THE EFFECT OF PARTICLE SIZES ON METAL ACCUMULATION IN THE MARINE SEDIMENTS OF CAPE TOWN HARBOUR

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Abstract: This study was carried out to assess the elemental concentration in sediment samples collected from six locations in the Cape Town Harbour. The effect of size variation was considered determinant on metals accumulated in sediments. An Inductively coupled plasma-Mass spectrometry (ICP-MS) instrument was used for the analysis. The following metals were investigated Mn, Cr, V, Ni, Co, and As. The order of heavy metal concentration in sediments was Mn > Cr > V > Ni > Co > As. Strong correlation was found between these metals (Mn and V in sizes <75 and ≥150), (As and Co in size ≥150), (Ni and Cr in size ≥150 and <75).

Key Words: Sediments; marine; harbor; modeling; correlation; residual plots; particle- size.

INTRODUCTION

One of the most important factors found, to date, about metal accumulation in sediment is size, this is mostly because it is usually related to weight. The specific surface area of sediments is independent on granulometric parameters and mineral composition. (Juraic, et al. 1980; Fatoki et al. 2012). Because of the increasing amounts of toxic metals from anthropogenic sources in the sedimentary environment. Various means have been devised for determining their effects on marine organisms. Fine particles have high specific areas that retain high amounts of metals (Wang et al. 2006). Also, small particles are often soluble and metals are more likely to traverse the gastric mucosa and be more easily absorbed in human tissues than for coarse fractions (Lin et al. 1998). It is generally believed that metals are associated with smaller grain-size particles (Martincic, et al., 1990). This trend is predominantly attributed to sorption, co-precipitation and complexing of metals on particle surfaces and coating. Smaller particle sizes have a larger surface to volume ratio and therefore contain higher concentration of metals. The specific surface area of sediments is independent on granulometric parameters and mineral composition (Juraic, et al., 1980). Because of the increasing amounts of toxic metals from anthropogenic sources in the sedimentary environment.

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The geochemical cycle of trace metals in the marine environment is determined by various mechanisms including the interaction of the metals with sediments. One of the most significant parameters influencing trace metal content in a sediment fraction is sediment size dimension. Therefore, a comparison of the contamination of the marine environment by anthropogenic trace metals in various geographic locations should be based on the standardization of several parameters, among them sediment size. Often analyses were conducted on a certain size fraction to correct natural variability (Solomons and Solomons 1980). However, Windom et al. (1989) considered that such a procedure can often lead to confusing results, since contaminant concentrations in a certain size fraction do not necessarily reflect its abundance in the total sediment. The generally accepted opinion is that the smaller the size of the sediment fraction, the larger the amount of trace metals bound to this fraction, and that trace elements are mainly present in the clay/silt particles with grain size less than 0.063 mm (Forstner et al. 1982). In general, this is because of the increase in specific surface area of the smaller fractions and to the surface properties of clay minerals (Forstner et al. 1982).

FIG. 1. Map indicating the sampling points and the position of Cape Town Harbour along the Atlantic Ocean and Indian Ocean.

Comparison of heavy metals distribution with grain size allows the identification of the area where the sediment fine fraction appears affected by heavy metal anthropogenic contamination (Leoni and Sartori 1996). Different results which reported that finer particles are associated with higher concentrations of heavy metals have, however, been reported by other investigators. The findings by Moore et al. 1989 and Combest (1991) demonstrated that metal concentrations did not increase with decreasing particle size, and the highest heavy metal concentrations were in the coarse silt and larger

size fractions. Reaction to the uptake of metals has obvious, toxic and chronic responses. Levels of metals resulting in chronic responses are difficult to detect or assess. These levels are of importance, since prolonged chronic levels or synergistic effect of various metals and/or other toxicants can result in undue stress on the organisms. The objective of this study is to evaluate the effect of particle size on the distribution of heavy metals in the Cape Town Harbour sediments. The total metal concentration was analyzed using inductively coupled plasma-Mass spectrometry (ICP-MS).

FIG. 2. Map indicating the sampling points and the position of Cape Town Harbour along the Atlantic Ocean and Indian Ocean.

