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Anthropogenic trace metal contamination of Port Kembla Harbour sediments

Abstract

Detailed geochemical investigation of the bottom sediments in Port Kembla Harbour using neutron activation (NAA) and X-ray-fluorescence (XRF) techniques has delineated areas severely impacted by anthropogenically-sourced trace elements including potentially toxic metals (Pb, Cu) and metalloids (As and Se). In the south western section of Port Kembla Outer Harbour, copper concentrations in excess of 6000 ppm (~30x ANZECC-ARMCANZ (2000) ISQG-high trigger value) are associated with significant (i.e. above ISQH-high) concentrations of lead, zinc and arsenic. The potential source of sediment contamination here is the Port Kembla Copper (formerly ERS and Southern Copper) smelter. Port Kembla Inner Harbour sediments are geochemically distinct from those in the Outer Harbour being generally characterised by lower gold, zinc, copper and lead values and below detection limit (5ppm) concentrations of selenium. Locally very high (20%wt) concentrations of iron, zinc (>2000ppm) and tin (1500ppm) are suggestive of inputs from BHP Steel. The depths to which sediment contamination extends are not firmly established but are at least 50cm in sections of the Outer Harbour. Trace element contaminants are generally concentrated in the finer (62.5µm) sediment fraction, however, significant contamination also is evident in the coarser (62.5-250µm) sediment fraction suggesting that at least some trace elements have an association with industrially-sourced particulate matter. Stable lead isotopic studies indicate the existence of and potential mixing of two major sources of anthropogenic lead in Port Kembla Harbour. Lead in the Outer Harbour has 206/204Pb of 17.5-17.8 (208/206Pb 2.13-2.09), close to that of recent Southern Copper emissions whereas Pb in the Inner Harbour has 206/204Pb of 18.0-18.6 (208/206Pb 2.09- 2.03) generally consistent with a steelworks source. Future management of this contaminated sediment will need to address issues including disturbance, that is the potential for acid generation from pyrite decomposition and appropriate methods of disposal.

Keywords

contamination, metal, trace, kembla, sediments, harbour, port, anthropogenic, GeoQuest

Disciplines

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ANTHROPOGENIC TRACE METAL CONTAMINATION OF PORT KEMBLA HARBOUR SEDIMENTS

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ABSTRACT

Detailed geochemical investigation of the bottom sediments in Port Kembla Harbour using neutron activation (NAA) and X-ray-fluorescence (XRF) techniques has delineated areas severely impacted by anthropogenically-sourced trace elements including potentially toxic metals (Pb, Cu) and metalloids (As and Se).

In the south western section of Port Kembla Outer Harbour, copper concentrations in excess of 6000 ppm (~30x ANZECC-ARMCANZ (2000) ISQG-high trigger value) are associated with significant (i.e. above ISQH-high) concentrations of lead, zinc and arsenic. The potential source of sediment contamination here is the Port Kembla Copper (formerly ERS and Southern Copper) smelter.

Port Kembla Inner Harbour sediments are geochemically distinct from those in the Outer Harbour being generally characterised by lower gold, zinc, copper and lead values and below detection limit (5ppm) concentrations of selenium. Locally very high (20%wt) concentrations of iron, zinc (>2000ppm) and tin (1500ppm) are suggestive of inputs from BHP Steel. The depths to which sediment contamination extends are not firmly established but are at least 50cm in sections of the Outer Harbour.

Trace element contaminants are generally concentrated in the finer (62.5µm) sediment fraction, however, significant contamination also is evident in the courser (62.5-250µm) sediment fraction suggesting that at least some trace elements have an association with industrially-sourced particulate matter.

Stable lead isotopic studies indicate the existence of and potential mixing of two major sources of anthropogenic lead in Port Kembla Harbour. Lead in the Outer Harbour has 206/204Pb of 17.5-17.8 (208/206Pb 2.13-2.09), close to that of recent Southern Copper emissions whereas Pb in the Inner Harbour has 206/204Pb of 18.0-18.6 (208/206Pb 2.09-2.03) generally consistent with a steelworks source.

Future management of this contaminated sediment will need to address issues including disturbance, that is the potential for acid generation from pyrite decomposition and appropriate methods of disposal.

