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Baseline

Richards Bay Harbour: Metal exposure monitoring over the last 34 years

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ABSTRACT

Richards Bay Harbour is South Africa's premier bulk cargo port. It was constructed in the Mhlathuze estuary in 1976 and over the past 34 years has become South Africa's most modern and largest cargo handling port. Although no official monitoring programme is in progress various studies by different groups have provided relevant data with respect to changing metal levels in brown mussel tissue (*Perna perna*) over the last 34 years. Eleven elements were analysed in brown mussels from the main channel in Richards Bay Harbour using ICP-MS. The results indicate that the metal concentrations in the mussel tissue remained relatively constant between 1974 and 2005. The mean metal concentrations increased significantly in 2005 possibly due to the construction of the new coal terminal and associated dredging activities. Mean metal concentrations in the 2008 sampling event were also elevated due to increased run off during an above average rainy season.

Metals enter the marine environment through a variety of point and diffuse sources (Kramer and Botterweg, 1994; He and Morrison, 2001). Metals are natural constituents of the marine environments which are generally found in low concentrations (Ansari et al., 2004). Increased concentrations in the marine environment could be derived from several major sources, most notably riverine influx (Forget et al., 2003), atmospheric deposition (Williams et al., 1998) and/or anthropogenic activities (Cheevaporn and Menasveta, 2003). As a result, the burden of metals is now a serious environmental concern (Franca et al., 2005). The focus of these concerns is primarily on estuaries and harbours due to their highly productive nature and their close proximity to the point sources of pollution (Kennish, 1997).

Measurements of metals by direct chemical analysis in water and sediment are limited in reliability (Bolognesi et al., 2004; Smolders et al., 2003). Consequently, after the initial suggestion by Goldberg (1975), many studies have utilised mussels to assess metal levels in the environment (Shulkina et al., 2003; Banaoui et al., 2004; LaBrecque et al., 2004). Mussels have been suggested to be the ideal bio-indicator organism in biomonitoring studies due to their sessile filter-feeding life style, coupled with their abilities to accumulate metals to much higher concentrations than those found in water and to not metabolise metals appreciably (Olivier et al., 2002).

Richards Bay Harbour is South Africa's premier bulk cargo port. It was constructed in the Mhlathuze estuary in 1976 and over the past 34 years has become South Africa's most modern and largest cargo handling port (Jury and Guyon, 2009). It also has the world's largest single coal terminal (Walmsley et al., 1998). Exports from the port include wood chips, aluminium, containers, coke (pitch, pet and metallurgical), steel (including scrap), granite, ferro alloys and fines, pig iron, and alusite, chrome, copper concentrate, rock phosphate, titanium slag, alumina, sulphur, coking coal, salt, fertiliser and loose bulk. Although no official monitoring programme is in place, studies by different groups have provided relevant data with respect to changing metal levels in brown mussel tissue (*Perna perna*) over the last 34 years. These include data from studies conducted before the construction of the harbour in 1974 (Hennig, 1985) and studies conducted between 1997 and 2010 (Wepener, 1997, unpublished data, 2000, unpublished data; Mills, 2005; Wepener, 2008; Degger, 2010).

Since Richards Bay Harbour is a relatively new harbour and data were collected on levels of metals in mussels from the original estuary that was converted into the harbour in 1976, it provides a unique opportunity to track any changes in metal contamination over time as a result of harbour developments and activities. Vermeulen and Wepener (1999) commented on the relatively unknown degree of contamination due to the lack in comparative data. The aim of this paper was therefore to relate the bioaccumulation changes in mussels from Richards Bay Harbour to the baseline concentrations measured pre-construction in 1974.

Historical metal bioaccumulation data for the Richards Bay-Lagoon (Mhlathuze Estuary), collected prior to the construction of the harbour were taken from Hennig (1985). The Hennig (1985) study made use of mussels that were collected from rocky outcrops in the mouth of the estuary. For the surveys between 1997 and 2009, brown mussels (*P. perna*) were sampled from the navigational buoys in the main shipping channel of Richards Bay Harbour (Fig. 1) during the pre-spawning period (February–April).

