Environ Geol (2007) 53:501–516
DOI 10.1007/s00254-007-0662-5

ORIGINAL ARTICLE

The chemistry of suspended particulate material in a highly contaminated embayment of Port Jackson (Australia) under quiescent, high-wind and heavy-rainfall conditions

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Received: 10 October 2006 / Accepted: 23 January 2007 / Published online: 8 March 2007

Abstract This study investigated physico-chemical characteristics of the water column and chemistry of suspended particulate material (SPM) under quiescent, high-wind and high-wind/heavy-rainfall conditions in Homebush Bay, a highly contaminated embayment of Port Jackson (Australia) to distinguish source and possible adverse effects to benthic and pelagic animals. Mean concentrations in surficial sediment were <1, 14, 181, 141, 37, 290 and 685 μg g^{-1} for Cd, Co, Cr, Cu, Ni, Pb and Zn, respectively. Sediment chemistry indicated these metals had multiple sources, i.e. the estuary, stormwater and industry. Mean total suspended solids (TSS) were 7, 17 and 20 mg L^{-1} during quiescent, high-rainfall and heavy rainfall/high wind conditions, respectively, whereas SPM Cd, Co, Cr, Cu, Ni, Pb and Zn concentrations varied between 13–25, 166–259, 127–198, 38–82, 236–305 and 605–865 μg g^{-1}, respectively under these conditions. TSS and total water metal concentrations were lowest during quiescent conditions. High TSS and metal loads in surface water characterised high-rainfall events. Wind-induced resuspension contributed the greatest mass of SPM and metals to the water column. Benthic animals may be adversely affected by Pb and Zn in sediment. Total water Cu and Zn concentrations may pose a risk to filter-feeding animals in the water column due to resuspension of contaminated sediment.

Keywords Suspended particulate matter · Heavy metals · Stratified estuaries · Well-mixed estuaries · Contaminant risk

Introduction

Estuaries readily trap sediments and fine material and associated contaminants tend to accumulate in areas of low energy in these environments (Hedges and Keil 1999). Suspended particulate material (SPM) is the major route of contaminant transfer between bed sediment, the overlying water and organisms within estuaries (Owens et al. 2001; Carter et al. 2003; Turner and Millward 2002). Resuspension of bed sediment by wind-driven waves and stormwater runoff following high-precipitation, are the main sources of SPM in microtidal estuaries. Sediment resuspension is the re-cycling of bottom sediment back into the water column by meteorological, hydrological, or anthropogenic processes and is well understood (Schoellhamer 1995, 1996). The hydrology of stratified estuaries produced through prolonged high precipitation is also well documented, e.g. Santa Monica Bay (Bay et al. 2003; Washburn et al. 2003), San Diego Bay (Schiff et al. 2003), Newport Bay, USA (Lee et al. 1999) and Fouha Bay, Guam (Wolanski et al. 2003). Although the physical processes associated with resuspension and stratification is well known, the chemistry of SPM under these conditions is less well documented (Taylor 2000; Simpson et al. 2002). Heavy metal distributions in surface sediment provide a long-term, integrated assessment of contaminant supply in depositional estuarine environments, whereas SPM chemistry gives information on short-term processes controlling supply and dispersion.

Extensive areas of Port Jackson (Fig. 1) are mantled in sediments containing high concentrations of a wide range

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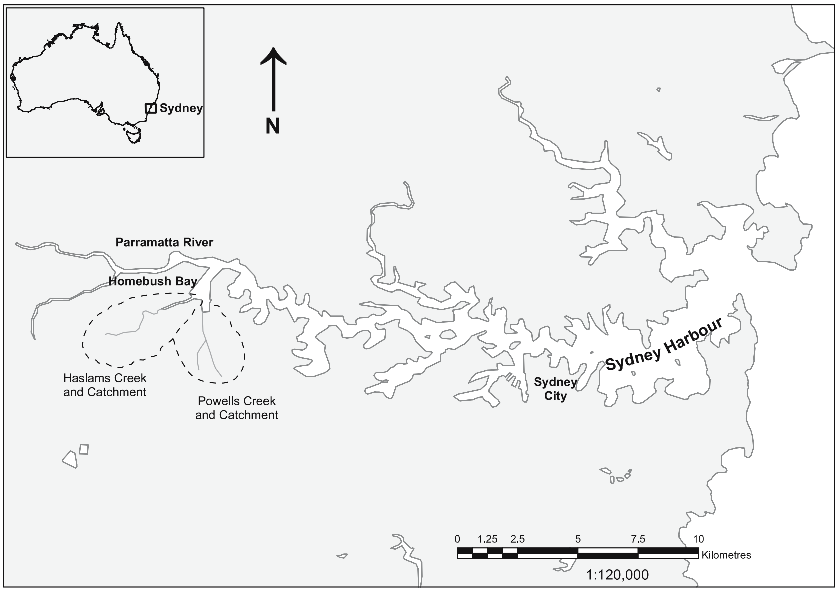


Fig. 1 Location of Homebush Bay and Haslams and Powells Creek catchments

of contaminants due to an abundance of fine sediment and high (86%) industrial and residential land use in the catchment (Birch and Taylor 2004). The distribution, concentration and provenance of contaminants in bottom sediments of Port Jackson have been the focus of extensive research in recent years (Irvine and Birch 1998; Birch and Taylor 1999; Taylor 2000; McCready et al. 2000), but contaminants within the water column have been largely neglected (Taylor and Birch 1999; Simpson et al. 1998). Homebush Bay is less than 2-m-deep in the eastern half, deepening to 5 m in the west (mean depth 2.9 m) and is one of the most-contaminated embayments in Port Jackson (Fig. 1). Sediments containing high concentrations of metallic and organic contaminants mantle extensive areas of the bay, especially in the shallow (<2 m) eastern part adjacent to the Rhodes Peninsula (JET 1990, 2001; USEPA 1991; Parametix Inc and AWT Ensight 1996; EVS Environmental Consultants 1998; Birch and Taylor 1999; McCready et al. 2000; URS Australia 2002).

Port Jackson is almost completely saline (well mixed) under conditions of no precipitation, but is well stratified after high-precipitation events (Wolanski 1977; Pitblado 1978; Pitblado and Prince 1977; Birch and Taylor 2004). Tidal resuspension in this microtidal (~2 m) estuary is negligible, but wind-wave resuspension is frequently observed in extensive parts of the many shallow, muddy embayments of Port Jackson, especially during the windy summer months (Pitblado 1978; Taylor and Birch 1999; Taylor 2000). The aims of the current research are to determine the distribution and chemical nature of SPM in Homebush Bay under quiescent, high-precipitation and high-wind conditions and to determine the relative contribution of catchment-derived stormwater and estuarine-sourced, resuspended bottom sediment to the water column under these conditions.

Materials and methods

Surficial sediment

Bottom sediment was sampled in May 2004 using a small stainless-steel box corer (15 × 15 × 15 cm) deployed by hand from a boat. Surficial samples ($n = 39$) were collected on ten east–west transects of Homebush Bay and locations were determined using a Garmin 12XL Global Positioning System (GPS) (Fig. 2). The upper (<2 cm) hydrous layer (>50% pore water) was selectively sampled to ensure that sediment likely to re-enter the water column during resuspension was collected. Samples were stored on ice in the field and transferred to a cool room (<4°C) on return to the laboratory (<6 h) to minimise effects of microbial activity and preserve sediment chemistry.

Textural analysis was undertaken by sieving through a <62.5 μm nylon mesh and applying a texture classification scheme based on the percentage mud (Lewis and McConchie 1994). Data were size-normalised (<62.5 μm) to minimise the confounding effects of variable grain size and to facilitate comparisons between SPM and surficial sediment (Förstner and Wittmann 1979; Birch 2003).

