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Assessment of human-induced change and biological risk posed by contaminants in estuarine/harbour sediments: Sydney Harbour/estuary (Australia)

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A R T I C L E I N F O

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ABSTRACT

A rapid, simple yet scientifically sound scheme providing two important types of information used in assessment of estuarine sediments is presented. The mean enrichment quotient (MEQ) (fine contemporary sediment metal concentration/fine fraction background metal concentration) for Cu, Pb and Zn provides the magnitude of human-induced change, (deviation from the pristine condition), while sediment quality guidelines (SQGs) assess the risk posed by sedimentary contaminants to the benthic community.

Maximum metal enrichment for sediment in Sydney estuary (Australia) is >100 times for Cu, Pb and Zn and the MEQ is >10 times for most of the estuary. Adverse effect on benthic populations due to Cu, Pb and Zn are expected in 2%, 50% and 36% of the waterway, respectively. SQGs for contaminant mixtures predict $\sim 2\%$ of the estuary has the highest risk of adverse effects, while 25% has intermediate risk. The scheme is well suited to initial assessments of estuarine sediments worldwide.

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1. Introduction

Estuaries are diverse and biologically productive ecosystems with high conservation and ecological value (Costanza et al., 1997, 2014; Boyes and Elliott, 2006). More than half the world population live within 100 km of a shoreline (Niemi et al., 2004) and estuaries are commonly the foci of industrial, commercial and recreational activities (Birch et al., 2015a, 2015b). Substantial anthropogenic stress has resulted in reduction in sediment and water quality, which threatens benthic and pelagic populations (Chapman and Wang, 2001). It is therefore important that the extent of contamination of estuarine environments is assessed accurately and that the level of threat to the health to these sensitive environments be determined comprehensively.

1.1. Environmental indicators

Choosing an appropriate ecosystem indicator to assess environmental condition is complex and requires an integrated strategy to quantify the effects of human activities on the marine environment (Magni, 2003; Magni et al., 2004). Ecosystem condition indicators need to be easily and inexpensively employed to accurately and appropriately measure numerous biological, chemical and physical processes. A weight-of-evidence approach is considered most effective for assessment of multiple indicators within a decision framework in assessing ecosystem health (Simpson et al., 2005).

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http://dx.doi.org/10.1016/j.marpolbul.2017.01.013 0025-326X/© 2017 Elsevier Ltd. All rights reserved. Ecosystem indicators used to assess anthropogenic stress are commonly compromised by significant natural spatial and temporal variability. This confounding results in inappropriate and erroneous ecosystem assessment rendering it difficult to discriminate between natural- and human-induced stress under complex conditions (Hogg and Norris, 1991; Olmos and Birch, 2010). The original (pristine) condition of an estuary is rarely known and to determine whether present distributions of flora and fauna indicate good or poor condition is difficult to establish (McLoughlin, 1985). To determine the magnitude of human-induced change and adverse biological effects requires historic baseline information (control sites), which is commonly not available, especially on high-population seaboards.

1.2. Sediments as ecosystem indicators

The use of sedimentary indicators in assessment of environmental condition has been poorly understood and greatly undervalued and underutilised. The preferred media for assessment of estuarine health has traditionally been the water column and biological indicators (Rainbow, 1995, 2006). However, water is dynamic and highly variable in the short- and long-term requiring large numbers of samples to spatially and temporally characterise this medium with confidence (Birch and Olmos, 2008). Water chemical concentrations are low and analyses are difficult and expensive, which compromises data quality and interpretation (Bubb et al., 1990; Siaka et al., 1998; Birch and Taylor, 2000b). Analysis of floral and faunal distributions is often difficult due to significant natural temporal and spatial variance and the chemistry of tissue is variable between individuals, species, genders, organs and age.

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Increasingly, sediment is being used preferentially to assess the status of aquatic environments rather than other traditional media. However, these materials should be used in combination with other screening tools for a comprehensive assessment of aquatic systems (Belin et al., 2014). The great advantage sediments have over other indicators of environmental condition is that they faithfully record and time integrate environmental events within the aquatic system, commonly referred to as 'the memory of sediments'. The integration of environmental events over time provides useful spatial and temporal information (Birch, 2007; Birch et al., 2013) and allows prediction of future environmental change and status (Birch et al., 2010, 2012, 2013). Sediments greatly affect the quality of overlying and interstitial water through physical (re-suspension, Peterson et al., 1997; Simpson et al., 2000), biological (bioturbation, Reible et al., 1996) and chemical (desorption and benthic diffusion, Rivera-Duarte and Flegal, 1994.) processes. Sediments are an extensive habitat and a large storage for contaminants and therefore have an extensive influence on the biological health in the marine environment. The majority of toxicants adsorb to fine-grained particles and therefore sediments are important in the transport of contaminants in the water column. Contaminants also have an affinity for finely-disseminated organic matter and therefore sediments influence the feeding habitats of benthic and pelagic animals, as well as being involved in the uptake processes. Sediment quality thus influences, to a large degree, the biodiversity and ecological health of marine systems. Although sediments are appropriate indicators of environmental status, these materials should be used in combination with other media for a comprehensive of aquatic systems. However, as yet no approach has been undertaken to develop indicators to assess potential interactions between the wide range of metallic and non-metallic contaminants, which may be contained within marine sediments. The vast number of combinations of these chemicals may result in synergistic, additive or antagonistic functional effects on benthic and pelagic communities, which remains to be addressed (Part, 2006).