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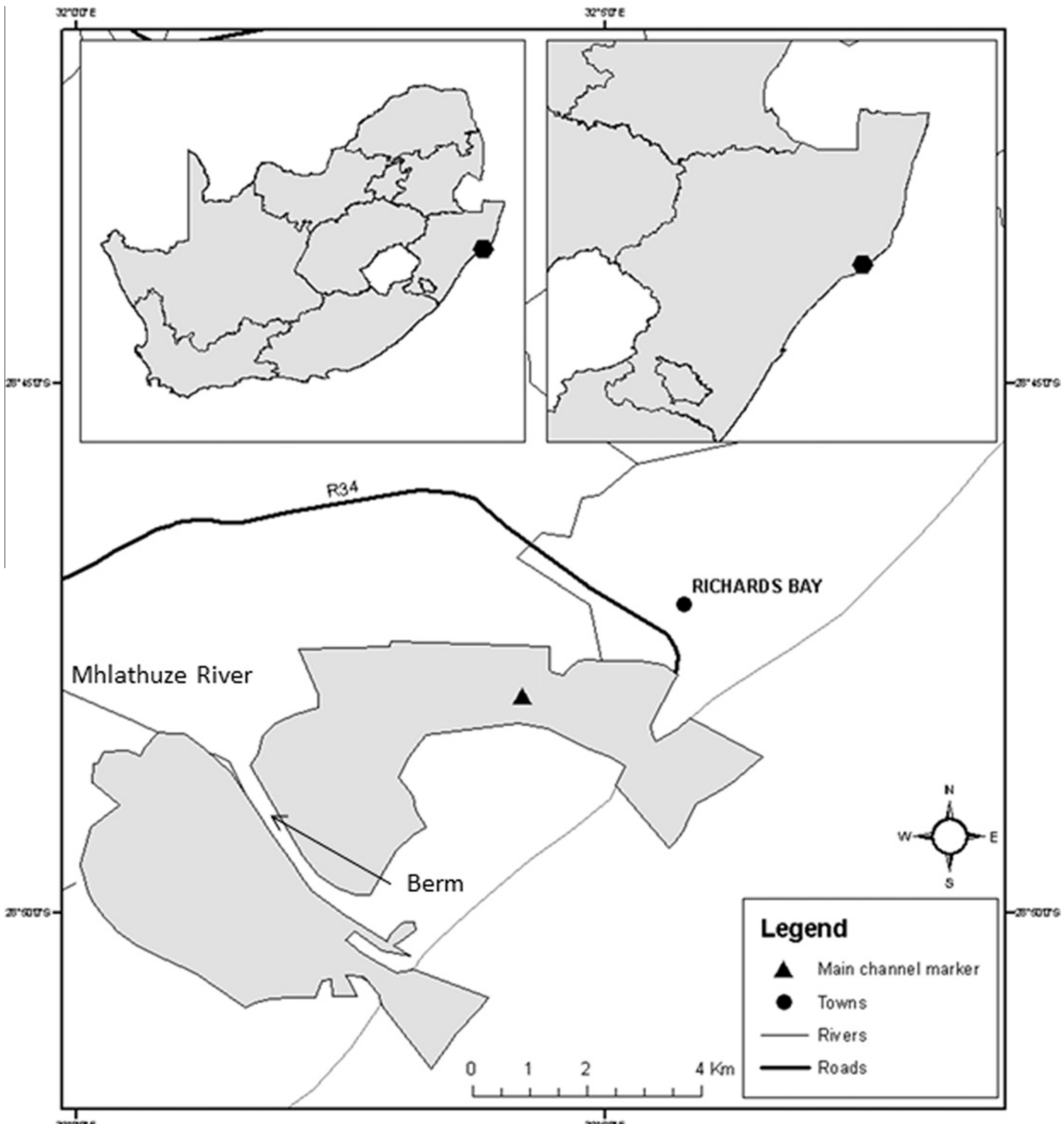

Fig. 1. Position of Richards Bay Harbour along the South African coastline indicating the main channel marker sampling site and the berm separating the Mhlathuze Estuary from Richards Bay Harbour.

These mussels were placed on ice and returned to the respective laboratories for analysis. Mussel tissue was prepared for analysis according to the methods set out by Blust et al. (1988).