Total acid-extractable metals (Cd, Co, Cr, Cu, Ni, Pb and Zn) were determined by *aqua regia* digestion of sediment (USEPA method 200.8) (Birch et al. 1999) using a Vista Varian Inductively Coupled Plasma Atomic Emission Spectrometer (ICP-AES). Analytical precision and accuracy were established using the International Certified Reference

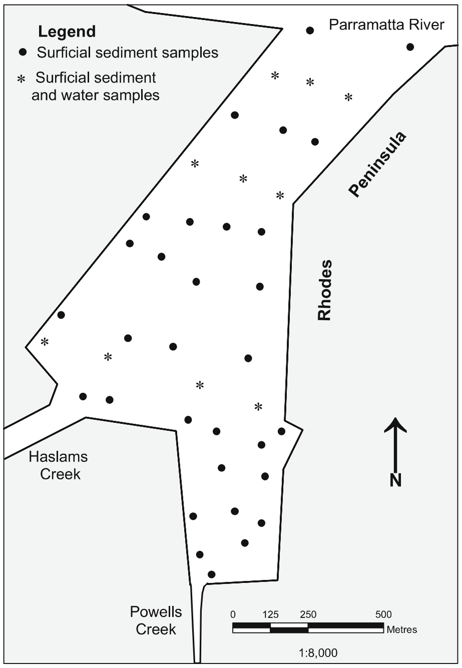


Fig. 2. Homebush Bay and sampling locations

Material AGAL-10 (Australian Government Analytical Laboratory) and an internal laboratory standard (ILS-3). Precision was <10% relative standard deviation (RSD) for all trace metals and recovery was 91–101%. Procedural blanks were below analytical detection (1 μg g^{-1}).

Water sampling

Water samples were collected at ten sites on three transects in Homebush Bay (Fig. 2) under three weather conditions; quiescence (25 June 2004), calm/heavy-rain (18 August 2004), and high-wind/heavy-rain (2 October 2004). Fieldwork for quiescent conditions took place during a dry (>10 days), calm period (wind <2 m s^{-1}) with low SPM (mean 7 mg L^{-1}). Sampling took place for the heavy-rain event during calm conditions (wind <2 m s^{-1}) following 2 days of rainfall (115 mm) on the Homebush Bay catchment. Water quality monitor data presented later shows the whole bay was occupied by a turbid, buoyant plume of stormwater) during this period. The wind during the high-wind/heavy rain survey was consistently >5 m s^{-1} and gusting up to 10 m s^{-1} with maximum wave heights of 30–40 cm and rainfall was 50 mm (BOM 2004).

Water samples were collected in 5 L polyethylene bottles pre-cleaned by soaking for 4 h in a warm detergent solution (Extran-MAO3), rinsing with deionised (DI) water and soaking in a 5% HNO_3 bath for at least 24 h. The bottles were soaked in ultra pure water for 24 h, dried in a laminar fume hood and stored in two airtight polyethylene bags.

An YSI 6920 field water quality monitor was used to measure physico-chemical properties, including dissolved oxygen (DO), turbidity, conductivity and pH at sites from which sediment samples were collected. Instrument probes were calibrated before each survey. Water samples were collected during a 2-h period on either side of high tide to reduce influence of tidal resuspension and to maintain consistent tidal conditions during sampling of different events. Sample bottles were removed from protective plastic bags and rinsed once with ambient water prior to water collection. Subsurface water was sampled from 0.5 m above the sediment bed using a metal-free submersible pump, whereas surface water was collected from just beneath the water surface to exclude floating debris. The pump was tested for possible metal contamination prior to use in the project. Ambient water was pumped for 3–5 min between samples to rinse the sample path and to prevent cross contamination. Sample bottles were filled to the top to exclude air, capped immediately and sealed in two polyethylene bags. Samples were taken upstream of the boat to avoid contamination.

Temperature (°C), pH, turbidity (NTU), specific conductivity (mS cm^{-1}) and DO (mg L^{-1}) were measured while water samples were being collected.

Water samples were refrigerated (<4°C) and kept in the dark in the laboratory to minimise effects of microbial activity. Samples were filtered through pre-dried 0.45 μm cellulose nitrate filter papers using a manifold filter system under vacuum and acidified with 5% HNO_3. TSS was determined by washing the filtrate twice using DI water to remove salt. Components of the filter system were acid-washed (5% HNO_3) between samples to prevent cross contamination. Filter papers were dried to a constant weight and placed in 50 mL test tubes and digested in 1:1 w/w $HCl:HNO_3$ (US EPA Method 200.8). SPM samples were analysed for Cd, Co, Cr, Cu, Ni, Pb and Zn by ICP-AES. In this work, TSS refers to the mass of suspended solids in water and is reported as mg L^{-1}, whereas SPM is used for suspended particulate metal concentrations and are provided as μg g^{-1} dry weight. Water samples were analysed at the National Measurement Institute, Sydney, which is a National Association of Testing Authorities (NATA) certified laboratory. Water was diluted 1:10 before analysis on a Perkin Elmer Elan 6000-DRC Quadrupole Inductively Coupled Plasma Mass Spectrometer (ICPMS) for Cd and Pb using isotopes Cd 114 and Pb (combined 208 + 207 + 206). Copper and Zn were analysed on a Finngan-Matt Element–1 High Resolution ICPMS using isotopes Cu63, Zn64. Detection limits are 1.0 μg L^{-1} for Cd Cu, Pb and Zn. Precision of analysis for Cd, Cu, Pb and Zn is 6, 7, 5 and 7.7% RSD. Measurement of uncertainty (MU) is 12, 14, 10 and 16% of the reported value with a confidence interval of 95%.

The quality of field and laboratory analytical procedures for SPM metals was assessed as for surficial samples using the same certified reference material (AGAL-10). For every batch of 15 SPM samples, an International Reference Material, a replicate and a blank were prepared to determine accuracy and precision, and to detect procedural contamination. Field and laboratory blanks were below detection (1 μg L^{-1}). Precision for Cd, Cr, Cu, Pb and Zn was <10% RSD and <15% for Ni and Co and accuracy, expressed as percentage recovery, was 93–102% for all metals.

Mass of metals in SPM (μg g^{-1}) was calculated by multiplying TSS and SPM metal concentrations and total water metal concentrations were provided as particulate mass per litre (μg L^{-1}), and exclude the dissolved phase.

Geographical information system (GIS)

Mapping SPM and surficial sediment data onto a cadastral map of Homebush Bay was performed using Arc Mapper GIS software. The Spatial Analyst tool was used to interpolate spatial data by Inverse Distance Weighting (IDW) applying a variable neighbourhood size and grid scale of 0.0001288895 m.

Possible biological effects

Possible adverse biological effects posed by metals contained in surficial sediments and SPM phases were assessed using local sediment quality guidelines (SQGs) (ANZECC/ARMCANZ 2000), which are based on a North American scheme (Long et al. 1995; Long and MacDonald 1998). SQGs provide two values, Interim Sediment Quality Guidelines-Low (ISQG-L) and -High (ISQG-H), which delineate three concentration ranges for a chemical. Concentrations < ISQG-L represent a minimal-effects range, below which biological effects are rarely observed, whereas concentrations > ISQG-L < ISQG-H is a range within which biological effects occur occasionally. Concentrations > ISQG-H is a probable-effects range above which, adverse biological effects frequently occur. The North American equivalents to ISQG-L and ISQG-H values are ERLs and ERMs. Water quality guidelines (WQGs) used for assessing total and dissolved water phases are hierarchal and require an assessment against total water concentrations first and if exceeded, dissolved water concentrations are tested (ANZECC/ARMCANZ 2000).