INTRODUCTION

Port Kembla Harbour is a heavily industrialised, artificial harbour located 4 km south of Wollongong and approximately 70 km south of Sydney at latitude 34° 29' south and 150° 54' east (Figure 1). It consists of an Outer Harbour created in 1898 by the construction of two breakwaters (Jones *et al.*, 2003) and an Inner Harbour formed by the dredging of Tom Thumb Lagoon in 1960. The harbour is constructed in Permian volcanic and volcanogenic strata of the Broughton Formation although creeks draining into the harbour are in part underlain by the Illawarra Coal Measures.

The Inner Harbour, and its narrow (155m) connection (the 'cut') through the Outer Harbour to the open ocean, has been dredged to a maximum water depth of 16.3 m (He and

Morrison 2001) to allow the entry of bulk ore carriers. Water depths in the Outer Harbour range from 4-16 m (He and Morrison, 2001). Water discharges into the Harbour are principally from Allans Creek with a catchment of 23km² (He and Morrison, 2001) and the Town Drain. Natural flows into the harbour are accompanied by the discharge of 900 ML/day of saline cooling water from the BHP main drain (Jones *et al.*, 2003) and by hot water discharge from the Port Kembla Copper smelter via the Darcy Road Drain. These discharges can result in significant increase in water temperature (8°C above general harbour temperature).

The port has experienced a century of industrial expansion and diversification. At present, Port Kembla Harbour is fringed by heavy industries including iron and steel manufacturing (BHP Steel), copper smelting (Port Kembla Copper, formerly South Copper and ER&S Pty Ltd), fertilizer manufacturing and coal and grain handling facilities. All of these industries are potential contributors to sediment contamination within the harbour.

Previous studies within the precinct of the harbour have focussed on atmospheric contamination (Beavington, 1977; Crisp 1981,1982; Chiaradia *et al.*, 1997). Crisp (1981,1982) identified three major sources of atmospheric contamination adjacent to the harbour.

These were steel manufacture (Fe, Mn, Ni and minor Cu and Pb), coal firing (coke and organic-rich particulates) and copper smelting (Cu, Pb, Zn, Cd, Fe and As). Equivalent published studies on the levels of sediment contamination in the harbour are sparse.

This paper presents an account of trace element contamination of Port Kembla Harbour sediments and addresses the issue of future management options for the contaminated material.

METHODS

Details of the methods used in this study are presented in Chenhall *et al.* (1994), Chiaradia *et al.* (1997) and Jones *et al.* (2003).

RESULTS

General Physical and Physico-Chemical Sediment Characteristics

Sand-dominated (e.g. >50%wt sand) sediments are present in the northern third of the Outer Harbour and are developed as a thin fringe along the eastern breakwater. Sandy sediments are also developed in the western section of the Outer Harbour adjacent to the dredge spoil area (Figure 1) and adjacent to the Darcy Road Drain. Mud-dominated sediments characterize the south-central section of the Outer Harbour. Sandy sediments are present throughout much of the Inner Harbour although mud-dominated sediments are developed throughout the 'cut' and adjacent to the mouths of Allans Creek and the Town Drain (Figure 1). The thickness of sediment deposited in the harbour, established by seismic profiling ranges from 0m in the navigation channel to a maximum of 2m adjacent to the northern breakwater (Jones *et al.*, 2003)

Mud-dominated sediment cores are generally characterized by a dark grey colour although a thin (1-3mm) rusty brown surface veneer of oxidised sediment is usually present. These sediments are generally devoid of macroscopic shelly organisms, however, live *Polychaete* worms were extracted during sampling. Sediment pore water pH ranged between