MATERIALS AND METHODS

Study Area

Cape Town competes with Alexandria harbour as the most famous port in Africa (Figs. 1 and 2). In fact, the harbour is one of the most beautiful harbours in the world that has a magnificent backdrop of Table Mountain framed by the mountainous Peninsula. The port is situated on one of the world's busiest trade routes and will always retain strategic and economic importance for that reason alone. Cape Town is also a busy container port, second in South Africa only to Durban, and handles the largest amount of fresh fruits. Fishing has a significant place in the economic activity of the port. The emerging oil industry in West Africa has also become a significant factor because one vessels visit the harbour for repair and maintenance facilities.

Instrumentation

Samples were analyzed with an Agilent 7,700 ICP-MS. The Agilent 7,700 instrument used with a Meinhardt nebulizer and silica cyclonic with continuous nebulisation. The operation parameters were: Plasma RF power: 1,550 W; sample depth: 8.0 mm; Carrier gas: 1.08 L/min; Nebulizer pump: 0.10 rps; Helium gas: 5.3 mL/min.

Sampling Procedure

Six sampling stations were chosen for the sediment sampling (Fig. 2). Sediment samples were collected with the aid of sample Boat Waveride DTC 787C (6.3 m stringray cat hull) supplied by Stringray Marine was powered by Suzuki 90 hp's 4 stroke engines and equipped with Van Veen Grab sampler. A Garmin GPS was used to get the sampling coordinates. The sediment samples were placed in plastic poly Zip block bag in an ice chest and transported to the laboratory. Three replicates were taken from each station.

Sample Preparation

Sediment samples were dried in the oven overnight at 105°C, until they were sufficiently dry to handle. A portion of each sample from the respective sampling sites was sieved

Table 1. Quality Control Assessment.

Analytes	Certified Value	Measured Value	% Recovery
V	9.67	9.33	96.48
Cr	9.73	9.67	99.43
Mn	9.67	9.25	95.61
Co	9.67	9.30	96.22
Ni	9.67	9.52	98.43
As	96.71	96.70	99.99

Table 2. Physiochemical parameters measured in the sampling sites.

Description	Sampling Site	Parameters		
		pH	Conductivity (mS)	TDS (ppm)
Duncan Dock	1	7.59	8.42	5,740
Duncan Dock	2	7.1	2.4	2,710
Inside Sea 500 m away from Point 4	3	7.5	3.19	2,160
Duncan Dock	4	7.25	8.32	5,680
Robinson DRY Dock	5	7.86	2.07	1,420
Synchrolift	6	7.77	7.85	5,330

into 3 sub-samples with grain-sizes of <75 , ≥ 75 and ≥ 150 micro-meters. This was performed by a dry sieving method, using a mechanical shaker sieve set. For the analysis of the sediment physico-chemical parameter, 10 g of dried sediment was mixed with 50 ml of distilled water and the pH, conductivity and total dissolved solids (TDS) were determined. A triplicate 0.5- g sediment sample was digested with aqua-regia (7.5 ml HCL and 2.5 ml HNO_3) in a micro-wave unit and heated using the following heating program:

250 W for 6 min, 400 W for 6 min, 600 W for 6 min and the vent time used was 5 min and the total digestion process was 24 min. The digested residues were filtered to get a clear solution of which 10 ml was then diluted to 100 ml using Milli Q water. The obtained solutions were analyzed for the following metals: Mn, Cr, V, Ni, Co and As by ICP-MS. The measurements were programmed and adjusted to analyze the samples in triplicates while blank samples were analyzed after 10 measurements.

FIG. 3a. Variation of particle size with pH.
FIG. 3b. Variation of particle pH with particle sizes.

Table 3. Concentrations of some selected heavy metals in different size fractions in sediments of the Cape Town Harbour.