In the laboratory the mussels were cleaned of epibionts and then shucked to remove the tissue from the shells. Whole mussel tissue samples were dried at 60 °C for 48 h and digested using HNO_3/H_2O_2 and microwave digestion. The digested samples were then diluted in 1% HNO_3 (AR) with Milli-Q water and metal concentrations were determined using a Thermo Inductive Coupled Optical Emission Spectrophotometer (ICP-OES) and an Inductive Coupled Plasma Mass Spectrophotometer (ICP-MS) in the case of the 1997, 2005, 2008 and 2009 samples. Metal concentrations in the 1974 samples were determined using Atomic Absorption Spectrophotometry (AAS). All samples were analysed for aluminium (Al), cadmium (Cd), cobalt (Co), chromium (Cr), copper (Cu), iron (Fe), manganese (Mn), nickel (Ni), lead (Pb), strontium (Sr), and zinc (Zn).

Indium was used as an internal standard to correct for interference from high-dissolved solids in the different matrices. Concentrations were expressed on a dry weight basis. Quality control of metal measurements in mussel tissue was verified by including process blanks and certified reference material (1997 and 2005 – CRM 278, mussel tissue Community Bureau of Reference, Geel, Belgium, and 2008 and 2009 – NRCDOLT3, Dog fish liver tissue, Community Bureau of Reference, Geel, Belgium). No certified reference material was used for quality control in the 1974 metal bioaccumulation study. All recoveries except the Zn and Cu in the NCRDOLT3 were within 10% of the certified value (Table 1). The Cu and Zn recoveries in the NCRDOLT3 tissue were within 20%. No correction factors were applied.

The significant variations between the metal concentrations in mussels collected between 1997 and 2009 from Richards Bay Harbour were tested by One-way ANOVA. Only the mean concentrations of metals in the 1974 mussels were available so no comparisons with the means of subsequent surveys were possible. Data were tested for normality and homogeneity of variance using Kolmogorov–Smirnoff and Levene's tests (Zar, 1996), respectively, prior to applying post hoc comparisons. Post hoc comparisons were made using the Scheffé test for homogeneous or Dunnett's-T3 test for non-homogenous data. The use of either one of the two tests resulted in the determination of significant differences ($p < 0.05$) between variables. The data were further subjected to Discriminant Function Analysis (DFA) and Principle Component Analysis (PCA).

Table 1
Metal concentrations reported in standard reference material and recoveries.

Element	BCR 278 mussel tissue (μg/g)[a]	Recovery (μg/g)	NRCDOLT3 Dogfish liver tissue (μg/g)[b]	Recovery (μg/g)
Cd	0.35 ± 0.01	0.36 ± 0.02	19.4 ± 0.7	18.73 ± 0.43
Cr	0.78 ± 0.06	0.76 ± 0.04		
Cu	9.45 ± 0.13	9.58 ± 0.12	31.2 ± 1.0	39.46 ± 2.92
Mn	7.69 ± 0.23	7.65 ± 0.13		
Pb	2.00 ± 0.04	2.0 ± 0.10	0.33 ± 0.046	0.338 ± 0.05
Zn	83.1 ± 1.7	79 ± 0.2	86.6 ± 2.4	75.11 ± 2.34

[a] Reference material used in the 1997 and 2005 study.
[b] Reference material used in the 2008 and 2009 study.

Where metal concentrations were below detection limits, 50% of the detection limit was included for the statistical analysis (United States Environmental Protection Agency (USEPA), 2000). Minimum values thus reported in data ranges will be minimum value recorded and not the 50% detection limits.

Clear temporal differences in metal bioaccumulation in brown mussels were observed. Aluminium, Co, Cr, and Ni had significantly higher metal concentrations ($p < 0.05$) during the 2005 sampling events when compared with the rest of the sampling events (Figs. 2 and 3). This is attributed to increased remobilization of these metals during dredging of the harbour substrates. The main dredging event took place in 2005 for the construction of the new bulk coal terminal. One of the environmental challenges facing the construction of the new coal terminal was the prevention of coal residue from the terminal discharging into the harbour. Steps were taken to upgrade the settling systems (Berry, 2009) however from the accumulation results it was evident that resuspension of sediments containing coal dust had occurred. According to Rosenberg (2009) metals such as Al, Co, Cr, Fe, Mn and Ni are associated with coal and coal sludge. According to Bocchetti et al. (2008) the remobilization of contaminants from sediments is a major risk during dredging activities. The dredging of contaminated sediments would dramatically change the physico-chemical properties of a water body and could induce the rapid mobility and bioavailability of heavy metals. The remobilization or release of these metals from the sediments could have adverse effects on aquatic organisms (Guerra et al., 2009).