Results

Surficial sediment

Surficial sediment mantling Homebush Bay were predominantly mud (<62.5 μm). Sediments (<62.5 μm) in the south east contain high concentrations of Pb (>900 μg g^{-1}) and Zn (>1,000 μg g^{-1}) and in the central eastern area sediments were rich in Cu (>150 μg g^{-1}) and Ni (>100 μg g^{-1}), whereas Cr (>200 μg g^{-1}) was most elevated in northern Homebush Bay (Fig. 3 changes in grey scale are due to variable neighbourhood interpolation size used) (Table 1).

Quiescent conditions

Physico-chemical characteristics of the water column

Water quality data were taken at the surface and 0.5 m off the bottom. Temperature and pH values were consistent spatially and with water depth, however EC, DO and turbidity varied considerably (Table 2).

TSS and the chemistry of SPM

The distribution of TSS in bottom and surface water were similar, but bottom water was twice surface water concentrations and TSS was highest in the south east of the bay (Fig. 4).

Cadmium concentrations were below detection for all samples. Surface water SPM metal concentrations were consistently higher than bottom water sediments, except for Co. Bottom water SPM heavy metals have similar spatial patterns, i.e. metal concentrations increased south- and eastwards (mean concentrations for Cr, Cu and Pb on the western side was 244, 109 and 193 μg g^{-1}, respectively compared to 277, 146 and 286 μg g^{-1}, respectively on the eastern side) (see Fig. 5, representing the distribution of all metals) (Table 3). The spatial distribution of Cu and Pb in surface water SPM increased towards the south east similar to bottom water, whereas surface water Zn, Cr, Co and Ni was highest in the north west (Fig. 5) (Table 3).

Dissolved metal concentrations under quiescent conditions

Concentrations of dissolved Pb and Cd were below detection (<1 μg L^{-1}) under quiescent conditions and dissolved Cu concentrations (mean 1.4 μg L^{-1}) were an order of magnitude lower than Zn (mean 16.6 μg L^{-1}) (Table 4).

High-precipitation conditions

Physio-chemical characteristics of the water column

Electrical conductivity varied considerably under high-precipitation conditions due to stratification of the water column, whereas pH remained nearly constant. Salinity of surface water (mean 18 mS cm^{-1}) was consistently lower than bottom water (mean 44 mS cm^{-1}) under high-pre-

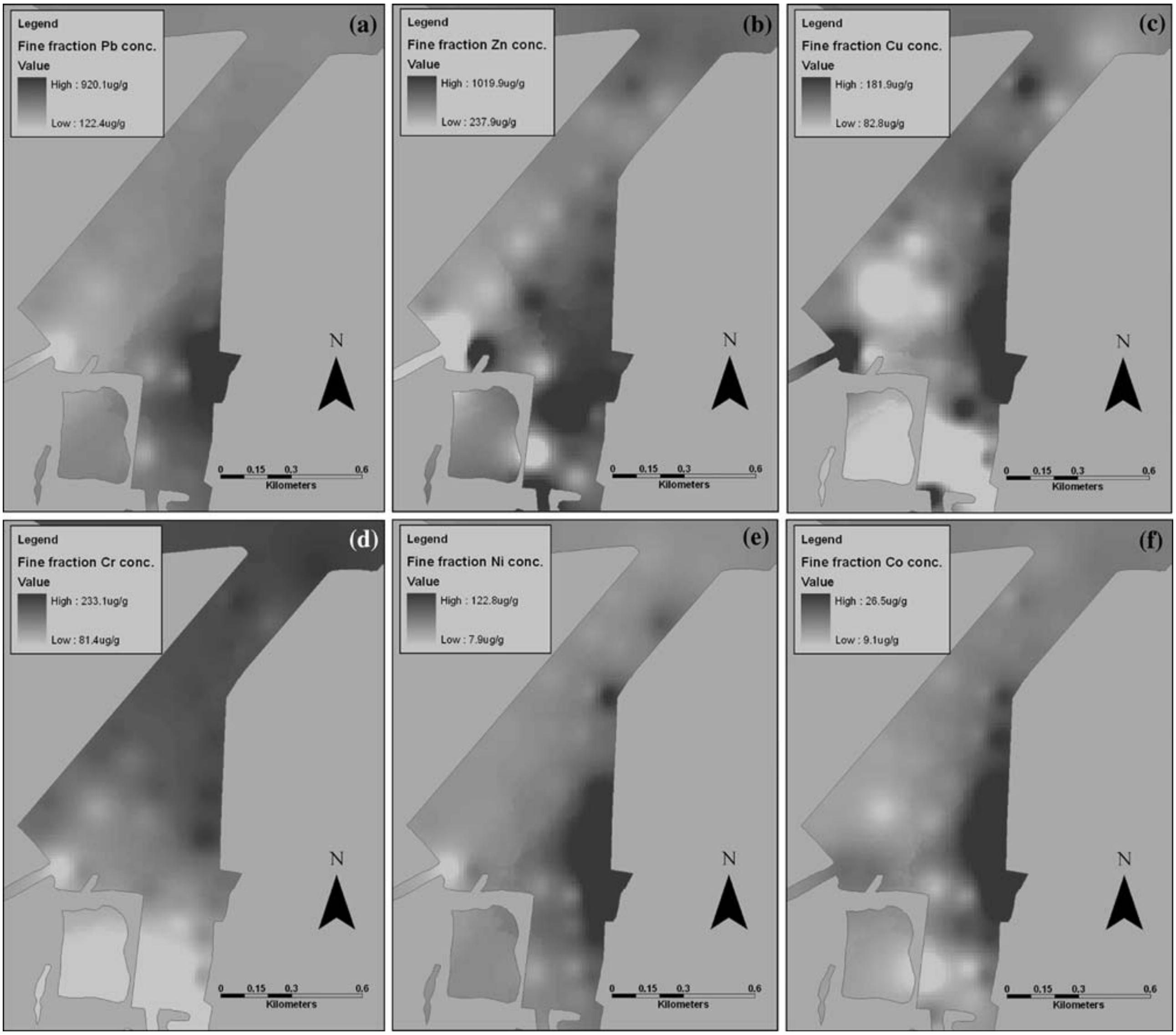


Fig. 3 Distributions of **a** Pb, **b** Zn, **c** Cu, **d** Cr, **e** Ni and **f** Co in surface sediment

Table 1 Chemistry of surficial sediment in Homebush Bay

	Cd	Co	Cr	Cu	Ni	Pb	Zn
Total sediment (<2,000 μm), *n* = 41							
Mean	<1	14	*181*	*141*	*37*	**290**	**685**
Range	bd–7.0	8–25	71–*226*	*75*–*170*	4–**118**	*65*–**886**	126–**850**
Normalised sediment (<62.5 μm), *n* = 41							
Mean	1.5	15	190	150	39	305	722
Range	bd–8.4	9–27	81–237	83–183	8–124	122–927	236–1021
Sediment quality guidelines							
ERL[a]	1.2	NA	81	34	20.9	46.7	150
ERM[a]	9.6	NA	370	270	51.6	218	410
ISQG-L[b]	1.5	NA	80	65	21	50	200
ISQG-H[b]	10	NA	370	270	52	220	410

Figures underlined are above ERL/ISQG-L

Figures in bold are above ERM/ISQG-H

Concentrations in μg g^{-1}, *bd* below detection (1 μg g^{-1}), *NA* not available

[a] Long et al. (1995),

[b] ANZECC/ARMCANZ (2000)

Table 2 Physico-chemical characteristics of the water column

	Temperature (°C)	Conductivity (mS cm^{-1})	DO (mg L^{-1})	pH	Turbidity	TSS (mg L^{-1})
Quiescent conditions, $n = 64^a$						
Mean	13	44.5	11.2	7.7	7.2	7
Range	12.3–13.7	11.2–57.0	4.5–17.2	7.0–8.0	1.4–10.3	3.2–18.5
High-precipitation conditions, $n = 20$						
Mean	NA	28	NA	7.6	29.4	17.2
Range	NA	13.5–51.3	NA	7.1–7.8	13.9–48.7	7.8–41.2
High-wind/heavy rainfall conditions, $n = 64$						
Mean	17.2	35.5	9.1	7.1	56.8	20.8
Range	15.8–18.1	18.2–49.2	6.3–11.2	6.9–7.4	3.3–38.3	11.2–41.6