		Heavy metal concentration in ppb ($\mu\text{g/g}$)					
Sample		Vanadium	Chromium	Manganese	Cobalt	Nickel	Arsenic
Location	Grain size	Mean $\pm$ SE	Mean $\pm$ SE	Mean $\pm$ SE	Mean $\pm$ SE	Mean $\pm$ SE	Mean $\pm$ SE
1	<75	233.78 $\pm$ 2.19	371 $\pm$ 8.32	1,176 $\pm$ 4.62	41 $\pm$ 0.68	149 $\pm$ 1.69	58 $\pm$ 0.66
	$\geq$ 75	183 $\pm$ 2.19	362 $\pm$ 2.39	951 $\pm$ 4.97	38 $\pm$ 0.19	142 $\pm$ 2.15	54 $\pm$ 0.58
	$\geq$ 150	219 $\pm$ 1.17	365 $\pm$ 7.18	1,087 $\pm$ 1.09	39 $\pm$ 0.38	136 $\pm$ 0.76	59 $\pm$ 0.76
2	<75	202 $\pm$ 4.99	323 $\pm$ 4.97	888 $\pm$ 2.72	38 $\pm$ 0.76	127 $\pm$ 2.84	60 $\pm$ 0.97
	$\geq$ 75	224 $\pm$ 2.49	409 $\pm$ 6.36	1,075 $\pm$ 2.29	42 $\pm$ 0.52	140 $\pm$ 2.36	66 $\pm$ 0.48
	$\geq$ 150	189 $\pm$ 2.49	324 $\pm$ 5.85	774 $\pm$ 1.59	45 $\pm$ 0.74	114 $\pm$ 1.46	54 $\pm$ 0.74
3	<75	84 $\pm$ 0.70	290 $\pm$ 6.86	584 $\pm$ 3.88	25 $\pm$ 0.23	134 $\pm$ 1.57	28 $\pm$ 0.18
	$\geq$ 75	50 $\pm$ 0.39	178 $\pm$ 3.97	313 $\pm$ 1.94	15 $\pm$ 0.54	58 $\pm$ 0.81	12 $\pm$ 0.11
	$\geq$ 150	46 $\pm$ 0.49	191 $\pm$ 2.99	306 $\pm$ 1.34	11 $\pm$ 0.07	58 $\pm$ 0.30	12 $\pm$ 0.19
4	<75	217 $\pm$ 2.27	327 $\pm$ 5.44	1,579 $\pm$ 4.18	45 $\pm$ 0.41	142 $\pm$ 1.44	68 $\pm$ 0.66
	$\geq$ 75	175 $\pm$ 3.44	277 $\pm$ 3.45	1,108 $\pm$ 8.01	33 $\pm$ 1.76	114 $\pm$ 2.10	54 $\pm$ 0.90
	$\geq$ 150	234 $\pm$ 3.42	336 $\pm$ 2.46	1,407 $\pm$ 1.79	39 $\pm$ 0.64	130 $\pm$ 0.89	64 $\pm$ 0.83
5	<75	285 $\pm$ 2.29	547 $\pm$ 1.33	1,038 $\pm$ 2.26	594 $\pm$ 4.67	189 $\pm$ 1.33	181 $\pm$ 2.07
	$\geq$ 75	267 $\pm$ 2.19	709 $\pm$ 6.05	1,026 $\pm$ 6.12	163 $\pm$ 5.72	240 $\pm$ 2.20	142 $\pm$ 1.25
	$\geq$ 150	248 $\pm$ 3.76	938 $\pm$ 6.41	1,108 $\pm$ 2.20	330 $\pm$ 4.70	289 $\pm$ 4.58	131 $\pm$ 2.37
6	<75	314 $\pm$ 5.86	693 $\pm$ 0.40	1,092 $\pm$ 2.89	102 $\pm$ 1.54	282 $\pm$ 2.98	150 $\pm$ 1.61
	$\geq$ 75	187 $\pm$ 1.34	927 $\pm$ 8.01	1,111 $\pm$ 2.32	228 $\pm$ 0.14	542 $\pm$ 5.93	132 $\pm$ 0.96
	$\geq$ 150	320 $\pm$ 1.78	1,214 $\pm$ 7.93	1,200 $\pm$ 1.94	202 $\pm$ 4.29	521 $\pm$ 3.73	168 $\pm$ 1.15

Quality Assurance

For quantification of the element of interest, the ICP-MS were calibrated daily using NIST traceable standards. A quality control standard was analyzed prior to the samples to verify the accuracy of the calibration standards, while control standards were used throughout the analysis to monitor accuracy and instrument drift. Internal standards for ICP-MS were introduced continuously with the samples along with standards to correct for drift because of high matrix load. Data

acquisition and processing was software controlled and exported in Excel format.

Statistical Analyses

The results were statistically analysed using SAS 9 software (Cary, NC, U.S.). Pearson's correlation was applied to evaluate the relationships between the variables and a correlation coefficient with $p \leq 0.05$, regarded as significant. Principal Component Analysis (PCA), used to analyse the data.

FIG. 4. Average metal concentration across locations.

FIG. 5. Average metal concentration in selected grain sizes in location 1.

