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Abstract

The distribution of trace metals (spatial and temporal) and sedimentary fractions were investigated to identify the concentrations and sources of trace metals within Kogarah Bay, NSW, Australia. A total of 59 surface sediments and six subsurface samples from core of the sediment were collected. The contamination factor and pollution load index indices used to evaluate environmental effects of trace metals. The study area was found to be uncontaminated with Cr and Ni, moderately contaminated with As and considerably contaminated with Cu, Zn and Pb. The concentrations of Cr and Ni were below both effect range low and effect range median, while As, Cu, Zn and Pb were slightly above effect range low. The highest concentrations of these trace metals such as Cu, Zn and Pb were found in the north, northwest and southeast of the bay, close to discharge points, stormwater outlets and around boatyards and watercrafts. The spatial distributions of metals were strongly related to muddy particles and organic matter. The temporal sediments of metals declined with increased sediment depth, which reflects accumulation of trace metals since European settlement in this area. Furthermore, the source of the trace metals was found to be stormwater outlets, gasoline fumes, boatyards and other human activities.

Disciplines

Medicine and Health Sciences | Social and Behavioral Sciences

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Source identification and assessment of sediment contamination of trace metals in Kogarah Bay, NSW, Australia

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Abstract

The distribution of trace metals (spatial and temporal) and sedimentary fractions were investigated to identify the concentrations and sources of trace metals within Kogarah Bay, NSW, Australia. A total of 59 surface sediments and six subsurface samples from core of the sediment were collected. The contamination factor and pollution load index indices used to evaluate environmental effects of trace metals. The study area was found to be uncontaminated with Cr and Ni, moderately contaminated with As and considerably contaminated with Cu, Zn and Pb. The concentrations of Cr and Ni were below both Effect Range Low and Effect Range Median, while As, Cu, Zn and Pb were slightly above Effect Range Low. The highest concentrations of these trace metals such as Cu, Zn and Pb were found in the north, north west and south east of the bay, close to discharge points, stormwater outlets and around boatyards and watercrafts. The spatial distributions of metals were strongly related to muddy particles and organic matter. The temporal sediments of metals declined with increased sediment depth, which reflects accumulation of trace metals since European settlement in this area. Furthermore, the source of the trace metals was found to be stormwater outlets, gasoline fumes, boatyards and other human activities.

Keyword Kogarah Bay. Sediments. Trace metals. Contaminations. Statistical analysis

Introduction

Marine and estuarine ecosystem sediments have been found to serve as a sink for trace metals (El-Said and Youssef 2013; Xu et al. 2014). They also play a major role as a source of trace metals in aquatic organisms when there are changes in environmental conditions such as pH, sediment redox potential, bioturbation and organic degradation (Qiu et al. 2011). As a result of such changes, trace metals can be released from sediments as free ions, and/or complex compounds into the water column (Zoumis et al. 2001). Trace metals are considered to be very toxic, bio-accumulative, and resistant to bio-chemical degradation.

Trace metals are harmful for microorganisms which can take up trace metals from ambient environments (Ruiz et al. 2006). These metals enter estuaries and coastal waters from various sources from discharge points such as residential wastewater, drainage dust deposition, industrial sources, stormwater and agricultural runoff (Zhao et al. 2012; Yang et al. 2012). Trace metals tend to be adsorbed, absorbed and accumulated in the fine to very fine sediment particle fraction, which eventually transfers to depositional areas (Mendiguchia et al. 2006). Moreover, the discharge of untreated urban wastewater can lead to the accretion of organic matter that may contain high levels of total phosphorus, nitrogen and carbon in the sediments, resulting in the reduction of dissolved oxygen in water (Ruiz et al. 2006; Zhao et al. 2014).

The main purpose of this paper is to provide a better understanding of the geochemical status, toxicological spatial and temporal distribution of trace metals in Kogarah Bay, as well as to determine the relationship between the spatial pattern of trace metals and distribution of sedimentary fractions. The study also evaluates the environmental effect in order to protect the ecosystem from accumulated pollution.