NA Not available; $n = 64^a$ except for TSS, $n = 22$

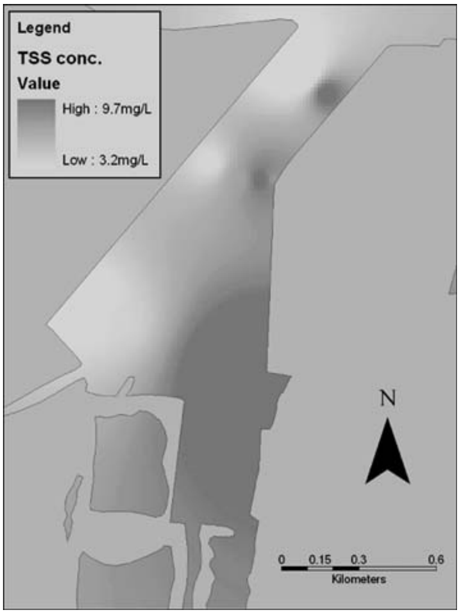


Fig. 4 TSS concentrations for surface water during quiescent conditions

cipitation conditions. Low salinity in top and bottom water in the eastern bay (mean 18.1 mS cm^{-1}) indicated that the stormwater plume occupied the majority of the water column in the shallow (1–3 m) parts of the bay, but not in the deeper (~5 m), western side under these conditions.

TSS and SPM metal concentrations

Mean TSS (20 mg L^{-1}) and turbidity (31 NTU) in surface water were higher than bottom water (mean 15 mg L^{-1} compared to 28 NTU) and TSS decreased with increasing depth towards the west in a similar manner to TSS during quiescent conditions (Fig. 6).

SPM metal concentrations varied considerably and were greater under high-precipitation conditions than during quiescent or high wind/rainfall events. Distributions of particulate metals in surface water were similar for all heavy metals, i.e. maximum concentrations were observed at the mouth of the bay (see SPM Cu distribution, which was typical of all metals, Fig. 7).

The distribution of particulate metals in bottom water varied substantially. Zinc and Pb concentrations increased northwards, Cu was most abundant along the eastern shoreline, Co and Cr concentrations were highest at the mouth of Haslams Creek, and Ni concentrations increased toward the south (Fig. 8).

Dissolved metal concentrations

Dissolved Cu and Zn concentrations were considerably higher during high-precipitation (mean 16.6 and 28.2 μg L^{-1}, respectively) than under quiescent (mean 1.4 and 2.7 μg L^{-1}, respectively) conditions. Cadmium and Pb were below detection.

High-wind/heavy-rain conditions

Physio-chemical characteristics of the water column

DO and EC varied considerably under high-precipitation/wind conditions due to stratification of the water column and both parameters were higher under these conditions than during the high-precipitation and quiescent events, whereas pH remained consistent under all weather conditions.

TSS and SPM heavy metal concentrations

TSS and turbidity in surface water were highest off Haslams Creek and along the eastern shoreline (mean 22.6 mg L^{-1} and 92 NTU) and decreased with increasing depth toward the west (means 19.1 mg L^{-1} and 20.3 NTU)

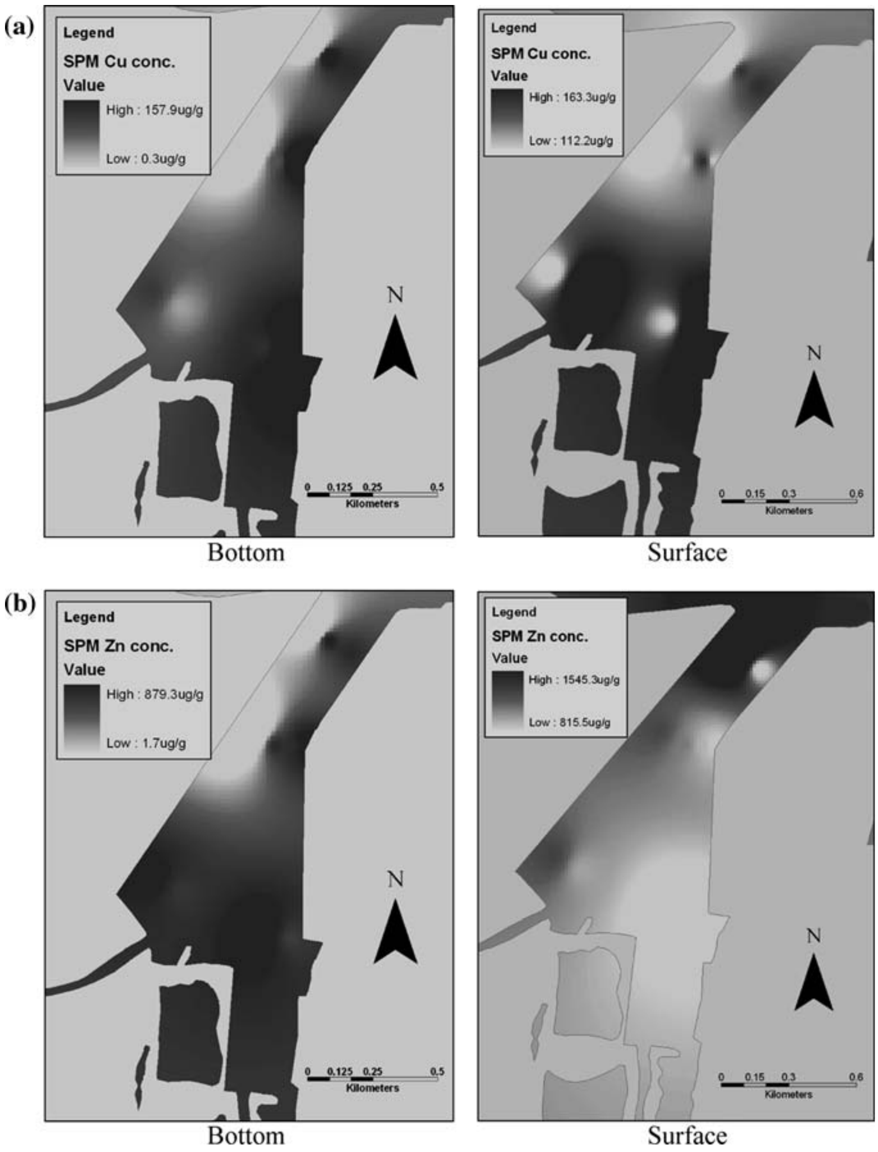


Fig. 5 Spatial distribution of **a** Cu and **b** Zn (representing Cr, Co and Ni) under quiescent conditions

Table 3 Particulate heavy metal concentrations

	Co	Cr	Cu	Ni	Pb	Zn
Quiescent conditions, n = 20						
Mean	13	259	127	**82**	**249**	**865**
Range	9–18	58–515	42–163	23–**158**	67–**348**	280–**1,081**
High-precipitation conditions, n = 20						
Mean	25	198	198	**64**	**305**	**853**
Range	15–142	84–**708**	127–**1,126**	31–**301**	197–**1,665**	**614–4,430**
High-wind/heavy rainfall conditions, n = 20						
Mean	18	166	119	38	**236**	**605**
Range	15–26	97–250	87–155	33–48	160–**318**	**497–798**

Concentrations in µg g^{-1}

Figures underlined are above ERL/ISQG-L

Figures in bold are above ERM/ISQG-H

(Fig. 9). TSS in bottom water increased adjacent to the mouth of Haslams Creek and in the shallow parts of the southern embayment.

