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Heavy metals contamination and risk assessment in sediments of Laucala Bay, Suva, Fiji

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ABSTRACT

Spatial distribution of metals (Cd, Cr, Cu, Fe, Ni, Pb and Zn) were studied in the surficial sediments from 20 sampling sites of Laucala Bay Suva, Fiji to assess the significance of wastewater effluent discharged from Kinoya Wastewater Treatment Plant (KWWTP) into Laucala Bay as a major source. The bioavailability of these metals was also assessed using the fractionation technique. The total concentrations of individual metals varied from 1.7–6.0 mg/kg Cd, 24.2–49.1 mg/kg Cr, 54–170 mg/kg Cu, 13,733–68,492 mg/kg Fe, 23.5–34.8 mg/kg Ni, 50–80 mg/kgPb and 15.5–157.0 mg/kg Zn. All the metals studied except Ni showed significant increase around the KWWTP effluent discharge sites indicating that prolong discharge of wastewater could be a major source of their accumulation in Laucala Bay. The multivariate statistical analysis confirmed that Cd, Cu and Pb could be attributed to similar sources like wastewater effluent. The study confirmed that the spatial distribution of the metals in Laucala Bay was affected by the hydrodynamic of Laucala Bay as pollutants were dispersed from the point sources. The Risk Assessment Code (RAC) assessed from the bioavailable fraction data confirmed that apart from Cd and Pb, the rest of the metals analysed had medium risk of toxicity. Owing to the persistent nature of metals and the results obtained from this study, there is a high probability of these metals to accumulate and increase in concentration in the future which could also lead to increase in their risk of toxicity to living beings.

1. Introduction

Metals have been classified as non-biodegradable pollutants which persist in the environment (Hung et al., 2012; Kumar, 2004; Shakya et al., 2006; Singh et al., 2011). Contamination of the coastal environment with metals is becoming a growing social and scientific concern worldwide (Shakya et al., 2006). There have been various cases of metal poisoning around the globe where the cause is anthropogenic enrichment of metals into the coastal environment (Harada, 1995; Nordberg, 2003). One such incident was the mercury (Hg) poisoning incident in Minimata, Japan where approximately 1043 death cases were reported (Harada, 1995). Similarly, a case of Cd poisoning was also reported in Toyama, Japan (Nordberg, 2003). The cities in the Pacific Island countries are undergoing rapid urbanization that places increased pressures on wastewater treatment infrastructure and solid waste management. If these infrastructures are not able to cope with increasing population then the surrounding aquatic environment will be contaminated with metals.

The coastal environments of Fiji are also facing problem of metals pollution (Maata and Singh, 2008). Laucala Bay is the coastal environment of interest for this study since it is known to have an interesting sediment environment with pollutant inputs from both marine and freshwater sources. There have been few studies done on metals pollution in Laucala Bay which concluded that the concentration of metals in the water samples was quite low due to the lower residence time of metals in water (Naidu et al., 1991). Other baseline study by Morrison et al. (2001) was on the sediments of Laucala Bay, Suva where the total contents of the metals: Pb, Al, Fe, Hg, Zn, Cu and Cd were analysed including in shellfish samples and concluded that Laucala Bay was not contaminated with metals. Nonetheless it is noteworthy that these studies in Laucala Bay are now outdated and does not take into account the rapid urbanization since the last three decades.

There is a possibility of overloading of metals through wastewater effluent discharged in Laucala Bay by the Kinoya Wastewater Treatment Plant (KWWTP). The KWWTP is the largest wastewater treatment facility operated by Water Authority of Fiji (WAF), which was designed with an initial capacity to serve population of 120,000 but is now serving a population equivalent of 155,000 with the current average dry weather flow of 31 Mega Litres/day in the Greater Suva Area (GSA) (WAF, 2018).The GSA, which consists of Suva City as well as the towns of Lami, Nasinu, and Nausori and their surrounding peri-urban areas, accounts for 57% of Fiji's urban population. Population

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Fig. 1. Sampling sites in Laucala Bay selected as representative of the entire Laucala Bay coastal environment (Source: Google Earth Pro).

growth in Suva over the past five decades has been modest but there has been a major growth in the peri-urban parts of the GSA where approximately 256,000 people reside. This is projected to increase to 350,000 by 2030 (FRA, 2014). Urban infrastructure and services have not kept pace with the rapid urban growth. The existing sewerage infrastructure covers only 36% of the GSA. The KWWTP is overloaded and is working at full capacity and faces challenges to process flows efficiently and therefore WAF has proposed to upgrade the existing KWWTP (WAF, 2018).