Materials and methods

Study area

Kogarah Bay, located 15 km south of Sydney (Fig.1a), is a tributary of the lower Georges River estuary. It has a length of about 2.3 km and a width between 500 m and 700 m (McLean et al. 2002). Overall, the bay has shallow water depths (<3 m, and the foreshores have been broadly modified (McLean et al. 2002). Kogarah Bay provides a significant environmental shelter and habitat for flora and fauna, breeding and nursery habitats for several species, and is of economic and social value (such as tourism, business enterprises, fisheries and aquaculture). The catchment areas of Kogarah Bay are urbanised with a population of approximately 60,400 in the local council area (Kogarah Council 2014). The sources of contamination are mainly emerging from catchment area and from recreational activities along the coast line such as watercrafts, boatyards and fishing which can in turn affect the water quality of the bay. Kogarah Bay is also affected by activities in the Georges River catchment area (800 km²; Fig.1b).

Fig. 1 a Surface and subsurface sediment samples in Kogarah Bay and b Botany Bay district, NSW, Australia

Sample collections and preparations

During the summer of 2012, a total of 59 surface sediment samples were collected by boat (Fig.1). Using a grab sampler, about 5 cm of surface sediment and six subsurface sediment samples from core of the sediment (40cm) were extracted by using push core in the highest concentrations of trace metals. The sediments samples were reserved for analysis. At each site the water depth and location were recorded using sonar and Geographical Position System (GPS) respectively. Grain size measurements were determined for all sediment samples using a Malvern Mastersizer 2000. This analysis was used to obtain the details of the source of sediments, distribution of grain size (sand, silt and clay), and to interpret geochemical results. Trace metals were measured by X-ray fluorescence spectrometer with a SPECTRO - analytical instrument (XEPOS) following (Norrish and Chappell 1977) procedure. Hierarchical cluster analysis (Zhang et al. 2013) was applied to distinguish characteristics of the samples. This statistical analysis was achieved using JMP software to include all variables.

Arc GIS desktop software was used to plot the sample sites within the study areas, and to provide advanced geostatistical analysis to create maps. The Kriging method of geostatistical analysis was used. This is a moderately quick interpolator that can be exacted or smoothed depending on the measurement error model. The method provides flexibility to evaluate graphs of spatial autocorrelation (Li and Heap 2008). Three mean parameters, the nugget (C_0), the sill ($C + C_0$) and the range, emerged from the models used. This identified the spatial structure of the variables at the given scale (Li and Heap 2008; Liu et al. 2013). The spatial heterogeneity of regional variables is characterized by the ratio of $C_0/(C + C_0)$ and provides spatial correlation among regional variables. Overall, a ratio less than 25% shows strong spatial correlation, a ratio ranging between 25% to 75% indicates moderate correlation, and a ratio of more than 75% indicates weak spatial correlation (Spijker et al. 2005). In the present research, the geostatistical approach uses a semivariogram to quantify spatial autocorrelation, and to provide conditions for the optimal spatial interpolation. The results indicated a ratio of 20% and thus demonstrating a strong spatial correlation. Kriging interpolation can be rigorously evaluated according to several cross-validation indicators as follows: the absolute value of the average mean standardized error (MSE) should be close to 0, there should be a minimum root mean square prediction error (RMSE), which should be close to the mean standard error (AME), and the standard root mean square (RMSS) should be close to 1 (Chen et al., 2013). These parameters are calculated using the following equations:

$$ME = \frac{1}{n} \sum_{i=1}^n (P_i + M_i) \dots\dots\dots 1$$

$$RMSE = \sqrt{\frac{1}{n} \sum_{i=1}^n (P_i - M_i)^2} \dots\dots\dots 2$$

$$MSE = \frac{1}{n} \sum_{i=1}^n [(P_i - M_i) / \sigma_i] \dots\dots\dots 3$$

where: n is the number of the sites; P_i and M_i are the predicted and measured values at sites i respectively, and σ_i is the Kriging standard error at sites i.