SPM metal distributions in bottom water were distinct from surface water. Highest concentrations of Cr, Cu, Pb and Zn (180, 147, 318 and 798 µg g^{-1}, respectively) in

Table 4 Dissolved heavy metal concentrations

	Cu	Zn
Quiescent conditions, $n = 8$		
Mean	1.4	16.6
Range	1.2–1.6	13.0–29.0
High-precipitation condition, $n = 6$		
Mean	2.7	28.2
Range	1.7–4.4	1.0–46.0
High-wind/heavy-rainfall conditions, $n = 6$		
Mean	7.8	23
Range	7.0–8.8	15–32
IWQGs	0.35	2.7

Cd below detection (1 µg L^{-1}); Concentrations in µg L^{-1}

Figures underlined are above IWQGs

IWQGs Interim water quality guidelines

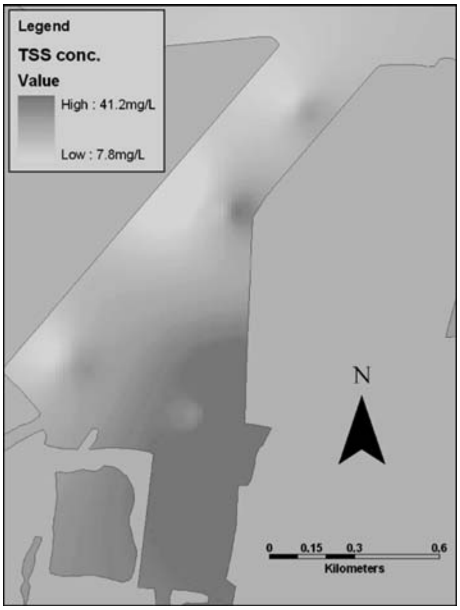


Fig. 6 TSS distributions in surface water following high-precipitation conditions

surface water were in the central bay and along the eastern shoreline (Fig. 10). Highest concentrations of particulate Ni (43 µg g^{-1}) and Co (26 µg g^{-1}) in surface water were in the middle and northern parts of the bay.

Maximum concentrations of SPM Cr, Cu, Pb and Zn (249, 155, 308 and 606 µg g^{-1}, respectively) in bottom water were in the southeast of the bay (Fig. 10) and concentrations decreased northwards, particularly along the northeast shoreline. Highest Ni SPM concentrations (43 µg g^{-1}) in bottom water were along the eastern shoreline and Co concentrations gradually increased northwards.

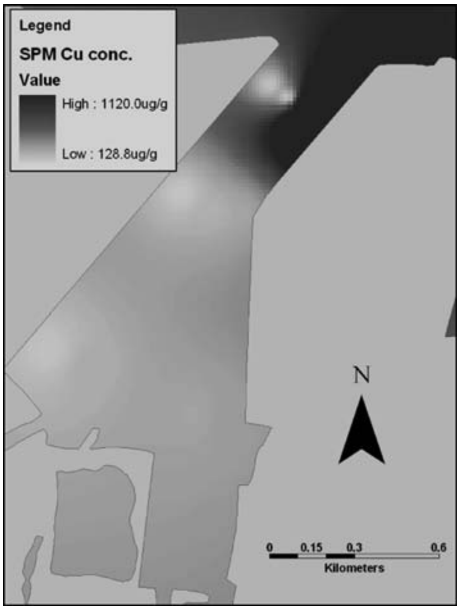


Fig. 7 Distribution of Cu SPM in surface water following high-precipitation

Dissolved metal concentrations

Cadmium and Pb were below detection in the dissolved phase and mean Zn concentrations (23.0 µg L^{-1}) were approximately three times higher than Cu (7.8 µg L^{-1}).

Discussion

Surficial sediments and the source of heavy metals

In the shallow, low-energy environments of Port Jackson, surficial sediment comprises an oxic, low-density, hydrous muddy material (>95% mud and >50% water) (Irvine and Birch 1998; Birch and Taylor 2000), commonly referred to as the ‘hydrous layer’, or the ‘readily resuspended sediment’ (RRS) (Luoma et al. 1990). This layer is up to 5-cm-thick in the harbour and is distinct from the darker, more cohesive and anoxic underlying substrate (Birch and Taylor 1999). The chemical nature of the hydrous layer is similar to the chemistry of SPM because particulates cycle between the estuary floor and water column dependant on ambient energy (Birch and Taylor 2004; Simpson et al. 2002). When surficial sediment is resuspended, the physical (mineralogy and grain size) and chemical (matrix and contaminants) characteristics of the hydrous layer are preserved, allowing the source of SPM to be determined (Förstner et al. 1989; Birch and Taylor 1999, 2000; Carter et al. 2003).

Fig. 8 Spatial distribution of **a** Zn (representing Pb), **b** Co (representing Cr), **c** Ni and **d** Cu concentrations in bottom water following high-precipitation

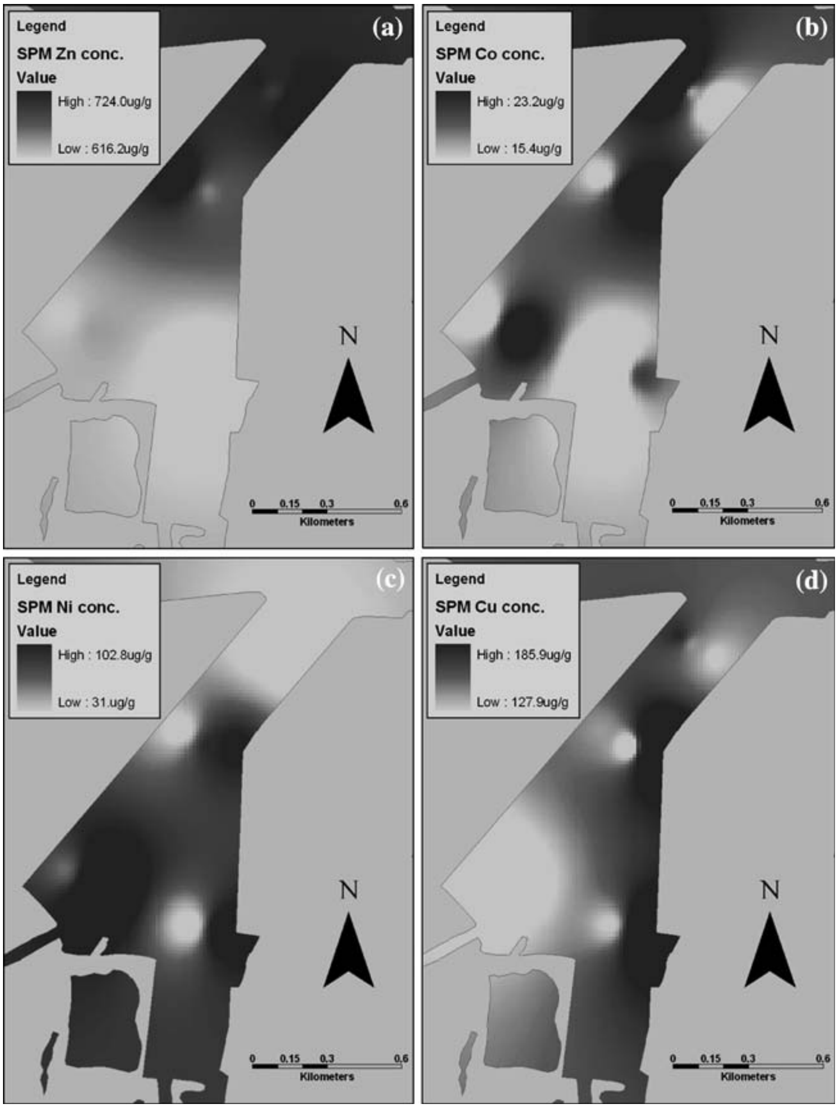


Fig. 9 TSS in the **a** bottom and **b** surface water during high-wind/rainfall conditions