Due to metals nature to persist long in the environment, the metals concentration is expected to have risen in 25 years when compared to the baseline study (Morrison et al., 2001). Thus, it was extremely desirable to study metals pollution assessment in the sediments of Laucala Bay, Suva, Fiji. This study was an excellent monitoring platform which has provided new insights in relation to the metals contamination level in the sediments of Laucala Bay because of wastewater effluent discharge from the KWWTP and other terrigenous, anthropogenic and natural sources as reported in this paper. In addition, a comprehensive study on the toxicity level of these metals was conducted through the use of Sediment Quality Guidelines (SQGs) and Risk Assessment Code (RAC). Effects range low (ERL) and effects range medium (ERM) were the SQGs used to assess the toxicity level of the metals. ERL represents the concentration below which toxic effects on aquatic species will be rarely witnessed while ERM value indicates the concentration above which adverse toxic effects occur frequently on aquatic species (Castillo et al., 2013). RAC was used to assess the ability of metals to become free from the sediments and become bioavailable. RAC assessed this by applying a scale to the percentage of the heavy metal in the carbonate and exchangeable fractions.

Multivariate statistical analysis and various indices were used for the analysis of data. This was done to better understand the contamination and ecological risks posed by metals and consequently understand the discrepancies of different risk assessment indices. The various indices used can be classified into contamination indices, background enrichment indices and ecological risk indices. The I_{geo} and EF were the two contamination and background enrichment indices used. The RAC and SQGs (ERL and ERM) were the ecological risk indices that were used in this study (Zhuang and Gao, 2014). It is obvious that the different assessment indices used for predicting environmental risks of metals were not consistent with each other. Yu et al. (2011) recommended that risk assessment process using SQGs on the basis of metals availability and site specificity should be considered. However, combination of statistical analysis and use of various indices helped to provide a comprehensive understanding of the heavy metal risks in Laucala Bay.

2. Materials and methods

2.1. Study area and sampling sites

The study area, Laucala Bay is located between the western side of the Suva peninsula and the eastern side of the Rewa River delta in Suva, Fiji. The topography of Laucala Bay is relatively flat but it gets hilly while approaching the center of the Suva peninsula, separating Laucala Bay and Suva harbour. Laucala Bay is bordered by reefs on the ocean side which restrict the exchange of water between the Bay and the open ocean; hence, inhibiting the dispersal of polluting agents (Naidu et al., 1991).

There are also various river mouths entering into Laucala Bay. This includes the Rewa River delta which is the widest river in Fiji that lies to the east of Laucala Bay. The distributaries of this river are Vunidawa River and Vunivadra River which enter into Laucala Bay. The western

side of Laucala Bay has the Vatuwaqa River, Nasinu River, Samabula River and Uluituni Creek opening into it (Morrison et al., 2001). An 800 m outfall pipe lies between Nasinu and Samabula River mouths which discharges the treated wastewater effluents from the KWWTP into Laucala Bay (Naidu et al., 1991). The predominant 7 sampling sites were in the area where the KWWTP's treated effluent is discharged. In addition, other sampling sites were chosen to provide significant comparison in terms of the heavy metal concentrations. These sites were mainly away from the land mass, close to the reefs, away from the effluent discharge area and close to the mouths of estuaries opening into Laucala Bay. Fig. 1 shows the exact location of the 20 sampling sites in Laucala Bay.

2.2. Sampling and preservation

A grab sampler was used to collect the surficial sediment samples from the 20 sites in Laucala Bay. Composite samples were obtained from each site where 3 grab samples collected from one site were mixed and placed into polyethylene bags for storage. This was done because composite samples are better representatives of the sampling site when compared to individual samples (Kinzelman et al., 2006; Petersen et al., 2004). The textural features of the sediments samples from each site were noted immediately after collection. It was observed that sediments obtained along the reef sites (sites 13, 14, 15) were mostly sandy while sediment samples from the rest of the sites appeared clayey. Physical parameters including pH, temperature, DO and salinity of the overlying water for each sampling site were measured in-situ.