Although sediments are appropriate indicators of environmental status in aquatic systems, their use has sometimes been problematic due to the lack of uniformity in analysis and difficulties in interpretation of sedimentary data. These issues have been largely overcome and interpretation has become clearer with establishment of accepted analytical protocols (Simpson et al., 2005, 2016) and a more advanced understanding of speciation and particularly partitioning in sedimentary systems. The difficulty in interpreting sediment-derived contaminant data due to the confounding effects of variable grain size is not always fully appreciated. Grain size is the dominant parameter controlling contaminant concentrations in sediments, including proximity to source. A procedure for reducing the confounding caused by variable grain size is essential for interpretation of sediment contaminant data and should be an integral part of the protocol for environmental assessment. Variability in grain size imposes considerable spatial and temporal variance on sediment-derived chemical data and determination of source, dispersion, the pristine condition, magnitude of anthropogenic change (as addressed in the current work), temporal change and comparability of contaminated systems cannot be made without some form of 'normalisation' of the data (see Section 2.1.3) (Forstner, 1982; Loring, 1991; Birch, 2003).

1.3. Assessing the status of estuarine condition

The condition of an estuarine environment may be described using a wide range of approaches, however from a management perspective it is important to know (1) by how much the system has deviated from the pristine condition (magnitude of anthropogenic change), and (2) the degree of risk of potential harm posed by sedimentary contaminants to biological communities. These two types of data are very different – one measures the amount of human-induced change and the other determines potential ecological risk posed by sedimentary anthropogenic

chemicals. These two aspects are frequently confused and these data types are often integrated (Caeiro et al., 2005; Mil-Homes et al., 2006).

The objectives of the present work are to briefly review methodologies used in the assessment of anthropogenic change and risk posed by sedimentary contaminants in marine/estuarine environments and then to apply the least complex and most effective approach to sediments of Sydney harbour/estuary, Australia.

2. Methods to assess the magnitude of anthropogenic change

2.1. Pre-anthropogenic, or background concentrations

To calculate the amount of deviation from the pristine condition in marine environments, it is first necessary to estimate the concentrations of chemicals in sediments deposited prior to human influence (pre-anthropogenic, pre-industrial or background concentrations). For anthropogenic chemicals, e.g. organochlorine compounds (OCs) and polychlorinated biphenyls (PCBs), pre-anthropogenic concentrations are zero. However, for chemicals, which have both anthropogenic and natural components, such as metals and polycyclic aromatic hydrocarbons (PAHs), it is necessary that the human contribution first be determined to establish the magnitude of change. The background concentration of PAH is mainly related to bush fire activity and is low (<50 µg/kg) and frequently regarded as zero. Metals are an intrinsic component of minerals comprising sediments and have a natural background concentration even in pristine environments. Pre-anthropogenic concentrations of metals have been determined by statistical and empirical methodologies (Matschullat et al., 2000).

2.1.1. Statistical methods to estimate background metal concentrations

Statistical methods used to determine anthropogenic change identify pristine from contaminated samples to establish a threshold, which separates natural from man-modified material. Statistical methods to determine anthropogenic contributions have been reviewed by Matschullat et al. (2000), Reimann et al. (2005) and Rodriguez et al. (2006).

Pre-anthropogenic metal concentrations are inferred using linear regression of a normalising element (Al, Fe) against total sediment metal concentration. Outliers are identified as those samples that fall outside the 95% confidence limit and are removed and regression recalculated until normality is achieved. Data falling within the predicted interval belong to a pre-anthropogenic population and samples above this band are enriched in metals due to human activities (Matschullat et al., 2000). Modal analyses, relative cumulative frequency curves, iterative standard deviation identification and outlier testing have been used in many studies to estimate background metal concentrations (Matschullat et al., 2000). More recently empirical cumulative distribution functions (ECDF) have been used to detect and remove outliers in large sedimentary metal data sets (Rodriguez et al., 2006). The mean of discrete uni-modal metal populations and the mean of the lowest sub-group for polymodal distributions were inferred as the pre-anthropogenic metal concentrations (Rodriguez et al., 2006).

Sediment chemistry data, however are commonly not normally or log-normally distributed due to the presence of multiple processes controlling elemental distribution and may comprise numerous subgroups (multimodal) (Reimann and Filzmoser, 2000). Also, all sediment samples in some harbour estuaries are contaminated to some degree and two distinct sub-populations (contaminated and pristine) are not present (Birch and Taylor, 2002a).

2.1.2. Empirical methods to estimate background metal concentrations

Empirical techniques use geochemical data from uncontaminated sediment derived from subsurface sediment below the anthropogenic section by coring (Thevenon et al., 2011), or from pristine surficial sediment in control areas of the same ecosystem. Use of pristine estuarine sediment is often not possible due to the global nature of atmospheric

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