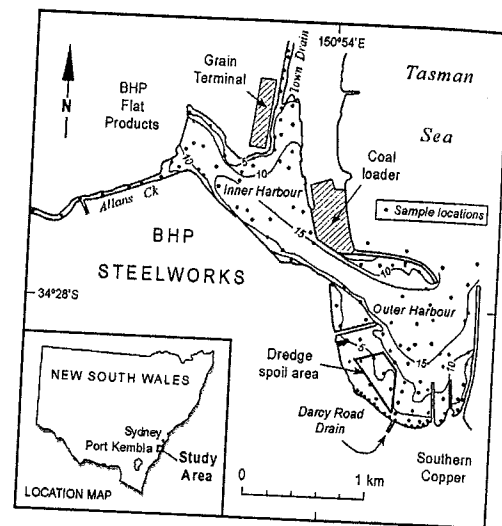


Figure 1 Location of Port Kembla Harbour

7.5 and 8.1 reflecting the marine-influenced nature of the sedimentary environment. Sediment underlying the oxidised surface veneer was reduced with redox potential showing a progressive decline to values of *circa* -450mV (relative to Ag/AgCl) at depths greater than 10cm below the water-sediment interface. These reducing conditions are consistent with the appearance of diagenetic pyrite at depths below 6cm in the sediment cores.

Binocular microscopy indicates the presence of anthropogenic components in the sediment, including coal, coke, pyrolytic carbon and pyrometallurgical ('kish') graphite, sinter, fly ash, electric arc furnace dust, iron oxide grains, slag and sulphide grains (Cu sulphides and diagenetic pyrite). Some of these contaminants are multi-sourced (e.g. coal from coal utilising/transporting sources) whereas others such as 'kish' and copper sulphide ore are uniquely sourced to steel manufacturing and copper refining respectively.

Bulk Near Surface Sediment Geochemistry

Figure 2 presents results for bulk sediment obtained by drop dredging. Nickel, Cu, Zn and Pb data were obtained by X-ray fluorescence spectrometry and the other elements analysed by neutron activation techniques. Neutron activation provides data for 31 elements. Many of these including REE, Rb, Th, Zr, K and Na are statistically strongly correlated with the finer sediment fractions in which they occur as natural components. Pre-industrial (natural or background) concentrations of trace element contaminants could not be determined from the surface sediments or from sediment cores which penetrated to depths of 60cm. Assessment of the degree of contamination is thus based on comparison with background concentrations from nearby Lake Illawarra (Table 1), the catchment of which contains geologically similar strata to those supplying sediment to the harbour.

Table 1. Background concentrations (ppm unless indicated) for elements from Lake Illawarra sediments and ANZECC&ARMCANZ (2000) Sediment Quality Guidelines (in mg/kg =ppm). ¹ Source: Payne *et al.* (1997); ² Present Study.

Element	Lake Illawarra ¹	Lake Illawarra ²	ANZECC&ARMCANZ (2000)	
			ISQG-low	ISQG-high
Fe	-	5.9 wt%	-	-
Co	-	20	-	-
Ni	-	-	21	52
Cu	33	37	65	270
Zn	68	84	200	410
Pb	17	18	50	220
Sb	-	0.7	2	25
As	-	15	20	70
Au	-	<5ppb	-	-
Se	-	-	-	-
Cr	-	-	80	370

Fe, Co and Ni (Figures. 2a, 2b and 2c)

Maximum concentrations of these elements occur in the southern section of the Outer Harbour adjacent to the Darcy Road Drain (Figure 1), however these elements generally display only moderate increase (2x-4x) above background. Iron concentrations of up to 20%wt are present in the western section of the Inner Harbour adjacent to the BHP-Steel bulk handling facility and probably originate from spillage and dispersal during steel manufacture. Nickel concentrations >50ppm are present at the mouth of Allans Creek.

Cu, Pb and Zn (Figures 2d, 2e and 2f)

From an environmental standpoint these metals represent the most significant contaminants in Port Kembla Harbour in terms of both their abundance and spatial distribution.