2.3. Analytical methods and quality control

The sediment samples were air dried, crushed gently in a mortar and pestle to disaggregate the sample, sieved in 63 μm plastic sieves and pulverized prior to digestion (Hseu et al., 2002). The ISO 11466 aqua regia extraction procedure was used for the digestion of the sediment samples for all the metals analysis (ISO, 1995). The digested samples were analysed with atomic absorption spectrometer (AAS, Perkin Elmer, AAnalyst 400).

The quality control measures such as the use of Standard Reference Material (SRM), percentage recovery, blank analysis, analysis in triplicates and method detection limits were employed in order to justify the reliability and validity of the methods used. The SRM (NIST 1646a) results obtained verified the accuracy of the method employed for heavy metal analysis. In addition, the recovery (%) for all the metals analysed were within the acceptable range of 90–100% (Addis and Abebaw, 2016) or 90–120% (APHA, 2017) which indicated that the results were valid with minimal amount of matrix interference. The analytical precision for the replicate samples achieved was between 0.15 and 7.7% which was within the acceptable relative standard deviation (RSD %) range that is ≤15% (Addis and Abebaw, 2016).

2.4. Sequential extraction and normalization

The content of metals bound to the bioavailable and the non-bioavailable fractions in the sediment samples were determined by following the Standards, Measurements and Testing Programme's (SM&T) fractionation technique previously known as the Community Bureau of Reference (BCR) technique (Pueyo et al., 2001). This procedure has been considered a reliable method in various sequential extraction studies conducted on metals (Castillo et al., 2013). The extracts from each fraction after extraction were analysed for the metals content using AAS.

In order to understand the level of anthropogenic input of the metals in sediment over their natural origin a normalization procedure was carried out through the calculation of the enrichment factor (EF) and the geo-accumulation index (I_{geo}).

2.4.1. Enrichment factor

Li has been certified as an equally good normalizer as Al, it was used as the normalizer in this study (Aloupi and Angelidis, 2001; Du Laing et al., 2009). The sediment samples were digested using the ISO 11466 aqua regia digestion procedure and analysed for Li. The results obtained were used to calculate EF using Eq. (1), where [M] refers to the concentration of the heavy metal of interest while [Y] is the concentration of the normalizing element (Li). These two concentrations were obtained both for the sample and the set of reference background where the average shale values of each heavy metal and the normalizing element from literatures were used as the background reference concentration (Ho et al., 2012; Silva-Filho et al., 2014; Turekian and Wedepohl, 1961).

$$EF = \frac{\left(\frac{[M]}{[Y]}\right)_{sample}}{\left(\frac{[M]}{[Y]}\right)_{background}} \quad (1)$$

2.4.2. Geo-accumulation index

The value for I_{geo} was calculated using eq. (2), where C_n is the concentration of the heavy metal (n) measured while B_n is the geochemical background concentration of that particular heavy metal (n) (Muller, 1969). The average shale value from the literature was used as the background concentration (Turekian and Wedepohl, 1961).

$$I_{geo} = log_2 [C_n/1.5\, B_n] \quad (2)$$

2.5. Total organic carbon (TOC)

The Walkley-Black titration method was used to quantify the TOC in the sediment samples (Gaudette et al., 1974). It is well known that TOC measurements formarine sediments are prone to chloride (Cl^-) interference. Therefore, silver sulphate ($AgSO_4$) was used during the digestion to remove any Cl^- interference present in the sample matrix whereby Cl^-precipitated out as AgCl (Schumacher, 2002).