Result and Discussion

Heavy metal percent recoveries from a sediment certified standard reference material were calculated for all the metals. The high percentage recoveries ranged from 95.61 to 99.99%. This was within acceptable limits for all elements examined and showed the accuracy of the digestion procedure (Table 1).

Physicochemical Characteristics of the Sediments

The sediment pH ranged from 7.10 to 7.86. Sediment samples located close to the estuary were more alkaline, because of infusion of salt water from the sea during high tide and may also be because of the presence of

carbonates of calcium and magnesium (Table 2, Fig. 3a, b) and the relationship between the pH and the particle sizes for each metal investigated.

Assume that the power relation of Wang and Fisher (1997) that body weights to metal bioaccumulation will also hold for body size versus accumulation

$$BCF = aw^b$$

$$\text{Let } S = BCF = aw^b$$

$$\text{Log}(S) = \text{Log}(a) + b\text{Log}(W)$$

b can be calculated from the above equation as the slope of the Log-log relationship between the BCF (Bioconcentration factor) and W (dry weight). Plotting S

FIG. 6. Average metal concentration in selected grain sizes in location 2.

FIG. 7. Average metal concentration in selected grain sizes in location 3.

(Bioconcentration factor) versus average size show how they vary with pH on Log Log graph (Fig. 3a–3f), for b value >0 the metal accumulation increases with size (+ve), while a b value <0 the metal accumulation decreases with size (–ve), and when the b value is close to 0, this indicates that the metal accumulation is relatively independent of particle size. The sediment conductivity ranged from 2.07 to 8.42 mS and the total dissolved solutes ranged from 1,420 to 5,740 ppm. The highest conductivity and TDS values (Table 2) were found in locations 1, 4 and 6, this may be attributed to various industrial activities taking place in these locations. Many studies have been carried out on heavy

metal distribution, and some authors have shown the effect of particle size on heavy metal distribution and have indicated that concentrations of metals in sediments increase with decreasing particle size. Chromium concentration in this study ranged from 219.8 to 944 ppb, with the lowest chromium concentration recorded in location 3 and the highest being in location 6 (Table 3). This high Cr concentration may be attributed to ship repairs and other industrial activities taking place at this location. Ni concentration ranged from 83.71 to 448.3 ppb, with the lowest Ni concentration being location 3 and the highest being location 6. This is because the municipal wastewater as this is the water

FIG. 8. Average metal concentration in selected grain sizes in location 4.

FIG. 9. Average metal concentration in selected grain sizes in location 5.

from sewage treatment plants. Vanadium concentration ranged from 60.3 to 274 ppb, the lowest V concentration being location 3 and the highest being in location 6. Vanadium contamination of soil may occur whenever petroleum or coal bi-products are accidentally discharged into the sediment environment.

Mn concentration ranged from 401.1 to 1,364 ppb, with the lowest Mn concentration being location 3 and the highest location 6. This may be attributed to shipments because potassium permanganate is a potent oxidizer and is used as disinfectant. Co concentration ranged from 17.12 to 362.3 ppb, the lowest Co concentration was recorded in location 3 and the highest being location 5. This may be because of the painting of

ships, because cobalt is used as a dying agent for paints. Arsenic concentration ranged 17.33 to 151.3 ppb, with the lowest As concentration being location 3 and the highest being location 5, this may be attributed to the lead and zinc production used in agriculture. Highest concentrations of these elements were found in the areas that experience intense traffic conditions. The Major “hot-spot” included Site 6 at Cape Town Harbour (grossly contaminated with Cr, Co and Ni). The least contaminated site was location 3. This was expected because this location was inside the sea. The order of heavy metal concentration in sediments was $Mn > Cr > V > Ni > Co > As$. The presence of elevated levels of Mn and Cr in almost all locations (Fig. 4) and the

FIG. 10. Average metal concentration in selected grain sizes in location 6.

FIG. 11. PCA showing distribution of the metals across locations.

increases potential for human exposure to heavy metals are serious matters of concern.

It is generally believed that metals are associated with smaller grain-size particles, (Whitney 1975; Gibbs 1977 and Martincic et al., 1990). This trend (Figs. 5–10) is followed by all analytes in location 1 except Mn. In location 5 this trend is followed by V, Co and As while chromium, manganese, and nickel reveal the opposite. This trend in location 6 is not followed by any element. This trend in location 5 is only followed by chromium and nickel, while vanadium, arsenic reveals the opposite and manganese and cobalt showing no trend. In location 6, this trend is only followed by chromium and manganese, while other elements reveal no trend as shown in Figs. 5–10.

