Environmental Pollution xxx (2015) 1–10

Contents lists available at ScienceDirect

Environmental Pollution

journal homepage: www.elsevier.com/locate/envpol

The impact of sediment bioturbation by secondary organisms on metal bioavailability, bioaccumulation and toxicity to target organisms in benthic bioassays: Implications for sediment quality assessment

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ARTICLE INFO

Article history:
Received 3 July 2015
Received in revised form
18 October 2015
Accepted 21 October 2015
Available online xxx

Keywords:
Bioassays
Benthic invertebrates
Contaminants
Speciation
Sediment quality guidelines

ABSTRACT

Bioturbation alters the properties of sediments and modifies contaminant bioavailability to benthic organisms. These naturally occurring disturbances are seldom considered during the assessment of sediment quality. We investigated how the presence (High bioturbation) and absence (Low bioturbation) of a strongly bioturbating amphipod within three different sediments influenced metal bioavailability, survival and bioaccumulation of metals to the bivalve *Tellina deltoidalis*. The concentrations of dissolved copper decreased and manganese increased with increased bioturbation. For copper a strong correlation was observed between increased bivalve survival (53–100%) and dissolved concentrations in the overlying water. Increased bioturbation intensity resulted in greater tissue concentrations for chromium and zinc in some test sediments. Overall, the results highlight the strong influence that the natural bioturbation activities from one organism may have on the risk contaminants pose to other organisms within the local environment. The characterisation of field-based exposure conditions concerning the biotic or abiotic resuspension of sediments and the rate of attenuation of released contaminants through dilution or readsorption may enable laboratory-based bioassay designs to be adapted to better match those of the assessed environment.

1. Introduction

The degree to which organisms modify sediments is influenced by sediment properties, biogenic activity (burrowing, feeding behaviour etc), and the frequency of sediment reworking. This is clearly shown by studies with amphipods (*Corophium volutator*), polychaete worms (*Lumbriculus variegatus*, *Arenicola marina*, *Nereis diversicolor*, *Nereis virens*, *Heteromastus filiformis* and *Tubifex tubifex*), bivalves (*Tellina texana*, *Macoma balthica* and *Cyclope neritea*), and sea cucumbers (*Holothuria whitmaei*) (Aller and Yingst, 1985; De Backer et al., 2011; Lagauzère et al., 2009; Peterson et al., 1996; Pischedda et al., 2008; Shiell and Knott, 2010; Volkenborn et al., 2010). Community structure and population density are further factors which influence the extent of sediment disturbance by organisms (Forbes, 1994; Thrush et al., 2006; Wethey et al., 2001).

Metal(loid) bioavailability and the rate of metal accumulation by benthic organisms is influenced by biokinetics, organism behaviour and physiology, and sediment chemistry, particularly factors that impact the partitioning between aqueous and solid phases and metal speciation in these phases (Fe/Mn (oxy)hydroxides and DOC) (Costello et al., 2015; Fisher et al., 1980; Simpson and Batley, 2007). The bioturbation of sediments by large benthic invertebrates alters sediment redox chemistry by mixing pre-stratified zones in the sediment, and increasing the penetration of electron acceptors such as dissolved O_2, NO_3^- and SO_4^{2-} into anoxic sediments (Aller et al., 2001; Granéli, 1979; Matisoff et al., 1985; Pischedda et al., 2008; Volkenborn et al., 2010). Redox changes can alter metal binding affinities between the solid and dissolved phases, significantly modifying the speciation and bioavailability of most metals in sediments (De Jonge et al., 2012; Doyle and Otte, 1997; Granberg

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http://dx.doi.org/10.1016/j.envpol.2015.10.033
0269-7491/

et al., 2008). The concentration of AVS has a major influence on metal bioavailability, and for sediments containing a molar excess of acid volatile sulfide (AVS) over simultaneously extractable metals (SEM, ΣCd, Cu, Ni, Pb, Zn), it is predicted that the porewater concentrations of these metals will be negligible and should not cause direct toxicity to benthic organisms (Ankley et al., 1996; Hansen et al., 2005; Lawrence et al., 1982). In addition, the activity of microbes such as *Desulfuromonadales* and *Geobacter sulfurreducens* in abandoned burrows also contributes to the release of metals from anoxic sediments to the pore waters and overlying water column (Kristensen, 2008; Meysman et al., 2006). Thus bioturbation processes can modify the exposure and risk posed by contaminants to the organisms and surrounding ecosystem (Atkinson et al., 2007; Ciutat and Boudou, 2003; Simpson et al., 2002).

Within sediment quality assessments programs, laboratory-based bioassays are frequently used to assess bioaccumulation and toxicity (ASTM, 2014, 2010). To prevent predation by indigenous organisms on the test species, methods generally specify the removal of large indigenous organisms before testing. While the exposure conditions in laboratory bioassays need not exactly resemble field conditions, the bioassays should aim to provide assessment outcomes that would be similar to those of sediments that remained in their natural field setting. In this study we hypothesise that the bioturbation activities of indigenous organisms may be sufficient to influence the outcomes of bioaccumulation and toxicity assessments, i.e. whether effects are detected, and potentially alter the outcome of assessment programs.

The specific objectives of this work were therefore to: 1) investigate how different bioturbation intensities alter the bioavailability, bioaccumulation and toxicity of metals, and 2) evaluate the influence that bioturbation by secondary organisms may have on the outcomes of sediment bioaccumulation and toxicity bioassays. The three different sediments were subjected to the presence of no organisms, a benthic bivalve (*Tellina deltoidalis*; low bioturbation), and this bivalve combined with a highly bioturbating amphipod (*Victoriopisa australiensis*; high bioturbation) and the metal bioaccumulation and toxicity (survival) to the bivalve was assessed. Changes in metal bioavailability were assessed through measurements of AVS, dissolved metal release to overlying water and bioaccumulation by the bivalve. Differences in metal bioaccumulation for the Low and High bioturbation treatments are discussed in relation to the observed differences in metal bioavailability and how assessment outcomes might be modified by varying degrees of bioturbation.