2.6. Statistical data analysis

The SPSS software was used to perform the statistical analysis of the results obtained from all the experimental analysis. This included the use of Kruskal-Wallis test to determine whether any significant difference lies between the concentrations of the heavy metal in the sediments collected from the wastewater effluent discharge site in comparison to the other sampling sites. In addition to this a post hoc was conducted to determine where exactly the difference exists. Kendall's tau-b correlation was used in order to comment on the correlation between the various confounding variables and the metals concentration obtained at each site (Johnson, 2009). Furthermore, the Principal Component Analysis (PCA) was used to compress the number of variables which in this case are the metals in order to explain the variance in their source. The results from EF and I_{geo} calculations were used to comment on the anthropogenic enrichment of the metals at a particular site. The results from the SM&T sequential extraction procedure was used to comment on the levels of metals in the bioavailable forms which could pose health risks to humans at a particular site studied.

3. Results and discussion

3.1. Total content of metals and comparison

The results obtained for the total heavy metal content in sediment samples are shown in Table 1.

The values presented in the Table 1 are the mean of the triplicate analysis with SD and calculated on the dry weight basis. The results in Table 1 suggest that KWWTP effluents are contributing towards the

Table 1
Total heavy metal concentrations at various sites in Laucala Bay. Sites 1–7 are sewage wastewater effluent discharge sites, sites 8, 11, 19, 20 are river mouths, sites 13–15 are sites close to the reef and the sites 9, 10, 12, 16–18 are coastal sites close to landmass.

Sites	Total heavy metal concentrations (mg/kg)						
	Cd	Cr	Cu	Fe	Ni	Pb	Zn
1	5.2 ± 0.4	36.2 ± 0.7	144.0 ± 3.0	51,649 ± 200	29.1 ± 0.4	80.0 ± 3.0	109.8 ± 0.5
2	4.9 ± 0.3	48.3 ± 0.4	147.0 ± 3.0	51,015 ± 100	29.0 ± 0.3	79.0 ± 2.0	127.0 ± 0.5
3	4.9 ± 0.1	42.4 ± 0.3	139.0 ± 2.0	49,598 ± 100	24.0 ± 0.4	79.0 ± 3.0	111.0 ± 3.0
4	5.3 ± 0.1	48.3 ± 0.4	137.0 ± 2.0	54,639 ± 100	24.3 ± 0.8	80.0 ± 4.0	120.3 ± 0.8
5	6.0 ± 0.1	47.8 ± 0.9	143.0 ± 3.0	57,014 ± 100	24.1 ± 0.9	79.0 ± 2.0	135.0 ± 2.0
6	5.2 ± 0.4	46.3 ± 0.9	138.0 ± 2.0	59,476 ± 100	23.0 ± 0.4	77.0 ± 3.0	157.0 ± 3.0
7	5.2 ± 0.2	46.4 ± 0.8	138.0 ± 3.0	58,179 ± 200	23.5 ± 0.8	77.0 ± 1.0	115.0 ± 2.0
8	4.4 ± 0.1	36.3 ± 0.1	170.0 ± 2.0	59,995 ± 100	34.0 ± 1.0	79.8 ± 0.8	90.7 ± 0.7
9	4.4 ± 0.2	42.1 ± 0.3	142.0 ± 4.0	68,492 ± 200	34.0 ± 0.5	65.0 ± 0.4	87.0 ± 2.0
10	4.1 ± 0.1	42.2 ± 0.3	101.0 ± 3.0	66,177 ± 100	29.0 ± 0.7	70.0 ± 3.0	76.9 ± 0.9
11	4.2 ± 0.1	49.1 ± 0.2	121.0 ± 3.0	63,186 ± 200	31.6 ± 0.7	51.0 ± 0.5	84.3 ± 0.6
12	3.1 ± 0.1	48.4 ± 0.3	61.0 ± 4.0	66,227 ± 200	34.6 ± 0.8	50.0 ± 0.4	86.0 ± 1.0
13	1.9 ± 0.1	24.2 ± 0.7	54.0 ± 2.0	13,733 ± 80	28.0 ± 0.4	50.3 ± 0.6	15.7 ± 0.5
14	1.7 ± 0.1	24.4 ± 0.4	44.2 ± 0.7	22,230 ± 60	34.8 ± 0.9	50.7 ± 0.8	23.0 ± 0.5
15	2.1 ± 0.1	24.7 ± 0.5	46.1 ± 0.5	20,455 ± 80	30.7 ± 0.9	50.0 ± 0.3	15.5 ± 0.8
16	3.3 ± 0.2	30.0 ± 0.4	56.0 ± 2.0	47,067 ± 100	24.0 ± 0.3	63.0 ± 2.0	75.0 ± 2.0
17	3.9 ± 0.1	36.1 ± 0.4	68.0 ± 1.0	49,591 ± 100	29.1 ± 0.7	62.0 ± 0.9	97.0 ± 4.0
18	4.1 ± 0.3	41.2 ± 0.2	73.0 ± 2.0	49,755 ± 200	33.0 ± 0.2	54.0 ± 0.4	119.0 ± 3.0
19	4.3 ± 0.1	31.0 ± 0.4	75.0 ± 2.0	43,494 ± 200	24.0 ± 0.2	74.0 ± 3.0	71.9 ± 0.9
20	3.1 ± 0.1	29.5 ± 0.6	76.0 ± 1.0	54,602 ± 100	24.7 ± 0.8	66.0 ± 0.4	52.5 ± 0.8