FIG. 12. PCA showing distribution of the metals in different sizes.

FIG. 13. graph of Eigen value vs. Component.

Statistical Analysis

The PCA as shown in Figs. 11 and 12 was used to group the elements according to how they accumulate in different locations and sizes. Strong correlation was found between these metals (Mn and V in sizes <75 and ≥ 150), (As and Co in size ≥ 150), (Ni and Cr in size ≥ 150 and <75) respectively. The strong correlation that was found confirmed that their contribution was from anthropogenic sources, i.e., industrial activities, storm and urban inflow, agricultural activities, and ship reparation that occur within and around the harbour. Component variations in heavy metal from eigenvalues are shown in Fig. 13. The eigenvalues decrease in this order $Mn > Cr > V > Ni > Co > As$ respectively. Ni, Co and As have very low eigenvalues. They can be said to be of natural origin while Mn, Cr and V are of anthropogenic sources because of their high level of component variation.

Pearson correlation coefficients for heavy metals in sediments collected from Cape Town Harbour is presented in Table 4. High correlation coefficients ($r > 0.8$) were calculated between (V and As); (Cr and Ni); (Cr and As) and (Co and As) respectively.

Plot of the residuals versus predicted Y (Fig. 14). The pattern shown indicates no problems because of the assumption that the residuals are normally distributed

Table 4. Pearson correlation coefficients between metal levels in sediment by ICP-MS.

	V	Cr	Mn	Co	Ni	As
V	1.00000	0.64936	0.76416	0.48935	0.51652	0.82518
Cr		<0.0001	<0.0001	0.0002	<0.0001	<0.0001
Mn		1.00000	0.37634	0.57221	0.93182	0.84624
Co			0.0050	<0.0001	<0.0001	<0.0001
Ni			1.00000	0.20873	0.36098	0.48503
As				0.1298	0.0073	0.0002
				1.00000	0.45953	0.80127
					0.0005	<0.0001
					1.00000	0.74655
						<0.0001
						1.00000

KEY: Upper value = correlation coefficient r.

Lower value = Significance level $p \leq 0.05$.

at each level of Y and constant in variance across levels of Y. SPSS does not automatically draw in the regression line (the horizontal line at residual = 0). A similar trend was observed for all the metals as shown in Fig. 14 for Arsenic. There is no variation for the residual plots drawn for each metals proving that the mean is closer to the residual and the data were in alignment.

CONCLUSION

The metal data for different size particles of the sediment samples collected from the different locations along the Cape Town Harbour suggested that the distribution of heavy metals in sediments had no obvious trend in terms of heavy metal concentration. Highest concentrations of these elements were found in the areas that experience intense traffic conditions. The

Model: Intercept + pH + Conductivity + TDS + Part_size + Location + pH * Conductivity + pH * TDS + pH * Part_size + pH * Location + Conductivity * TDS + Conductivity * Part_size + Conductivity * Location + TDS * Part_size + TDS * Location + Part_size * Location + pH * Conductivity * TDS + pH * Conductivity * Part_size + pH * Conductivity * Location + pH * TDS * Part_size + pH * TDS * Location + pH * Part_size * Location + Conductivity * TDS * Part_size + Conductivity * TDS * Location + Conductivity * Part_size * Location + TDS * Part_size * Location + pH * Conductivity * TDS * Part_size + pH * Conductivity * TDS * Location + pH * Conductivity * Part_size * Location + pH * TDS * Part_size * Location + Conductivity * TDS * Part_size * Location + pH * Conductivity * TDS * Part_size * Location

FIG. 14. A representative Observed * Predicted * Std. Residual Plots.

major “hot-spot” included Site 6 (Synchrolift) at Cape Town Harbour (grossly contaminated with Sn, Cr, Co and Ni). The least contaminated was location 3. This was expected as this location was inside the sea. The order of heavy metal concentration in sediments was $Mn > Cr > V > Ni > Co > As$. Strong correlation was found between these metals (Mn and V in sizes <75 and ≥ 150), (As and Co in size ≥ 150), (Ni and Cr in size ≥ 150 and <75), respectively. The strong correlation that was found confirmed that their contribution was from anthropogenic sources i.e., industrial activities, storm and urban inflow, agricultural activities and ship reparation that is taking place in and around the harbour.

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