The choice of background values plays an important role in interpreting geochemical data. Several authors have used the average shale values or the average crustal values (Hakanson 1980; Pekey et al. 2004), because they have not had background values. In the present study, the background values obtained from the core were used as reference baseline for the elements. The CF and PLI (Table 1) were calculated using the following equations:

$$CF = C_{\text{metal}} / C_{\text{background}} \dots\dots\dots 4 \text{ (Reboredo 1993)}$$

$$PLI = (CF_1 \times CF_2 \times CF_3 \dots\dots CF_n)^{(1/n)} \dots\dots\dots 5 \text{ (Harikumar et al. 2009)}$$

Where: CF is contamination factor, C_{metal} is concentration of metal in surface sediments and $C_{\text{background}}$ is the concentration of metal in subsurface sediments, n is number of metals and PLI is pollution load index.

Table1 Contamination factor, Pollution load index and their categories

Contamination factor	Classification and description	PLI values	Pollution status
$CF < 1$	Low contamination	$0 < PLI \leq 1$	Unpolluted
$1 \leq CF < 3$	Moderate contamination	$1 < PLI \leq 2$	Unpolluted to moderately
$3 \leq CF < 6$	Considerable contamination	$2 < PLI \leq 3$	Moderately polluted
$CF \geq 6$	Very High contamination	$3 < PLI \leq 4$	Moderately to highly polluted
---	---	$4 < PLI \leq 5$	Highly polluted
---	---	$PLI > 5$	Very highly polluted

Results and discussion

Grain size distribution

Sediment grain size and water depth varied within the bay. As illustrated in Fig.2a, a high percentage of sand was found to be located at the edges and shoreline of the bay, which has shallow water depth (<1.6 m, Fig.2b). The waves and currents at these sites are highly active and can disturb the fine particles. However, muddy (silt and clay) percentages are dominant within the inner and south-west of the bay (Fig.2c), where the water depth is high and the wave slightly lower. Therefore, fine and very fine particles can precipitate within the inner bay.

Fig. 2 Spatial distribution of **a** sand percentages, **b** water depth (m) and **c** muddy percentages in Kogarah Bay

Distribution patterns of trace metals pollution

Trace metals are analysed in this study and the minimum, maximum and mean values are presented in Table 2. Prediction maps of trace metals Cr, Ni, Cu, Zn, As and Pb are shown in Fig.3. The concentration of these metals generally exhibited similar spatial distribution patterns. The highest concentrations of these metals are located in the north and north-west of the bay, and close to the discharge points and stormwater outlets from catchment areas. Notably, the concentration of trace metals increased around boat moorings and boatyards. Trace metals were also concentrated within the inner bay which is dominated by muddy particles (silt and clay) and organic matter that can absorb and accumulate trace metals on clay minerals at these sites. Conversely, the concentration of trace metals decreases toward the edges and shoreline of the bay for two reasons. First, the edges of the bay are dominated by the sand fractions. Second, the currents and waves become more active at the edges and can disturb fine particles, which are then transported and gradually deposited in deeper areas. Furthermore, the level of these metals varied in the inner part of the bay based on mobility of each metal, sediment types and environmental condition.

Table2 Basic statistic of metals with ERL (Effect Range Low) and ERM (Effect Range Median) values in ppm

Trace Metals	Cr	Ni	Cu	Zn	As	Pb
Min.	6.6	1.3	4.8	10.6	1.5	5.4
Max.	91.4	28	100	433	27.5	235.2
Mean±SD	33±23	12.2±6	36.7±24	158±99	12.3±7	87±59
Limit of detection	0.5	0.5	0.5	0.5	0.5	0.5
ERL	81 (5)	20.9 (6)	34 (31)	150 (36)	8.2 (34)	46.7 (39)
ERM	270 (-)	51.6 (-)	270 (-)	410 (1)	70 (-)	218 (1)

NA not available and (N) number of samples exceeded ERL and ERM

On the other hand, the variation of these metals at the edges and mouth of the bay was found to be relatively small. Trace metal concentrations are compared with deleterious biological effect values, where the effect range low (ERL) and effect range median (ERM) in estuarine and marine sediments are suggested by U.S. National Oceanic and Atmospheric Administration (NOAA; Ligerio et al. 2002; Bakan and Özkoç 2007), for evaluation of the effects of trace metal pollution on environmental ecosystems. The concentrations of trace metal Cr and Ni were less than ERL values, which show no contamination from low pollution for these metals. The concentration of trace metals Cu, Zn, As, and Pb were above the ERL value, which is considered to be moderate in contamination for these metals in the north, north-western and inner part of the bay Fig.3. However, Zn and Pb concentrations in some sites KO26 and KO45 within the bay exhibited extreme contamination, which exceeded ERM value for deleterious biological effects (Table 2). This is because of gasoline fume from cars and boats activities.