metals load in Laucala Bay. However, just by referring to the concentration of a particular heavy metal at the studied sites it cannot be proven that indeed KWWTP is a significant source of heavy metal in comparison to other sources. Thus, multivariate analysis was performed in order to justify that a significant difference indeed exists.

The concentrations of metals obtained in the present study were compared with the previous study done by Morrison et al. (2001). After determining the percentage difference it is evident from Table 2 that there is a positive increase in all the metals concentrations at the KWWTP site. This is because of the continuous discharge of wastewater effluents at the respective site where metals tend to accumulate in sediments which act as reservoirs of metals (Maata and Singh, 2008; Sany et al., 2013). The concentration of Cd and Pb showed a large positive increase (%) at all the sites studied. This could probably be due to the increased anthropogenic input of Cd and Pb through the wastewater discharge and other riverine inputs. It was also seen that there was an increase in Cu concentration at all the sites except for Uluituni Creek, Vatuwaqa River and the sites between this creek and the river. It is possible that the anthropogenic release of Cu at these three sites have minimized which led to the decrease in their concentrations. This decrease could be attributed to the closure of the Raiwaqa sewage treatment plant, which was previously discharging wastewater effluent into the Vatuwaqa River but now no longer carried to Laucala Bay which could consequently leads to the decrease in the accumulation of Cu in the sediments around the Vatuwaqa River (Morrison et al., 2001). Thus, the discharge of the wastewater effluent plays a critical role in the metals contamination of the receiving water body. However, the trend observed for Cu was not apparent for the other metals studied suggested that the wastewater discharge from Raiwaqa was not a significant source for other metals as for Cu. It is noteworthy that there has been an increase in the squatter settlements in the Vatuwaqa area. These areas discharge most of their household wastes into the nearby Vatuwaqa River which has caused increased load of the other metals significantly.

Table 2
Percentage difference between the present study and the baseline study. Sites 1–7 are sewage wastewater effluent discharge sites, sites 8, 11, 19, 20 are river mouths, sites 13–15 are sites close to the reef and the sites 9, 10, 12, 16–18 are coastal sites close to landmass.

Sites	Difference (%)				
	Cd	Cu	Fe	Pb	Zn
1–7	589.47	73.01	18.00	927.31	10.62
8	486.67	137.10	7.83	303.61	−12.98
10	355.56	29.82	10.70	718.18	−30.72
11	600.00	91.15	15.33	825.59	−3.10
12	400.00	7.02	3.97	748.90	−20.37
13	156.76	4.65	1.70	676.23	−60.75
14	153.73	84.94	1.93	415.77	−28.13
15	169.23	45.43	2.22	463.7	−64.77
16	392.54	−31.46	3.54	375.47	−47.92
18	561.29	−9.32	4.22	394.05	−7.75
19	532.35	−15.54	4.81	727.74	−53.01
20	400.00	9.04	2.93	812.86	−51.83