Cadmium, Cu, Mn, Pb, Sr and Zn all exhibited the highest levels during the 2008 sampling event. These elevated levels of metals could be attributed to increased run off from the surrounding area into the harbour since rainfall recorded in 2008 was particularly high (between 200 mm and 500 mm) during the sampling period (Weather SA, 2010). The previous years' rainfall for the same period was on average between 107 mm and 109 mm.

Mean Al concentrations (Fig. 2) ranged between 186.5 μg/g in (2005) and 89 μg/g in (1997). The Al concentrations were all below the pre-harbour levels of 1974 of 568 μg/g. This would imply that the source of Al decreased due to the cessation of sediment

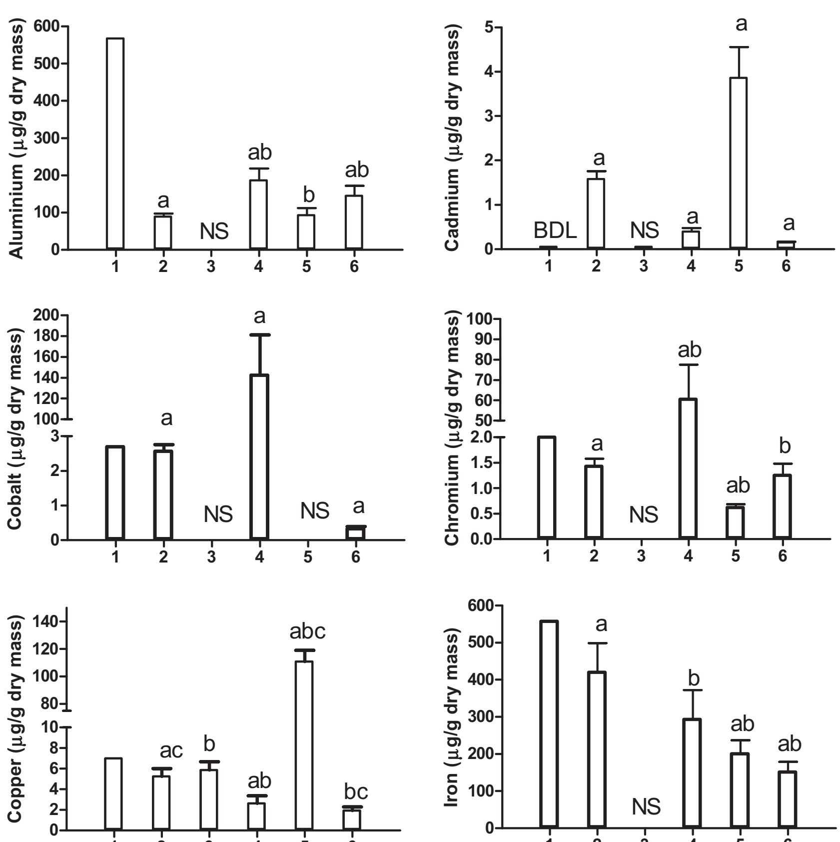

Fig. 2. Aluminium, cadmium, cobalt, chromium, copper and iron concentrations (mean + standard error in μg/g dry mass) in *Perna perna* sampled during 1 = 1974; 2 := 1997; 3 = 2000; 4 = 2005; 5 := 2008 and 6 := 2009. Means with common alphabetical subscript differ significantly ($p < 0.05$). No sample is represented by NS.