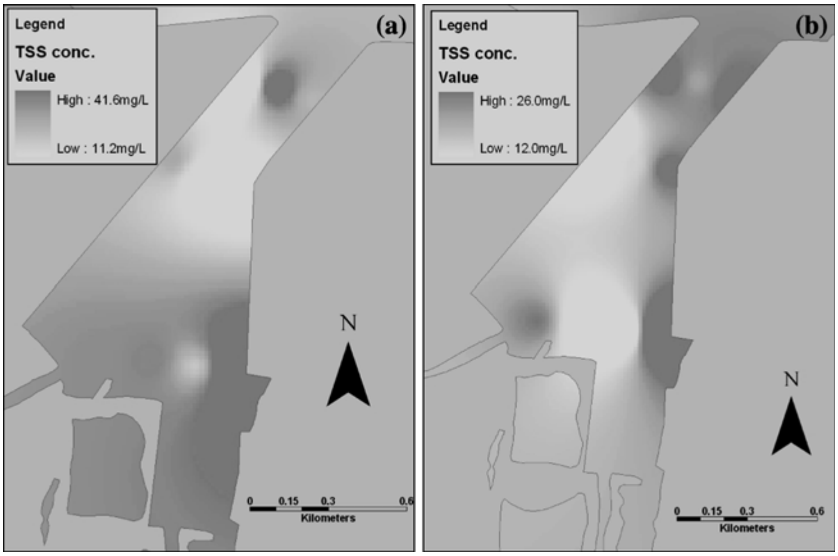
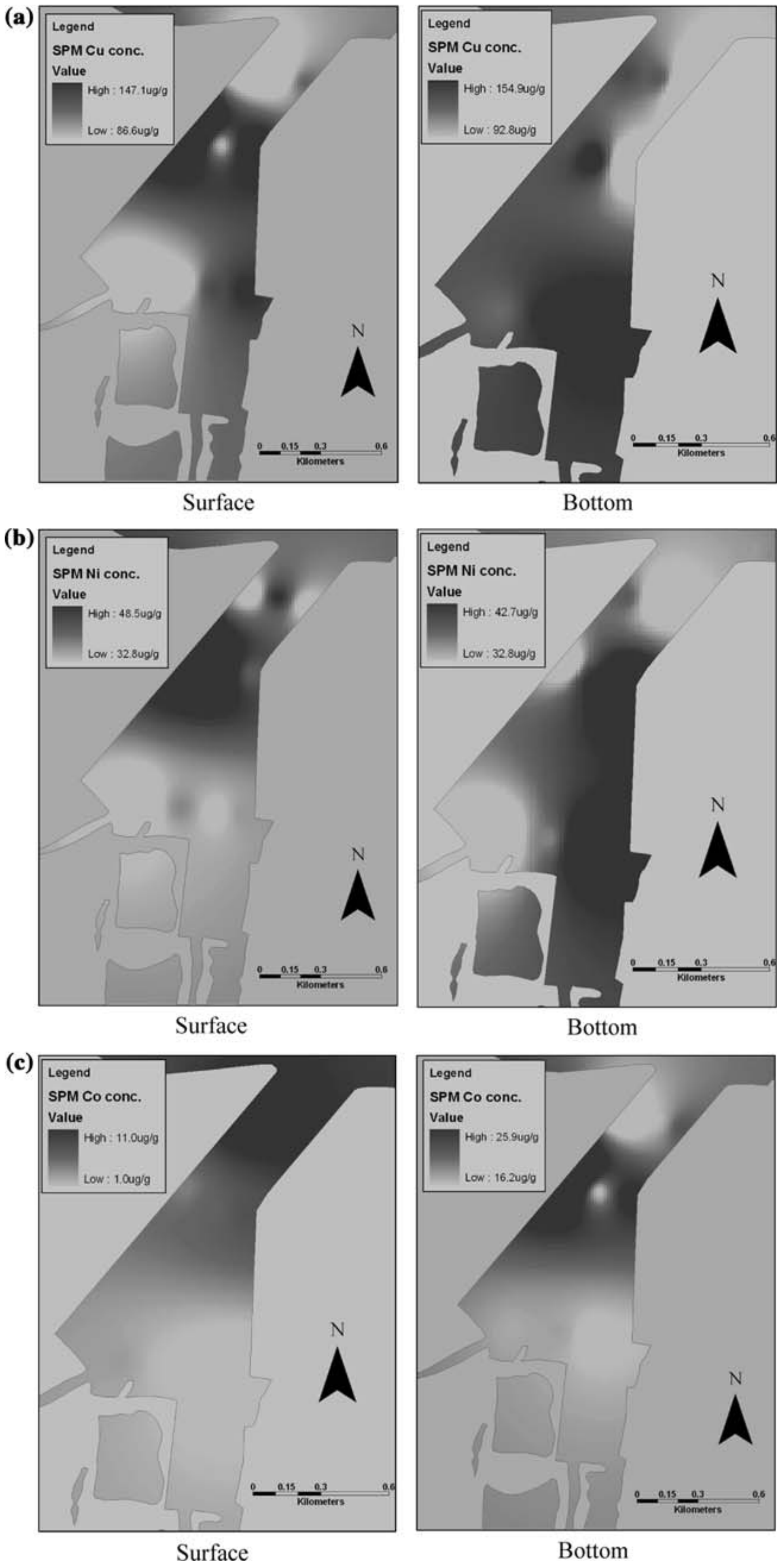


Fig. 10 Concentrations of **a** Cu (representing Cr, Pb and Zn), **b** Ni and **c** Co during heavy-wind/rainfall conditions (**b**) Zn during wind/rain conditions



Surface Bottom

Surface Bottom

Surface Bottom

Increasing concentrations of Co, Cr, Cu, Ni, Pb and Zn towards the eastern shoreline of Homebush Bay suggest that the industrial complexes on the Rhodes Peninsula are important sources of these metals in surficial sediment (Fig. 3, Table 1). Maximum sedimentary Pb concentrations in the south east of the bay indicate industry in southernmost part of the peninsula was the major source of this metal. Stormwater discharge into Port Jackson from a heavily urbanised and industrialised catchment is an important contributor to the contamination of sediment in the estuary (Birch and Taylor 2000; 2004). Haslams Creek is probably the source of sediment containing high-concentrations of Cd and Cu located off the mouth of this creek and strongly increasing Cr concentrations at the mouth of Homebush Bay suggest that Parramatta River is supplying this metal to the bay.

Quiescent conditions

Results obtained during quiescent conditions provide baseline data against which the influence of wind resuspension and stormwater discharge on SPM may be assessed. Waters of Port Jackson are under quiescent conditions the majority of the time and therefore these SPM characteristics represent typical base-case physiochemical conditions for the estuary.

Mean TSS (7 mg L^{-1}) during quiescent periods in the current investigation was lower than mean values reported previously, i.e. 20 mg L^{-1} (AWT 1994; Ferguson et al. 1995) and 20–30 mg L^{-1} (Taylor 2000) for other bays in Port Jackson. However, low-precipitation TSS concentrations determined over a full annual period are similar to the current investigation of Homebush Bay, i.e. 10 mg L^{-1} (Hatje et al. 2001). Mean TSS at the surface (6 mg L^{-1}) was lower than at 0.5 m off the bottom (8 mg L^{-1}) and TSS was highest in the east, suggesting that, despite the low-energy conditions, some resuspension was taking place in the shallow (<1 m) parts of the bay during quiescent conditions.

In contrast to TSS, metal concentrations in surface water SPM were higher than bottom water SPM. This inverse relationship between TSS and SPM chemistry has been observed previously (Turner 1996; Zhou et al. 2003; Benoit et al. 1994) and is a consequence of the diluting effect of more abundant metal-poor coarse material in bottom water. Nevertheless, higher mean total water metal concentrations in bottom water than surface water for all heavy metals indicate that particulate metal transport was predominantly associated with bottom water under quiescent conditions.