2. Material and methods

2.1. General methods

All chemicals used were AR grade or equivalent analytical purity. Deionised water (18 MΩ cm, Millipore) was used for all solutions. Glass and plastic consumables used for analysis were new and acid-washed via soaking in 10% (v/v) HNO_3 (>24 h, BDH, AR Grade), followed by thorough rinsing with deionised water. Glass beakers used for bioassays were washed in a dishwasher (Gallay Scientific) with detergent followed by acid-washing (5% HNO_3 (v/v)) and rinsing using reverse-osmosis purified water.

Physicochemical parameters of dissolved oxygen (DO), temperature, salinity and pH were routinely measured using WTW instruments (Wissenschaftlich-Technische Werstätten) calibrated according to manufacturer's instructions: an Oxi 330 Oximeter, LF320 Conductivity meter, and a pH 320 m. Dissolved ammonia was measured using a rapid test kit (API Fish Care, LR8600). Overlying water samples were immediately filtered (0.45 μm cellulose nitrate, 25 mm, Minisart, Sartorius) and acidified to 2% HNO_3 (v/v, Tracepur, Merck). Total suspended solids (TSS) were determined gravimetrically; overlying water samples (350 mL) were filtered (cellulose nitrate ester filters, 0.45 μm, *in vacuo* Sartorius polycarbonate filter rig), dried (70 °C, 48 h), and weighed.

Subsamples of homogenised bulk sediment were collected prior to sediment transfer into test vessels, and at the end of the study as mini-cores (1 cm diameter, 4 cm depth; 10 mL, polycarbonate vials) from each test vessel. The subsamples and mini-cores were immediately frozen (−20 °C) until analysis. The gravimetric determination of the dry:wet sediment ratio (D:W) and fine sediment fraction (<63 μm) was conducted as per Belzunce-Segarra et al. (2015). Total recoverable metals (TRM) were determined after low-pressure microwave-assisted (MARS 5, CEM) aqua regia digestion (3:1 HNO_3:HCl). Total organic carbon (TOC) analysis was conducted using a CO_2 evolution method. Dried and crushed samples were acid-treated to remove inorganic carbonates followed by high temperature combustion (LECO furnace) in the presence of strong oxidants/catalysts using infrared detection. Within a nitrogen filled glove box, the frozen sediment mini-cores were extruded and sections taken from the surface (~1.5 cm from the top) and at depth (~1.5 cm from the bottom) for analyses of AVS and AEM (Simpson, 2001).

Biological tissues (~0.1 g) were freeze-dried (24 h, Christ Freeze Drier), digested in a HNO_3:H_2O_2 digestion solution (2 mL concentrated HNO_3 (Tracepur, Merck): 1 mL concentrated H_2O_2 (Merck) for 18 h at 25 °C) and microwave-heated for 1 h (MARS 5, CEM, programmed RT − 60 °C, 12 min; 60–65 °C, 10 min; 65–70 °C, 10 min; 70 °C for 10 min) before a 10-fold dilution using deionised water for metal analysis.

Metal analyses in waters and acid digests were performed on a combination of inductively coupled plasma − atomic emission spectrometry (ICP-AES, Varian 730-ES) and inductively coupled plasma-mass spectrometry (ICP-MS, Agilent 7500ce). For the purpose of QA/QC, 10% of all samples analysed were blanks and 30% were duplicates. Certified reference material recoveries for both sediment (ERM®-CC018, European Reference Materials (ERM)) and biological tissues (DORM-3, *Mytilus galloprovincialis*, NRCC), were analysed with the respective samples and were within 75–125% of expected 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 sourced from the southeast coast of New South Wales (NSW), Australia, was filtered (1 μm) and analysed (ICP-AES) before use to ensure that metals of interest were below 1 μg L^{-1}. Clean and contaminated sediments (0–15 cm depth) were collected from Lake Illawarra, Port Kembla, Bonnet Bay and Kings Bay, NSW. Sediments were sieved on-site (2 mm plastic mesh) to remove coarse material (e.g. detritus and leaves) and any large fauna, then thoroughly homogenised and stored in polyethylene bags at ~4 °C in the dark. Lake Illawarra (S1) and Kings Bay (S3) sediments were used unmodified, whereas sediment from Port Kembla was mixed 1:1 with Bonnet Bay sediment to achieve the desired contaminant concentrations (S2) as discussed by Belzunce-Segarra et al. (2015).

The deposit-feeding amphipods *V. australiensis* (Chilton, 1923) (2–3 cm body length) were collected from Lake Illawarra (34°3′S, 150°49′E), a large coastal estuarine system. *V. australiensis* inhabits estuarine, littoral, mud flats and seagrass sediments of south-eastern Australia (Dunn et al., 2009; Lowry and Springthorpe, 2005). The amphipod inhabits fixed burrows, and feeds on subsurface sediments as they excavate and redeposit the detritus back into their burrows. The deposit-feeding bivalve *T. deltoidalis*

Please cite this article in press as: Remaili, T.M., et al., The impact of sediment bioturbation by secondary organisms on metal bioavailability, bioaccumulation and toxicity to target organisms in benthic bioassays: Implications for sediment quality assessment, Environmental Pollution (2015), http://dx.doi.org/10.1016/j.envpol.2015.10.033