It is evident from Table 2 that the increase (%) of Fe in Laucala Bay over a period of around 25 years was quite low i.e. < 20%. This low increase could be due to minimal anthropogenic enrichment or low accumulation due to the natural sources such as weathering processes (Eliseo Ochoa-Valenzuela et al., 2009; Levei et al., 2014). On the contrary, the concentration of Zn as seen in Table 2 has declined from the time of the baseline study (Morrison et al., 2001) and the only increases were seen at the KWWTP sites which could be possibly due to the continuous discharge of the wastewater effluents into the Bay. It is in line with a study done by Gaynor and Gray (2004) on the sediments of the Avoca River in Ireland where 35% decrease in Zn concentration was determined between the year 1994 and 2001. The decrease in the Zn concentration in Laucala Bay could be because of the decrease in its anthropogenic sources with the implementation of the Environmental Management Act (EMA) by the Government of Fiji. According to this strategy, all the wastes from industries should be discharged into the sewerage system and should not be directly discharged into the nearby rivers or creeks (SPREP, 2001). This probably explains the decrease in other sites but an increase in KWWTP.

In order to get an idea on the severity of metals pollution in Laucala Bay, the results obtained for the total metals content in the present study were also compared to the results of other studies in the Pacific region as shown in Table 3. The concentration of Cd obtained in the current study seemed to be greater than that of previous studies except for the study done in Rewa River, Fiji. It is evident from Table 3 that the concentration of Cr is comparable to the concentration obtained in

Table 3
Comparison of the total heavy metal contents in Laucala Bay with other studies done in the Pacific region.

Study sites	Average heavy metal concentration (mg/kg)							Reference
	Cd	Cr	Cu	Fe	Ni	Pb	Zn	
Laucala Bay	1.7–6	24.2–49.1	44.2–170	13,733–68,492	23–34.8	50–80	15.5–157	Present study
Vitogo River	1.2–2.2	35–100	82–150	90,000–190,000	–	6.8–10	54–220	Gangaiya et al., 1988
Great Astrolabe reef, Kadavu	1.9	20	29.6	–	8.7	7.3	29.5	Morrison and Naqasima, 1999
Rewa River, Fiji (2000)	0.4–11	–	4.5–60	2200–7800	–	4.5–106	5.5–584	Deo, 2000
Lami, Fiji	–	–	5–1980	–	–	4.2–516	52–514	Gangaiya et al., 2001
Tonga	0.1–0.9	–	2–180	1550–114,000	2–25	3–31	6–208	Morrison and Brown, 2003
Tanapag Lagoon, Saipan	1.83–2.08	38.1–44.6	55.9–67.9	–	62.7–83.2	46.4–60	58.2–71.4	Denton et al., 2006
New Zealand	–	2.4–94	14–48	–	7.6–17.1	23–130	97–896	Redfern, 2006
Kiribati		0.9–12.6	0.3–13.9	–	–	3.4–13.3	1.2–77	Redfern, 2006
Suva Harbour	–	–	21.4–143	14,000–48,700	–	22.1–93.5	40.2–269	Maata and Singh, 2008
Samoa- Commercial Port	–	–	1090–3710	–	–	90–130	–	Imo et al., 2014
Samoa-Fishing Port	–	–	1240–3410	–	–	1320–1850	–	Imo et al., 2014

Tanapag Lagoon, Saipan which indicated a common level of anthropogenic enrichment. The concentration of Cu when compared to the other studies displayed similar results to the study conducted in Vitogo River, Tonga, Saipan and Suva Harbour (Denton et al., 2006; Gangaiya et al., 1988; Maata and Singh, 2008; Morrison and Brown, 2003). This was mainly because these areas have been also quite urbanized and industrialized which are contributing source of Cu in the coastal environment. Similarly, Pb, Zn, Fe and Ni showed comparable results with other studies as shown in Table 3 and this could yet again be attributed to similar sources.

Table 3 also shows that the ranges of concentrations of Cu, Pb and Zn at Laucala Bay were far less than the concentrations obtained in the Lami River sediments. It is noteworthy that Lami River is densely industrialized and due to lack of enforcement of the National Liquid Waste Management Strategy and other environmental policies, industries were discharging most of their wastes into this river (SPREP, 2001; Gangaiya et al., 2001). Since the Lami River is quite small as compared to Laucala Bay there is less dispersion of pollutants leading to the increased accumulation of Cu, Pb and Zn (Gangaiya et al., 2001). With the implementation of National Liquid Management Strategy it would be interesting to investigate the current metal concentrations in Lami River. When compared to the pristine environment i.e. the Great Astrolabe Reef in Kadavu, the concentrations of all the metals analysed in Laucala Bay sediments are quite high. Thus, it can be concluded that the metals pollution load of Laucala Bay is of concern although the concentration is less than that of the Lami River.