All SPM heavy metals in bottom water showed similar spatial patterns, i.e. concentrations increased towards the shallow eastern shore, suggesting that resuspension of metal-rich surficial sediments bordering the highly industrialised Rhodes Peninsula is an important source of metals to the water column. SPM metal concentrations in surface and bottom waters had similar spatial distributions for Cu, and Pb in the southern parts of the embayment, indicating that resuspension influenced the whole water column in shallow-water areas. High SPM Zn, Ni, Cr and Co concentrations in surface water in the northern part of the bay suggest that some particulate metals were entering Homebush Bay from Parramatta River in the upper water column.

High-rainfall conditions

Estuaries with large drainage areas generally remain well-mixed following high-precipitation events, e.g. the River Po, Italy (Davide et al. 2003), River Geul, the Netherlands (Leenaers 1989) and River Tawe, Wales (Bird 1987), however microtidal estuaries with small, highly impervious catchments may develop a freshwater plume and a stratified water column following high-rainfall, e.g. Fouha Bay, Guam (Wolanski et al. 2003) and Santa Monica Bay, USA (Washburn et al. 2003). Port Jackson is unusual in that it is well mixed under low-precipitation conditions and is strongly stratified following heavy rainfall (>50 mm day^{-1} for at least 2 days) (Wolanski 1977; Pitblado 1978; Birch 1996; Taylor 2000).

The nature of TSS and associated contaminants transported by stormwater reflects land use types within the catchment (Carter et al, 2003) and the chemistry of SPM introduced into estuaries has been used to discriminate between contaminant sources within drainage basins (Wall and Wilding 1976; Peart and Walling 1986).

Lower conductivity of surface water than bottom water (mean 18 mS cm^{-1}; bottom water mean 29 mS cm^{-1}) during the high-rainfall event indicated that stormwater was dispersed over most of the bay and low salinity surface and bottom water suggested that stormwater occupied the whole water column in the shallow (<1 m), eastern bay at these times.

Higher mean TSS (20 mg L^{-1}) and turbidity (31 NTU) in the surface stormwater plume than in bottom water (15 mg L^{-1} and 28 NTU, respectively) suggests that resuspension of the bottom sediment during heavy rain introduces less SPM into the water column than stormwater discharge. TSS was high in surface and bottom water in the southeastern part of the bay, supporting the contention that stormwater occupied the whole water column in shallow areas under high-precipitation conditions. TSS in bottom water decreased with increasing water depth towards to west, suggesting that stormwater resided as an upper, buoyant plume in the deeper parts of the bay.

Mean Co, Cu, Pb and Zn concentrations of particulate and total water were generally higher in surface water than in bottom water, indicating metals were primarily transported in surface water following heavy rainfall (Elbaz-Poulichet et al. 1991). High SPM concentrations in surface water at the mouth of Homebush Bay (typified by Cu in Fig. 7) suggest that Parramatta River was supplying all metals studied to the bay under high-rainfall conditions, whereas Co, Cr, Pb and Zn were introduced in bottom water. Low particulate metal concentrations in stormwater discharged from Haslams and Powells Creeks during high-flow conditions compared to surficial sediment were possibly due to dilution by large volumes of relatively clean rain water from the catchment after 2 days of heavy rainfall (Bird 1987; Leenaers 1989; Chambers 2000; Davide et al. 2003).

High Co, Cr, Cu and Ni concentrations in bottom water SPM in the south and the general similarity in particulate metal concentrations in bottom water and surficial sediment indicates that resuspension was the likely source of the majority of bottom water SPM, even under high-rainfall conditions.

High-wind/heavy-rain conditions

Marginally lower salinity in surface water (mean 31.5 mS cm^{-1}) than bottom water (mean 39.6 mS cm^{-1}) indicated that minor stratification of the water column was preserved under the influence of strong wind (gusting >10 m s^{-1}) and 30–40 cm-high waves during the heavy wind/high precipitation event. Higher TSS and turbidity in the bottom water relative to surface water suggests that sediment resuspension was contributing a higher sediment load to the water column than stormwater, except directly off the mouth of Haslams Creek, where stormwater supplied more SPM to surface water than resuspension did to bottom water. Evans (1994) found that the proportion of resuspended matter in the water column exceeds material supplied by stormwater runoff by a factor of 10–20. High TSS and turbidity (mean 22.6 mg L^{-1} and 92 NTU, respectively) in surface water and elevated TSS (>40 mg L^{-1}) in bottom water in eastern Homebush Bay indicate that resuspension effected the whole water column in the shallow parts of the embayment during these conditions. During rainfall and no wind, the water column is stratified and the upper water column was decoupled from the bed and inflowing metals are transported into the bay in surface water. With high rain and wind, the water column is partially mixed and stratification is week and incoming sediment and metals interact with the bed resulting in deposition and resuspension.

Metal SPM distributions in bottom and surface water were different under high-wind/rainfall conditions, suggesting that contaminants were derived from different sources. Total water metal concentrations in bottom water were higher than surface water for all metals and the chemistry of bottom water SPM and surficial sediment was similar during high-wind/heavy-rainfall conditions. Maximum concentrations of metals in bottom water were in the shallow southeastern part of the bay, similar to the spatial trends of bottom water during high-wind conditions. These results indicate that bottom water SPM is derived from surficial sediment and that resuspension contributed more metals to the water column than stormwater under these conditions in a similar manner to bottom water during the high-wind event.

Concentrations of Co Cr, Cu, Ni, Pb and Zn in surface water were highest in central Homebush Bay (represented by Cu in Fig. 10), suggesting that fresh surface water, entering the bay from the two southern creeks and from Parramatta River in the north at the end of the rainy period contained minor metals and diluted surface water SPM metal concentrations.

The turbidity–TSS relationship

Turbidity and TSS were strongly related in Homebush Bay during dry-weather ($R^2 = 0.814$) and under heavy-rainfall ($R^2 = 0.6095$) conditions, but a correlation was absent ($R^2 = 0.007$) during the high-wind event. A week separation for upper and bottom water TSS data suggests that the lack of correlation under windy conditions may be due to increased ambient energy and the introduction of coarser-grained particles into the water column through wind-induced resuspension. Different particle sizes have different light-scattering characteristics, which influence turbidity measurements, possibly disrupting the close TSS–turbidity relationship observed under quiescent and high-rainfall conditions (Papacosta 2002).

Contaminant partitioning

Dissolved metal concentrations were consistently below detection for Cd and higher for Zn than Cu under all environmental conditions assessed. Dissolved Zn concentrations (mean 6.5 μg L^{-1}) reported by Hatje et al. (2003a, b) for Port Jackson under ‘‘low-flow’’ conditions were less than that measured in current study (16.6 μg L^{-1}), whereas mean Cu concentrations (1.6 μg L^{-1}) are similar (1.4 μg L^{-1}). The difference in dissolved metal concentrations are possibly because Hatje et al. studied deep-water, central channel waters of Port Jackson where sediment and SPM concentrations are considerably lower than the metal-enriched environments of the off channel embayments, e.g. Homebush Bay.

Dissolved Cu concentrations increased with increasing ambient energy, i.e. from quiescent to high-precipitation to high rainfall/high wind conditions, and dissolved Zn concentrations were highest during high precipitation. SPM is transported primarily as flocs comprising a composite structure of organic/inorganic particles and microbial communities (Burban et al. 1990; Azetsu-Scott and Johnson 1992; Phillips and Walling 1995, 1999; Droppo et al. 1997; Droppo 2001). During transportation, particles are in a constant state of physical and chemical change. Flocculation and de-flocculation alters the surface area of the particle available for adsorption, which may affect partitioning of trace-contaminants between solid and dissolved phases (Brassard 1994). Increased ambient energy (e.g. turbulence due to wind and wave energy) may result in floc disaggregation and an increased concentration of contaminants in the dissolved phase (Brassard 1994). Colloidal particles (<0.45 μm) are effective in binding metals in solution and the affinity of colloids and SPM strengthens this trend. Metal-rich particles introduced into the water column during resuspension may also release adsorbed phases due to changes in environmental conditions, including pH and redox conditions, light availability, temperature and salinity (Förstner et al. 1989; Simpson et al. 1998, 2002; Byrd 1990; Turner 2003; Zhou et al. 2003; Filgueiras et al. 2004).

