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Temporal trends and bioavailability assessment of heavy metals in the sediments of Deception Bay, Queensland, Australia



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ABSTRACT

Thirteen sites in Deception Bay, Queensland, Australia were sampled three times over a period of 7 months and assessed for contamination by a range of heavy metals, primarily As, Cd, Cr, Cu, Pb and Hg. Fraction analysis, enrichment factors and Principal Components Analysis–Absolute Principal Component Scores (PCA–APCS) analysis were conducted in order to identify the potential bioavailability of these elements of concern and their sources. Hg and Te were identified as the elements of highest enrichment in Deception Bay while marine sediments, shipping and antifouling agents were identified as the sources of the Weak Acid Extractable Metals (WE-M), with antifouling agents showing long residence time for mercury contamination. This has significant implications for the future of monitoring and regulation of heavy metal contamination within Deception Bay.

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Contamination of marine sediments by heavy metals (in particular, toxic metals) is a growing concern due to their toxic effects on local fauna and flora (Che et al., 2003; Liu et al., 2003). Marine sediments act as a sink for heavy metals, concentrating them and acting as a filter through precipitation and sequestration (Grecco et al., 2011; Nicholson et al., 2003; Satpathy et al., 2012). In addition to acting as heavy metal sinks, a number of simultaneous processes, including re-suspension and bioturbation, occur in marine and estuarine environments which can make them available for uptake into the biosphere (Birch and Taylor, 1999; González-Fernández et al., 2011).

Deception Bay (27°8′S, 153°6′E) is the northernmost embayment within Moreton Bay (27°15′S, 153°15′E), Queensland and is bordered to the north by Bribie Island, to the south by the Redcliffe Peninsular and to the west by Deception Bay. The area receives water inputs from the Caboolture River and Pumicestone Passage, with the Caboolture River being the major terrestrial sediment input (Dennison and Abal, 1999). Mineral sands flowing up the coast of Queensland flow into Deception Bay through the Northern Passage in Moreton Bay, acting as a source of elements such as Th and U (Arogunjo et al., 2009).

There are significant possible metal sources at Deception Bay, including two shipyards, an anchorage within the mouth of the Caboolture River, as well as a Marina with an associated fishing fleet within Pumicestone Passage. The Caboolture region which drains into the Caboolture River and into Deception Bay has undergone exponential growth since the 1980s, with an influx of population resulting in increase in vehicular traffic and light industry.

Elements which are more likely to be bioavailable should be the focus of sediment risk assessment. However, it is more common in research to identify Total Metals (T-M), Total Recoverable Metals (TR-M) or to do a complete fraction analysis using the Tessier or BCR (Community Bureau of Reference) methods (Baptista Neto et al., 2000; Cox and Preda, 2005; Jones and Turki, 1997; Martin et al., 1987; Preda and Cox, 2001, 2002). Total Metals and Total Recoverable Metals methods tend to rely on harsh reagents such as hydrofluoric acid (HF) or Aqua Regia (AR) digestions which not only extract the most bioavailable metals but also attack the more inert minerals in the sediment (such as quartz), which releases metals that are not normally bioavailable, resulting in over-reporting of risk (Ahdy and Youssef, 2011; Lee et al., 2012; Shikazono et al., 2012; Shilla and Dativa, 2011). The use of sequential methods, such as the Tessier protocol (Tessier et al., 1979) or the BCR protocol (Cuong and Obbard, 2006) have identified chemistry problems, most notably re-precipitation of metals (Whalley and Grant, 1994).

Weak Acid Extractable Metals (WE-M) has witnessed some use in the literature. However, the type of reagent is important, as some metals (most notably Pb and Hg) are not soluble as chlorides (from HCl). The use of nitric acid (HNO₃) may lead to oxidation of metals which are not otherwise available (Agemian and Chau, 1976; Sutherland, 2002).



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The use of chelating acids (such as EDTA) has been reported, but these methods have not seen widespread application because dilute HCl has been found to be more efficient (Malo, 1977). In some cases, the use of dilute HCl has even been more efficient than the use of sequential extraction methodologies (Sutherland, 2002) in assessing the weakly bound elements.