Fig. 3 Distribution of trace metals (a Cr, b Ni, c Cu, d Zn, e As and f Pb) in surface sediments in Kogarah Bay. Where: arrows indicate stormwater outlets

Between 0 and 30cm the concentrations of trace metals such as Cu, Zn, and Pb decrease dramatically with increasing sediment depth. However, after this depth, the concentrations are negligible. Concentrations of Ni and As decrease slightly with depth. Cr fluctuated in concentration with sediment depth (Johnston and Chrysochoou 2014). This may imply that the source of Cr is from heavy minerals such as hematite (Fig.4). Nonetheless, concentrations of Cu, Zn, and Pb are derived from anthropogenic activities. Trace metals have accumulated since European settlement which has led to increasing pollution over the years. The surface sediments are trap for trace metals especially in anoxic conditions and iron oxide-hydroxide.

Fig. 4 Variation of trace metals with sediment depth (cm) in subsurface sediments

Assessment of contamination and environmental effects of trace metals in sediments

Both contamination factor and pollution load index can provide a quick and easy assessment of the quality of the sediments at a site (Tomlinson et al. 1980). Figs.5 and 6 illustrate the CF and PLI of the trace metals in surface sediments ranging from low to considerably contaminated through the sample site. Trace metals Cr and Ni were found to be low contaminated, whereas metals such as Cu, Zn, As, and Pb were found to be moderately to considerably contaminated within Kogarah Bay. As illustrated in Figs.5 and 6, areas close to channels, discharge points and sites with many boats were moderately to considerably contaminated. These results are reliable indicators of the geochemical composition of the sediments. The CF for Cu, Zn and Pb is very high contaminated in samples KO16, KO20, KO26, KO29, KO39, KO41, KO42, KO45, KO51, KO50 and KO53 which is due to these sites being located close to moored boats and boatyards that are painted and repaired to prevent fouling.

Fig. 5 Contamination factor of trace metals in surface sediments in Kogarah Bay

Fig. 6 Pollution load index of trace metals in surface sediments in Kogarah Bay

Fig.7 and Table 3 show the findings of hierarchical cluster analysis (HCA). Three groups of variables can be recognised from these statistics. Based on similar characteristics, sediment fractions, trace metals and mineral compounds can be classified. Group A (red) contains samples from deep water and has low percentages of sand, high percentages of mud and the highest concentration of trace metals. Samples in this group, which are located close to discharge points, in the inner bay and around sites that have many boats (Fig.7), reflect anoxic conditions. Group B (green) samples have high percentages of sand, low percentages of mud and high levels of trace metals, especially Pb. This may have high percentages of organic matter, which can be a trap for trace metals (Fernandes et al. 2011). In contrast, Group C (blue) consists of samples from shallow water, with high percentages of sand, low percentages of mud and low concentration of trace metals. These samples are located at the edges and shoreline of the bay, and indicate an oxic condition.

Table3 Percentages and concentrations of variables by HCA

Variables	Cluster A	Cluster B	Cluster C
Depth	3.1	2.5	1.4
Sand%	9.9	21.7	60.2
Muddy%	90.1	78.3	39.8
Cr	37.1	38.2	29.5
Ni	17.0	15.0	7.9
Cu	45.9	46.4	28.6
Zn	222.9	199.5	98.8
As	19.5	15.3	5.8
Pb	137.7	112.3	42.9

Fig. 7 Sketch for wind direction, stormwater and environmental depositions. Where: blue arrow indicates wind directions and red arrow indicates discharge points

The concentrations of Ni, Cu, Zn, As, and Pb in surface sediments in Kogarah Bay suggested that the source of pollution had emerged from the catchment area, which includes residential, industrial part, roadsides, as well as boatyards and watercrafts. These trace metals can have deleterious effects on fauna, flora and aqua microorganisms.