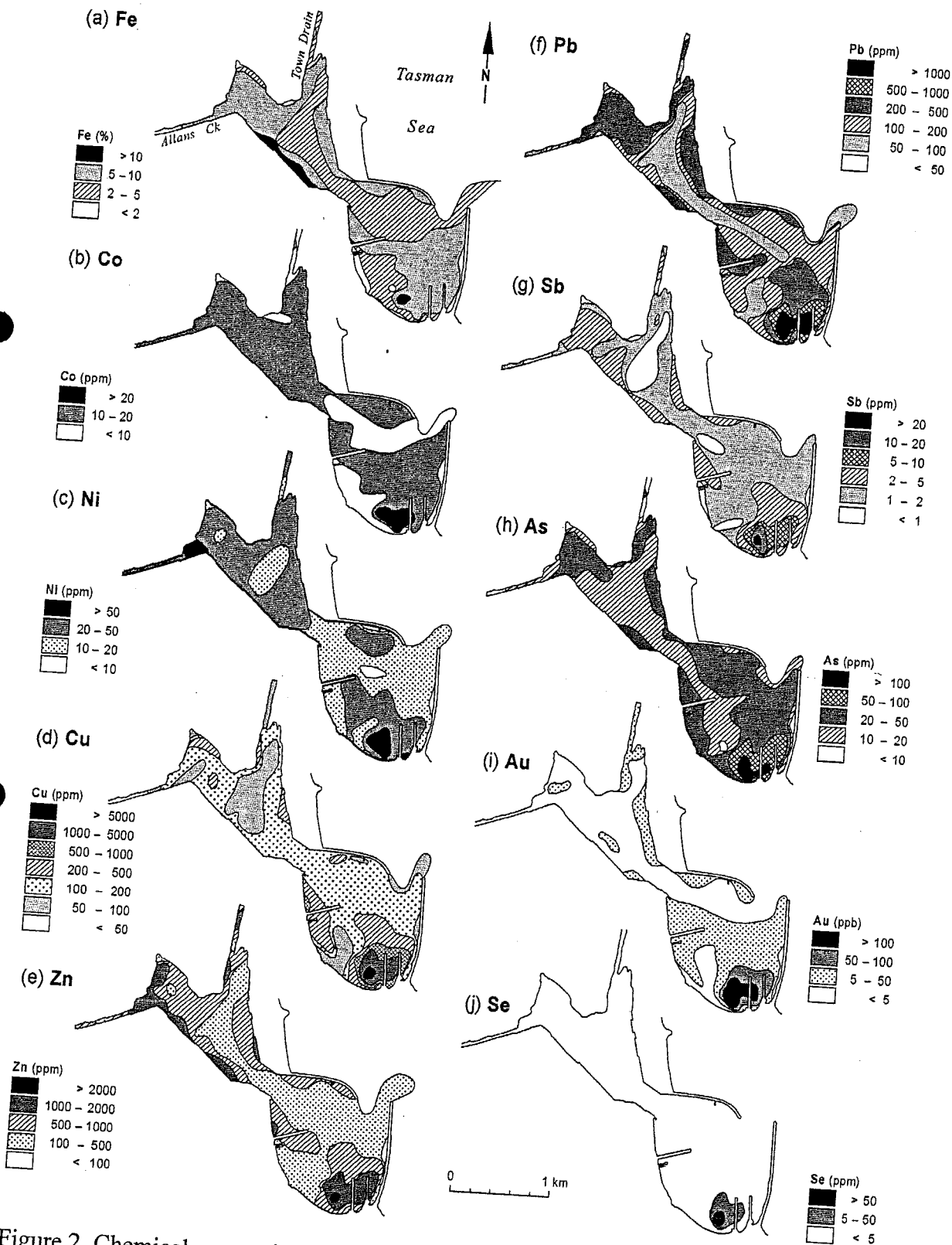


Figure 2. Chemical composition of near surface sediments from Port Kembla Harbour

ANTHROPOGENIC TRACE METAL CONTAMINATION

A concentration maximum of Cu, Zn and Pb (greater than 170x, 23x and 55x background respectively) is present in the Outer Harbour adjacent to the Darcy Road Drain. A contaminant halo (Figures 2d-2f) of these metals extends in an easterly direction from the drain towards the eastern breakwater. The northern portion of the Outer Harbour and most of the Inner Harbour are characterized by Cu concentrations 3x-6x background. Significant Zn contamination (>10x background) is present in the Inner Harbour adjacent to the mouth of Allans Creek and BHP bulk handling facility. Lead concentrations greater than 6x background are present in the northern section of the Outer Harbour and most of the Inner Harbour (apart from the navigation channel).

Sb, As, Au and Se (Figures 2g, 2h, 2i and 2j)

The distribution patterns of these elements are generally very similar with concentration maxima centralised in the Outer Harbour adjacent to the Darcy Road Drain. Antimony and As concentrations are raised above background values throughout Port Kembla Harbour. Selenium contamination is restricted to a 3km² area in the Outer Harbour. This distribution pattern may well be partly influenced by the relatively insensitive detection limit (5 ppm) for this element.