Historically, there has been little research into heavy metal contamination in Moreton Bay, with the latest work being an analysis of intertidal sediment cores, which found that there was some enrichment of Pb, Zn, Cd and Ni in intertidal regions. This finding was linked to settlement of the area (Morelli et al., 2012). The only other significant investigation on sediment contamination in Moreton Bay was undertaken in 1979 (Wallace and Moss, 1979), which examined Pb and Hg, but found little contamination. Both of these studies focussed on a limited number of sampling sites and provided limited information about the spatial variation and environmental risk of heavy metal contamination in the Bay.

Sampling sites were selected to achieve a systematic coverage of the sampling area. The depth of the sampling sites had to be considered, as the depth of Deception Bay varies from less than 0.5 m at high tide around sites DB5 and DB6 to more than 10 m at other sites, which had significant impact on the efficiency of the sampling dredge.

Sediment samples were taken from the sampling area (Fig. 1) using a Van Veem 7.5 kg sample dredge, before being placed in cleaned plastic sampling bags and stored on ice. Once the samples were in the laboratory, they were freeze-dried and sieved to less than 100 μ m to remove coarse materials such as pebbles and pieces of shell prior to digestion. Generally, the samples were fine-grained, with the only debris left after sieving being pieces of shell or larger rocks, which made up less than one percent of the total materials.

A background sample was taken from the upper reaches of the Caboolture River near Rocksberg at 27°6′30″S, 152°50′58″E; a second background site was taken from the Bongaree Jetty (27°5′2″S, 153°9′28″E) to account for sand coming from Pumicestone Passage and a third background sample was taken from Woorim Beach. (27°5′2″S, 153°12′26″E) to account for sand entering Deception Bay from the north. These samples were treated in the same way as the sediment samples. Previous work on the background sites identified Caboolture River as the largest contributor to sediment deposition in Deception Bay (Brady et al., 2014; Hancock, 2001a,b; Milford and Church, 1977) and this resulted in the use of the Caboolture River background site rather than the other two background sites, which had much smaller sediment contributions to the area.

Weak-acid Recoverable metals were digested using a method similar to that has been well reported in the literature (Agemian and Chau, 1976; Hu et al., 2011; Sutherland, 2002) by placing 0.05 g of sample into a pre-cleaned 50 cm³ centrifuge tube, and 50 cm³ of 1 M nitric acid (twice distilled) was added. The samples were then tumbled end-over end overnight (for a minimum period of 6 h) to ensure complete extraction of the weak-acid soluble fraction. Samples were then centrifuged at 3500 RPM for 5 min and analysed using an Agilent 8800 ICP-MS/MS.

Total Recoverable Metals (TR-M, which recover elements that are not bound within the quartz structure) digestions were completed by placing 0.05 g sample into a Teflon sample tube, adding 3 cm³ twice distilled HNO₃ and 1 cm³ twice distilled HCl and 0.1 cm³ 1000 mg/L Au standard (as a preservative for Hg). The samples were then digested using a Milestone Ultrawave microwave digester by ramping to 260 °C in 20 min and holding for 40 min at 260 °C. The samples were then placed into a 50 cm³ centrifuge tube, diluted to 50 cm³ using ultrapure water (18.2 M Ω resistivity) and centrifuged at 3000 RPM for 5 min before analysis using an Agilent 8800 series ICP-MS/MS. This collected data is available in the Supplementary Information provided.

In order to ensure the integrity of the sampling program, field and trip blanks were used, along with random duplicate samples (which were selected before each sampling run) being collected and analysed during each sampling cycle. Reagent and water blanks were analysed according to NATA (2012) and US EPA (2001) recommendations. In addition, the CRM (Certified Reference Material) MESS-3 (NRC, Canada) was analysed for the elements of interest and was treated to the same digestion procedures as the samples, with a ratio of 1 CRM to every 20 samples for the



Fig. 1. Deception Bay sampling site locations, with anchorages marked by arrows.

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