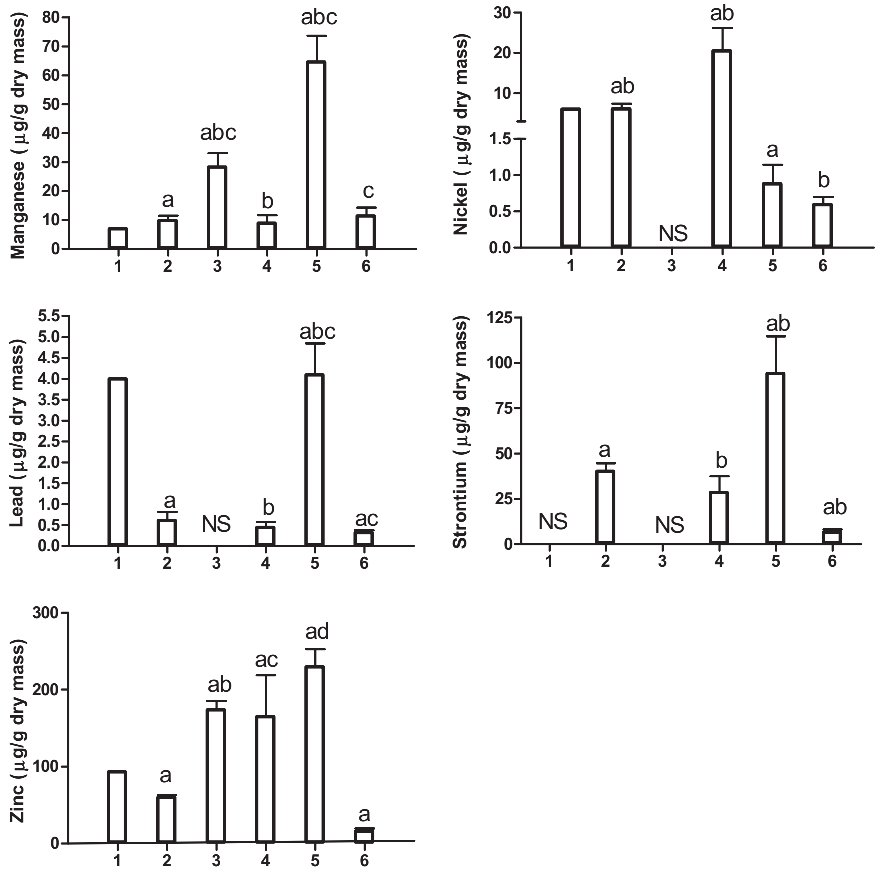

Fig. 3. Manganese, nickel, lead, strontium and zinc concentrations (mean + standard error in µg/g dry mass) in *Perna perna* sampled during 1 = 1974; 2 := 1997; 3 = 2000; 4 = 2005; 5 := 2008 and 6 := 2009. Means with common alphabetical subscript differ significantly ($p < 0.05$). No sample is represented by NS.

transport from the upper catchment into the system. With the construction of the harbour, the estuary was separated from the harbour by means of a berm (as depicted in Fig. 1), thus preventing any water from the catchment entering the harbour. This is clearly demonstrated in the lower levels of Al measured in whole mullet (*Liza dumerelii*) from Richards Bay Harbour (Vermeulen and Wepener, 1999) when compared the levels in the same species during the same period (April 1996) in the adjacent Mhlathuze Estuary (Mzimela et al., 2003). Mean Co concentrations (Fig. 2) ranged between 142 µg/g in (2005) and 0.33 µg/g in (2009). Cadmium levels (Fig. 2) ranged between 3.9 µg/g (2008) and 0.14 µg/g (2009). Mean Cr levels (Fig. 2) ranged between 60.6 µg/g (2005) and 0.62 µg/g (2008). The Mean Cr levels for the different sampling events were all within ±2 µg/g of the 1974 level of 2 µg/g except for the 2005 samples with the concentration being some 30 times greater. Mean Cu levels ranged between 110.8 µg/g (2008) and 1.9 µg/g (2009). Copper levels (Fig. 2) were all below the 1974 base line level except those of the 2008 sampling event. Mean Fe concentrations ranged between 420.2 µg/g (1998) and 151.1 µg/g (2009). The mean Fe concentrations (Fig. 2) were all below the 1974 pre-construction concentration of 557 µg/g. These lower levels in the harbour are similar to the findings for the Fe in whole mullet when they were compared to levels in whole fish from the adjacent Mhlathuze Estuary. This once again points to the river-borne sediments that are the main source of Fe. The mean Mn levels (Fig. 3) ranged between 64.7 µg/g (2008) and 8.9 µg/g (2005). Mean Ni concentrations (Fig. 3) ranged between 20.5 µg/g (2005) and 0.6 µg/g (2009). Mean Pb concentrations (Fig. 3) ranged between 4.1 µg/g (2005) and 0.3 µg/g (2009). Mean Sr levels (Fig. 3) ranged between 92.2 µg/g (2008) and 6.8 µg/g (2009). Mean Zn concentrations (Fig. 3) ranged between 229.7 µg/g (2008) and 16.1 µg/g (2009).