Sediment Cores

Geochemical analysis of sediment cores indicates that the harbour sediments are highly heterogeneous in three-dimensional space. Core S6C9 (Figure 3) representing bulk sediment from a site close to the Darcy Road Drain indicates that significant contamination of the sediments with Cu, Zn and Pb is by no means confined to the near surface layer since the contamination extends to the base of the core (48cm). Considerable depth-compositional variation (ranges given in brackets) is also demonstrated by Sb (12-52 ppm), Cr (240-396ppm), Au (164-616 ppb), As (140-298 ppm) and Se (31-135 ppm) in this core.

Concentration fluctuations as a function of depth may well represent temporal variation in pollution load, however concentration fluctuations may also be affected by sediment disturbance and diagenetic controls.

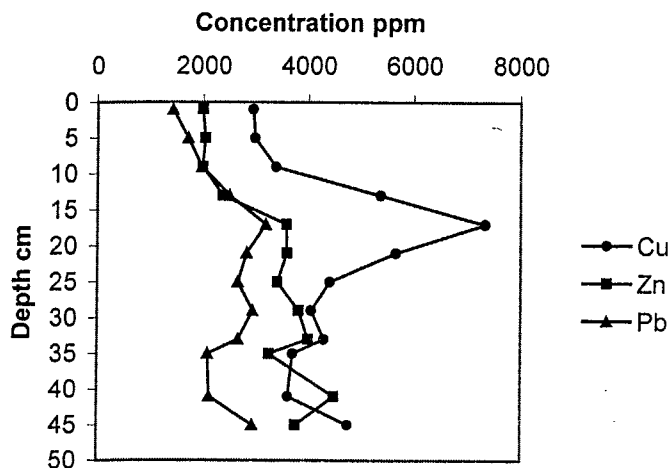


Figure 3. Bulk sediment composition of S6C9

Results for sieved sediment from core DRD1 are presented in Figure 4. The data presented in Figure 4 indicate that trace element contamination, whilst more pronounced in the <63µm sediment fraction is by no means restricted to this grain size range. Indeed the >63µm fraction contains substantial Zn, Cr, Sn and Au. Essential geochemical similarities and differences between the sediments of the Inner and Outer Harbours are well illustrated in Figure 5. Sediments from the Outer Harbour (Darcy Road Drain) are distinguished from those of the Inner Harbour (Allans Creek mouth) by their characteristic Au, Se As and Sb signature. Zinc, Cr and Sn are abundantly distributed at both sites, although a Sn-Cr signature characterises sediments from Allans Creek mouth.

Figure 4 Mean concentrations (n=9) of elements in the <math><63\mu\text{m}</math> and $>63\mu\text{m}$ sediment fractions from core DRD1 located in mud-dominated sediment near the Darcy Road Drain. Au concentration in ppb.

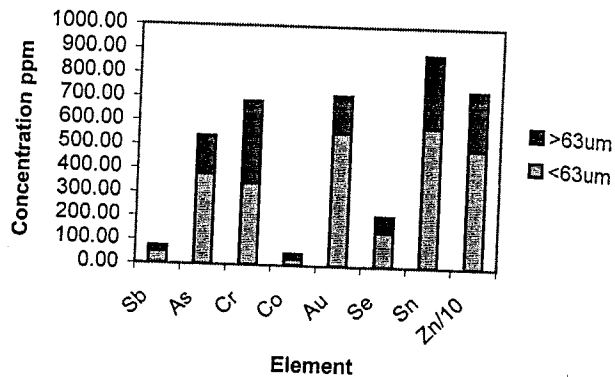


Figure 5. Geochemical characterization of sediments from the Inner and Outer Harbour at Port Kembla. Figure 5 depicts the mean concentration of nine 2cm sediment slices for the <math><63\mu\text{m}</math> sediment size fraction for core DRD1 (Outer Harbour) and core AC1 from the mouth of Allans Creek (Inner Harbour). Au in ppb.

