estuary (Sydney, NSW, Australia) for 31 days. Dimensions and photographs of the chambers and cages are provided in the Supporting Information. The cages were suspended from a floating boat pontoon that allowed cages to maintain a submerged depth of 40 cm, irrespective of tidal cycles. There were two replicates of each sediment sample (sediments 1–8) and these were randomly distributed in the cages.

Water quality parameters were measured twice weekly in the field for pH (7.5–8), salinity (31–33 PSU), temperature (21–24 °C) and dissolved oxygen concentration (> 80% saturation). Suspended particulate matter (SPM) was collected using sediment traps (100 mL polycarbonate vials, 4 cm diameter × 9 cm high) secured within the inner section of each cage on day 1 and changed on days 5, 10 and 20 (16 vials in total). Dissolved metals were monitored using diffusive gradients in thin films (DGT). Piston covers were obtained from DGT Research Ltd (Lancaster, UK) and DGTs were prepared and assembled following standard procedures (Zhang and Davison, 1995; http://www.dgtresearch.com/) with Chelex