There are no known published data to allow for comparisons with other South African harbours and therefore we have compared the metal concentrations in brown mussel from Richards Bay Harbour with metals in mussels from known metal contaminated regions. Mean Cd, Mn, Pb and Zn levels in mussels during the 2008 sampling period were greater than those found by Liu and Kueh (2005) for *Perna viridus* in Victoria Harbour, Hong Kong that is known to be subjected to metal contamination. With the exception of Cu all the maximum concentrations measured in *P. perna* from Richards Bay Harbour were below the metal concentrations recorded in *Mytilus edulis* from the metal polluted Western Scheldt Estuary in The Netherlands (Mubiana et al., 2005). The majority of the metal concentrations in Richards Bay Harbour mussels were comparable to the levels recorded in *M. edulis* from the Eastern Scheldt Estuary (Mubiana et al., 2005).

The lowest metal concentrations were measured during the 2009 sampling event. We attributed this to decreased run off from the surrounding cargo terminal, smelter and coal terminals due to lower rainfall. Mean precipitation ranged between 10 mm and 25 mm for March 2009 as opposed to between 200 mm and 500 mm for the previous year (Weather SA, 2010). Since there is no riverine inflow into the harbour the only terrestrial input is through direct storm water runoff from the activities in the immediate vicinity of the harbour.

The mean metal concentrations were subjected to a DFA with the grouped centroids clearly indicating the difference in heavy metal concentrations between 1974 and the other sampling events (Fig. 4). The DFA indicates 97.4% of the similarity between the

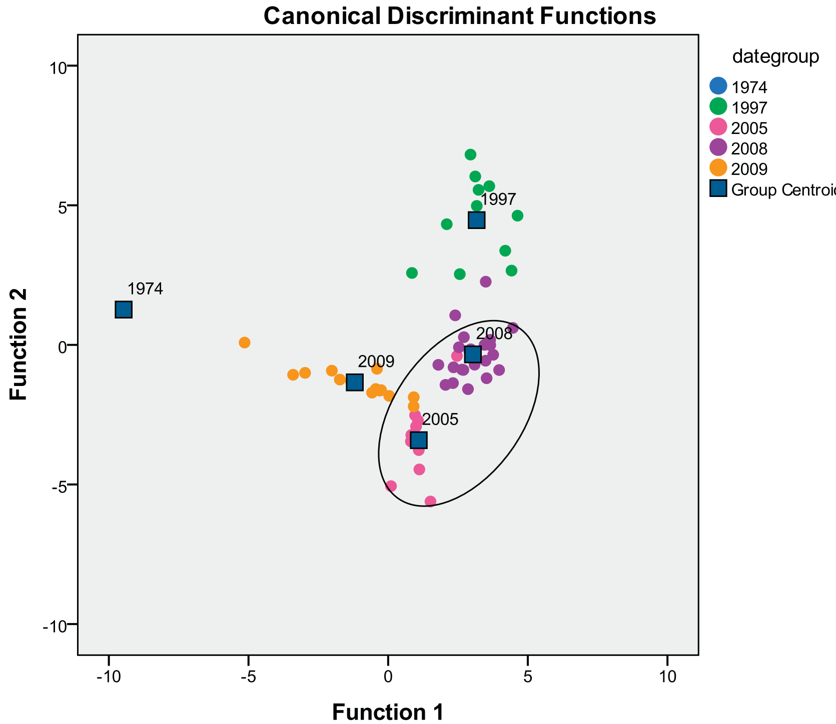

Fig. 4. Discriminant function analysis plot indicating 97.4% of the data from the first two functions.

different sampling periods in the first two functions and thus only these functions are discussed. Fig. 4 also indicates that mussels during the 1997 sampling have been affected by a different set of drivers thus placing them further away from the other sampling events and in a different quadrant being the in the (positive:positive) point of the plot. Although the 2009 centroid is closely positioned to the 2005 and 2008 centroids it is in a different quadrant and thus also is affected by a different set of drivers. This close grouping and situation of the 2005 and 2008 centroids indicates a relationship between the drivers affecting these sampling periods. This is most likely the result of similar metal exposures or bio-available fractions of the respective heavy metals. The same trend was observed by Bocchetti et al. (2008) with metal concentrations in caged *Mytilus galloprovincialis* decreasing after the cessation of dredging in the Piombino Harbour, Tuscany, Italy.