Possible biological significance

Contaminant uptake by pelagic and benthic animals is mainly via the gills for the dissolved phase and by ingestion for the particulate phase. The distribution of contaminants between these phases is therefore important in determining biological risk (Birch and Taylor 2002; Karickhoff et al. 1979; Luthy et al. 1997). Dissolved, total water and particulate metal concentrations are compared to water and sediment quality guidelines to assess possible biological significance.

The Australian and New Zealand sediment quality guidelines (ANZECC/ARMCANZ 2000) comprise two levels; the lower value, ISQG-Low, denotes the concentration below which, adverse biological effect are seldom observed and the ISQG-High level distinguishes concentrations above which, adverse biological effects are frequently observed amongst most species.

Mean Zn concentrations for surficial sediment are twice ERM/ISQG-High values and mean Cr, Cu, Ni and Pb concentrations are one to five-times above ERL/ISQG-Low levels (Table 1). SPM metal concentrations are generally higher than surficial sediment and mean concentrations are up to twice ERL/ISQG-High for Ni, Pb and Zn and three times ERL/ISQG-Low for Cr and Cu. The mass of SPM in Homebush Bay was between 17 and 50 tonnes for quiescent and high wind/heavy rainfall conditions, respectively, and the mass of metal at these times was 2–9 tonnes Cu; 1.4–2.7 tonnes Ni; 4–13 tonnes Pb and 15–36 tonnes Zn, respectively (Table 5). Water quality guidelines require that total water metal concentrations are assessed and if exceeded, are compared to dissolved concentrations. Total water Cu and Zn concentrations were exceeded under all meteoric conditions studied and Cr and Co was exceeded for maximum TSS during high-precipitation and high-precipitation/high wind events (Table 6). Dissolved Cu and Zn concentrations were also exceeded for all meteoric situations, except at minimal TSS for the high-precipitation event.

Table 5 Mass of TSS and SPM heavy metals in water column

TSS	Co	Cr	Cu	Ni	Pb	Zn
Quiescent conditions						
17.8	0.2	4.5	2.2	1.4	4.3	15.0
High-precipitation conditions						
43.7	1.1	8.5	8.5	2.7	13.0	36.6
High-wind/heavy rainfall conditions						
51.0	0.9	8.8	8.7	2.0	12.4	31.9

Mass in tonnes

If these sediment and water quality guidelines are indicative of adverse biological effects, then metals of concern for ingesting, sediment-dwelling animals are Pb and Zn and possibly Ni, whereas mean Cu and Cr concentrations were at levels where detrimental biological effects are sometimes observed. Detritus- and filter-feeding species would be at risk from ingesting Cu- and Zn-rich particulates in the water column at all times, and animals with an exposure route mainly via the gills would also be at risk to high dissolved Cu and Zn. These results highlight the need for additional research on resuspension, especially on detritus- and filter-feeding animals, in highly contaminated estuarine environments.

Conclusions

The current work investigated the chemistry of surficial sediment and SPM under quiescent, high-precipitation and high-wind/heavy-rainfall conditions in Homebush Bay, a highly contaminated embayment of Port Jackson.

Surficial sediment metal distributions indicated long-term, integrated source and dispersion patterns in Homebush Bay. Identified sources included the Parramatta River (Cr, Zn and possibly Cu), industry on the Rhodes Peninsula (Cu, Pb, Ni and Zn) and Haslems Creek (Cu).

TSS and SPM chemistry provided information on short-term, process-related supply and dispersion. TSS and total water metal concentrations in bottom water indicated minor sediment resuspension, even under quiescent conditions in the shallow, southern parts of the bay, whereas

Table 6 Total water heavy metal concentrations

	Co	Cr	Cu	Ni	Pb	Zn
Quiescent conditions, *n* = 21						
Mean	0.08	1.8	**0.91**	0.29	1.79	**5.72**
Range	<0.04–0.25	0.59–0.48	**0.40–2.82**	<0.16–1.00	0.60–4.94	**3.48–13.86**
High-precipitation conditions, *n* = 20						
Mean	0.46	3.34	**3.72**	1.2	5.82	**15.28**
Range	0.12–**2.99**	1.03–**14.86**	**1.00–23.64**	0.27–6.32	1.85–34.97	**4.93–93.02**
High-wind/heavy rainfall conditions, *n* = 27						
Mean	0.36	3.61	**2.54**	0.77	5.01	**12.57**
Range	0.18–0.71	1.67–**9.99**	**1.30–6.41**	0.41–1.59	2.66–12.82	**7.11–25.76**
IWGs	0.91	9.0[a,b]; 3.1[c,d]	0.324	32.64	47.01	2.74

Concentrations in µg L^{-1}, Cd frequently below detection and omitted

Bold, above IWGs

IWGs Interim water guidelines (ANZECC/ARMCANZ 2000)

[a] Interim level; [b] C^{rIII}; [c] $Cr^{V!}$; [d] Level 1

surface water SPM chemistry suggested that some metals (Co, Cr, Ni and Zn) were being supplied to Homebush Bay from Parramatta River.

Higher TSS, turbidity and total water metal concentrations in surface water than bottom water, as well as large differences in SPM metal concentrations between surface water and bottom sediments demonstrated that stormwater discharge was the dominant process of metal transfer during the high-precipitation event. All metals studied were supplied to Homebush Bay from Parramatta River in surface water and Co, Cr, Pb and Zn were also introduced to the bay in bottom water during the high-rainfall event monitored. Minor resuspension was observed during these conditions in the shallow, southern part of the embayment.

Stratification was only partially preserved during the high-wind/heavy-rainfall event investigated and surface water was diluted by relatively clean water entering the bay from the north and south after several days of heavy rainfall. Higher TSS and total water metals in bottom water than in the surface water plume, indicated that resuspension of bottom sediment was a greater contributor of TSS to the water column than stormwater during this event, especially in the shallower regions of the bay.

Assessment of SPM metals provided information on source and dispersion of contaminants during individual events, which occur as part of a continuum of meteorical conditions and cannot be duplicated. This approach identifies real-time processes, e.g. resuspension and bottom and surface water transport that studies of surficial sediment can not provide. Nevertheless, studies of bottom sediment give useful and important long-term, integrated assessment of sources and dispersion, not possible using single, synoptic surveys. These approaches are complimentary and both should be employed to obtain a full understanding of short- and long-term processes involved in supply and accumulation of contaminants in aquatic environments. Integration of these two approaches indicated that a major contributor of SPM metals to the water column in the shallow southeast of Homebush Bay was resuspension of highly contaminant bottom sediments derived from industry on the adjacent Rhodes Peninsula. During the monitored precipitation period, Parramatta River provided more SPM metals to Homebush Bay than the rivers draining the local catchment probably due to the Parramatta catchment being substantially larger and more industrialised. This conclusion will have to be validated by more detailed temporal sampling to determine whether these distributions are not due to timing of rainfall events in different catchments.

Acknowledgments The authors thank Chris Golding for advice and David Hobson, Bronwyn Buttlen and Marco Olmos for assistance with GIS. Dave Mitchell assisted with fieldwork and equipment and special thanks to Tom Savage who assisted in the laboratory and in the field. Dr. John Hunt and Mr. Jim Maniord of Thiess Environmental are thanked for their support. Reviewers of this manuscript are thanked for their useful comments.

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