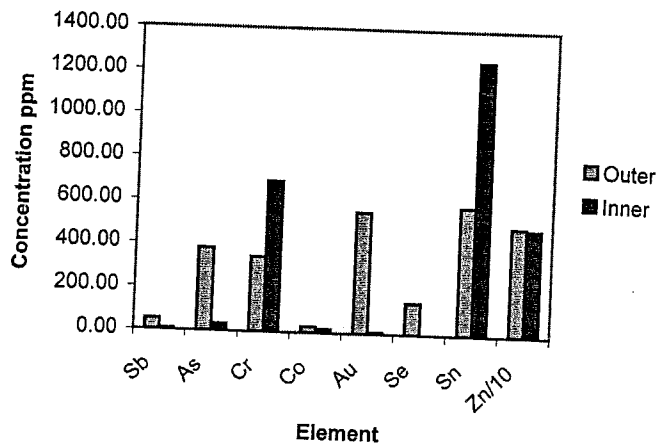
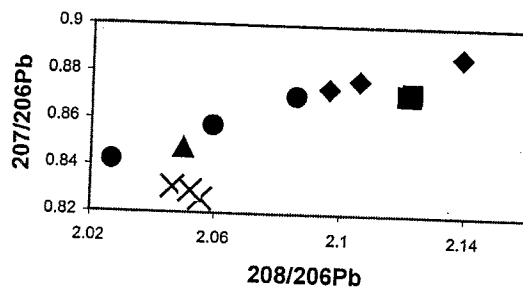


Figure 6 Pb isotopic composition of the sediments



Isotopic data presented in Figure 6 allow recognition of two major Pb sources in the harbour sediments. Sediments in the Outer Harbour (diamonds) plot near the isotopic composition of copper smelter dusts (solid squares) sampled in 1993, although the long-term isotopic compositional variation of Pb from this source is still being investigated. Two samples from the Inner Harbour (solid circles) plot near the isotopic compositions of Pb from Permian-sourced, pre-industrial sediments (crosses) from Lake Illawarra and Permian coal-utilising industries BHP Steel (triangle) and the defunct Tallawarra Power station. The linear distribution of harbour sediment 207/206Pb vs 208/206Pb is probably indicative of mixing of Pb sources.

DISCUSSION

Contamination Sources and Pathways

Data presented in this study indicate the presence of two major sources of sediment contamination in the harbour. Contamination from Port Kembla Copper (formerly ER&S Pty Ltd and Southern Copper) is most clearly reflected in sediments adjacent to the Darcy road Drain by elevated concentrations of a group of elements including Cu, Au, As, Pb and Se. With the exception of Au, all of these elements were recognised as copper smelter-sourced atmospheric contaminants by Crisp (1981,1982). Sediments from the Inner Harbour are geochemically distinct from those of the Outer Harbour, being characterised by generally lower Cu, Pb, As and low (commonly non-detectable) concentrations of Au and Se. Locally high Fe (Jones *et al.*, 2003) together with Zn, Sn and Cr (at the mouth of Allans Creek) strongly suggest an input from steel manufacturing. General support for these source appointments is provided by Jones *et al.* (2003) who analysed bag house dusts from Port Kembla Copper (high Cu, Zn, Pb, As, Se and Au) and blast furnace dusts (high Zn, low Cu, Pb, As and below detection Se and Au) from BHP Steel.

Recent analytical developments at Becquerel Laboratories have facilitated the analysis of Sn and although its poor detection limit (300ppm) may impede a comprehensive appraisal of the degree of Sn contamination, it is clear that Sn is a significant contaminant in the sediments. Potential sources of Sn could include the anti-fouling agent tributyltin and very likely, the steelworks-based tin plate mill. Similarly a thorough assessment of Cr contamination was precluded by the use of Cr steel crushing apparatus in the surface sediment study. Sediment core chromium concentrations adjacent to Allans Creek point to a generic steelworks source although the processing activity (e.g. Cr refractory usage) is at present unresolved. Other potential contributors to trace element loading include diverse sources (e.g. urban runoff, Cr-steel manufacture) located outside the industrial complex (Jones *et al.*, 2003).