Superimposing the drivers (metal data) onto a PCA (Fig. 5) indicates that Ni, Co and Cr are the driving forces between the dissimilarity between the 1997 and 2005 sampling event and the rest of the sampling events. Aluminium, Fe and Zn to a lesser extent are also driving factors behind this difference, with the former two particularly evident in distinguishing the pre-harbour construction mussels from the rest of the surveys. These metals are all associated with metal bound to the natural alluvial derived sediments from the catchment that was predominant in the preconstruction mussel samples. The higher Al and Fe levels are also present in fish tissues from the adjacent Mhlathuze Estuary that receives the run off from the catchment. A number of metals, i.e. Al, Co, Cr, Fe, Mn and Ni are associated with run off from coal and coal sludge. This is a logical increase as the sediments around the new coal terminal were disturbed during the dredging for foundations. The disturbing

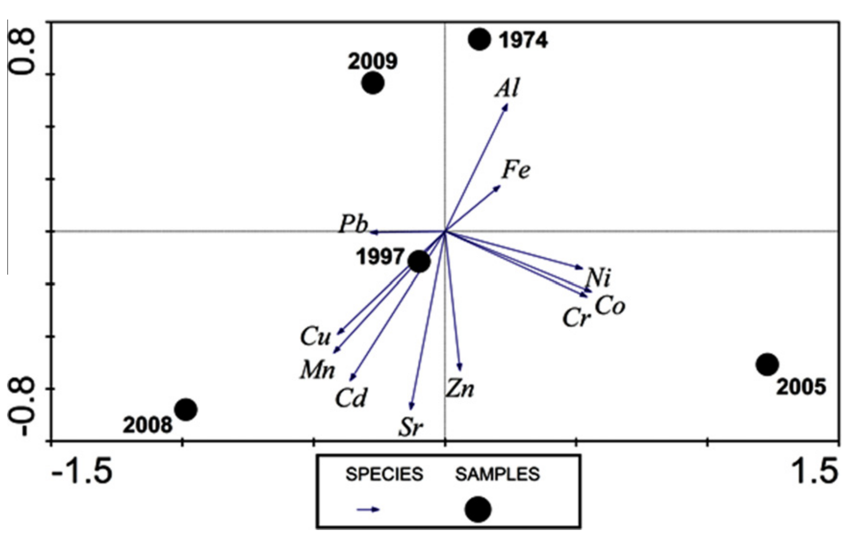

Fig. 5. PCA ordination of the different sampling events with the specific drivers superimposed onto it. Eigen values for the PCA indicate that 83.6% of the variance is indicated on the first two axes if the PCA.

of the sediments would remobilise any coal residues that would have settled out, thus remobilising the specific metals. The Cu, Mn, Cd and Sr concentrations were responsible for the dissimilarity between the 2008 and to a lesser extent 2005 and the other sampling periods. These metals are associated with increased run off from the industrial regions within the harbour as a result of increased precipitation during the 2008 wet season. This would have resulted in increased concentrations of these metals in the water and subsequently the mussel tissue. The position of these metals in the PCA corresponds with the data represented in the DFA (Fig. 4).

The results from this study clearly show that the activities associated with Richards Bay Harbour have a minimal effect on the metal inputs and subsequent biological exposure in the harbour. Although the mean metal concentrations have changed from the relative reference condition in 1974, the longer term monitoring results indicate that the increased metal exposure are associated with particular events such as dredging and run off due to increased levels of coal sediments and sludge, but that the concentrations decrease once the activity ends. The results indicate that although Richards Bay Harbour has grown to be South Africa's largest and busiest port, the activities do not appear to have much effect on the heavy metal accumulation of mussels in the main channel as the results of the 2009 sampling survey would indicate.

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