Trace metals can enter into human bodies via the food chain, inhalation or dermal absorption, and may lead to critical health disorders that affect the nervous system, endocrine system and/or the immune system (Romero et al. 2013; Zhao et al. 2014).

The trace metal concentrations that were measured in Kogarah Bay were compared with the concentrations of trace metals from marine sediments (Table 4) in previous Australian studies (Birch and Taylor 1999; Jones et al. 2003) and around the world (Hamer and Karius 2002; Pekey 2006). The Huon estuary is essentially a non-industrialised zone and might contain a low level of potentially toxic trace metals. In contrast, the Port Jackson estuary, Izmit Bay and Bremen Bay have high concentrations of trace metals and are expected to be toxic. This is due to the large amounts of organic and inorganic wastes from residential and industrial areas being dumped into these water bodies. Such wastes are discharged into the bays from catchment area, boatyards and watercrafts (Irvine and Birch 1998; Hamer and Karius 2002; Jones et al. 2003; Pekey 2006). As Table 4 illustrates, the sediment in Kogarah Bay is polluted with trace metals such as Cu, Zn and Pb while the sediments in the Huon estuary has fewer pollutants than the sediments in the Port Jackson estuary, Izmit Bay and Bremen Bay.

The main anthropogenic pollution sources of trace metals in the samples of the study area can be attributed to discharge points and stormwater drains from roadways and residential areas, as well as the number of marine services and the large numbers of moored watercrafts. Kogarah Bay, which is deemed to be a non-sheltered environment exposed to the open sea and to high winds from north and north-west, provides a useful example of

the distribution of pollutant concentrations by current and wave activity. As a consequence, the trace metals are gradually precipitated within deeper sites to the inner and west of the Bay (Fig.7).

Table 4 Comparison results of the study area with other estuaries in Australia and the worldwide

Location	Country	Cr ppm	Ni ppm	Cu ppm	Zn ppm	As ppm	Pb ppm	References
Huon Range Mean	Australia	5-80 71	2-28 20	7-32 17	2-66 40	4-25 16	2-48 25	(Jones et al., 2003)
Port Jackson Range Mean	Australia	BDL	5-24 22	9-1053 188	108-7622 651	BDL	38-3604 364	(Irvine and Birch, 1998)
Bremen Bay	Germany	131	60	87	790	NA	122	(Hamer and Karius, 2002)
Izmit Bay	Turkey	74.3	NA	67.6	930	NA	102	(Pekey, 2006)
Kogarah Bay Range Mean±SD	Australia	6.6-91 33±23	1.3-28 12±20	4.8-100 36±24	10.6-433 158±99	1.5-27 12±7	5.4-235 87±59	This study

Where: BDL below detection limit and NA not available

Conclusions

To evaluate the environmental status of Kogarah Bay in NSW, Australia, and to identify potential sources of pollution of marine sediments, trace metals were analysed in both surface and subsurface sediments. The trace metals of interest were Cr, Ni, Cu, Zn, As and Pb. Multivariate statistical analyses were applied for the contamination factor, pollution load index and hierarchical cluster analysis. The findings demonstrate that the Bay has low contamination of Cr and Ni, but a high level of contamination with Pb, and moderate to considerable contamination with Cu, Zn and As. Statistical analysis of sediment samples recognised three groups of environments. The first and second group represent moderately to considerably polluted environments. The third group shows unpolluted environments. In addition, stormwater outlets and discharge points are considered to be the source of trace metals in Kogarah Bay, together with emissions such as gasoline fumes from vehicle and boat exhausts which have accumulated over time since European settlement. In further studies, there is a need to analyse water and organisms to determine acceptable levels of trace metals in order to protect aquatic-organisms from trace metal contamination. Physical, chemical and biological treatment methods are recommended to remove and/or reduce contamination from stormwater outlets before discharges into the bay.

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