In 1986 the State Pollution Control Commission estimated that 88 tonnes/day of fugitive particulate matter was generated during steel manufacture and another 11 tonnes/day from copper smelting (SPCC, 1986). Much of this settled on the harbour and adjacent sites and was subsequently transported into the harbour by surface runoff (Jones *et al.*, 2003). Wind-blown dusts, spillages and water-transported particulates from raw material stockpiles and contaminated soils adjacent to the industrial complex are also probable mechanisms of contamination. Allans Creek and the Town Drain contribute contaminants in the form of urban runoff and the latter drains an industrial waste site known to contain Cu, Pb and Zn contamination. Thus diverse pathways (modes of transportation and deposition) have probably contributed to sediment contamination in the harbour.

Trace Element Speciation and Retention

In many sedimentary environments, trace elements including heavy metals commonly show a strong preferential association with the clay and organic matter components of mud-sized (<62.5 μm) material (e.g. Förstner & Wittmann 1981). Many studies concerned with sediment contamination thus concentrate on the fine (<62.5 μm) sediment fraction (e.g. Birch and Davey 1995; Haynes *et al.*, 1995). A lack of strong positive correlation between the finer silt and clay-sized sediments and trace metals including Cu, Pb and Zn (Jones *et al.*, 2003), together with the sieved core sediment data (Figure 4) indicates that trace metals either are additionally associated with the coarser sediment components possibly in faecal pellets, or are concentrated in Fe oxyhydroxide coatings on sand sized particles or are affiliated with coarse diagenetic pyrite developed below 6cm depth in the sediments. However in this highly industrialized setting, a significant trace metal component may also reside in industrially-derived particulate matter present in the coarser (>63 μm) sediment fraction. This contention

is supported by the positive relationship between ash (industrial particulates) and trace metal contents of sediments established by (Chenhall *et al.*, 1994) for neighbouring Lake Illawarra sediments.

In these forms (and given the anoxic, alkaline nature of the bottom sediment) they are not likely to contribute greatly to the bioavailable metal concentrations in the water unless the bottom sediment is stirred up by dredging or shipping activity

Severity of Contamination

Trace metals and metalloids in Port Kembla Harbour can be compared to the Australian and New Zealand Sediment Quality Guidelines (ANZECC & ARMCANZ, 2000; Table 1, this study). Some elements (e.g. Se, Sn) have no appropriate guideline values and cannot be assessed. Most of the elements that are strongly associated with Cu (e.g. Zn, Pb, As) exceed both the ISQG-low and ISQG-high values, especially in the south eastern portion of the Outer Harbour, where maximum concentrations of Cu, Pb, Zn and As are 5~30x the ISQG-high value. According to ANZECC & ARMCANZ (2000) protocols the bulk of the sediment in Port Kembla Harbour should be the focus of future scientific investigation including bioavailability and biotoxicity testing and based on these findings possible intervention.

CONCLUSIONS CONCERNING FUTURE MANAGEMENT

Future management plans for Port Kembla Harbour will need to address the issues of the degree of sediment contamination, adverse impacts caused by sediment disturbance (e.g. dredging) and the acid generation potential of the sediment governed by the presence and abundance of diagenetic pyrite. Transfer of these contaminated sediments from an anoxic to oxic (land-based) environment would result in pyrite oxidation (sulphuric acid generation) and release of toxic metals and metalloids into the hydrosphere (Forstner and Kersten, 1988). Recent work (Muhammad, private communication, 2003) has indicated that up to 60kg of lime would be required to neutralise 1 tonne (wet weight) of sediment should oxidation of the sediment take place.

As indicated in Jones *et al.* (2003) Port Kembla Port Corporation has in the past (refer to Figure 1) utilised 'sub-sediment deposition' (Forstner, 1995) whereby the contaminated sediment was capped underwater with inert (non-contaminated) material. If future objectives are to dredge and deepen the harbour then this probably is a non-viable option and the 'inert cap' would need to be monitored in terms of recontamination (Jones *et al.*, 2003) and breaching by burrowing organisms.

Another option is to dispose of the dredged sediments at sea. The Port Corporation has exercised this option in the past. At present the Port Corporation is seeking permission to dredge and dump spoil from the Inner Harbour by this method of disposal. The outcome of this proposal is pending.

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