3.2. Multivariate statistical analysis

Prior to performing a Kruskal-Wallis test and a Post hoc test, the 20 sites studied were divided in to 3 categories as: 1. KWWTP effluent discharge sites (1–7), 2. Sites close to river mouths and in close proximity to land (8–11 and 16–20) and 3.Sites (13–15) away from the land but close to the reefs.

3.2.1. Kruskal-Wallis test and post hoc test

A Kruskal-Wallis test performed at 0.05 significance level showed that a significant difference exists across the categories of sites in terms of Cd, Cr, Cu, Fe, Pb and Zn concentration. Since the significance level for Ni was > 0.05, it was concluded that no significant difference in Ni concentrations existed among the 3 categories of sites. The Post hoc test was also conducted at 0.05 significance level for the studied metals which showed significant difference among 3 categories summarized in Table 4.

Table 4 clearly shows that the concentrations of Cd, Cu and Pb were significantly different incase of categories 1 and 2 and categories 1 and 3. However, the concentration of the same metals did not show significant difference between categories 2 and 3. This indicates that the effluent discharged by the KWWTP is a significant source of Cd, Cu and Pb in Laucala Bay. The concentrations of Cd, Cu and Pb did not differ significantly for the sites closer to the land, near river mouths and away from the land that is close to the reef sites. Although the sites close to the reef are away from major anthropogenic and terrigenous point sources it clearly shows that the fate of transport of these metals to the reef sites is dependent on factors which govern sediment movement. These factors include wind and wave action as a result of extreme weather events i.e. flooding, cyclones, hurricanes or increased water flow alterations and bioturbation (Reuther, 2009). Thus, once released through the terrigenous and anthropogenic sources, these metals could be transported towards the reef system in Laucala Bay through wave action due to extreme weather conditions such as storms, hurricanes, earthquakes and flooding due to heavy rainfall. Upon comparison, it was noted that the average concentration of Cd, Cu and Pb at the reef sites in the present study is greater than that of the study done in the Great Astrolabe Reef. Since the data does not correspond with that of the pristine environment it is highly unlikely for the sources of these metals near the reefs to be of lithogenic origin. Therefore, hydrodynamics or wave action due to the extreme weather conditions play an important role in dispersing the sediments with metals from terrigenous and anthropogenic sources towards the Nukubuco and Suva Reef.

Table 4
Results from Post hoc test for pairwise comparison among 3 categories (cf. Section 3.2).

Pairwise comparison	Significance value					
	Cd	Cr	Cu	Fe	Pb	Zn
1–2	0.003	0.154	0.020	0.739	0.016	0.081
1–3	0.035	0.002	0.023	0.008	0.018	0.000
3–2	0.565	0.032	0.521	0.021	0.500	0.008

The pairwise comparison (Table 4) for the concentrations of Cr, Fe and Zn, showed no significant difference at the KWWTP sites (category 1) and the river mouths and close to land sites (category 2). However, the concentration of the same metals differed significantly between the KWWTP sites and the sites away from the land (category 3) and similar observations were made between category 2 and category 3 sites (Table 4). This observation concluded that the sources of Cr, Fe and Zn were mostly sewage effluent discharge and discharge from the land based sources through riverine inputs.

3.2.2. Kendall's tau-b correlation test

A Kendall's tau-b correlation test was used to comment on the

Table 5
Correlation matrix of the studied metals and physicochemical parameters.

	Cd	Cr	Cu	Fe	Ni	Pb	Zn	TOC (%)	pH	DO	Salinity	Temperature
Cd	1	0.106	0.672**	0.295	0.000	0.649**	0.171	0.756**	0.703**	−0.788**	−0.075	0.322
Cr		1	0.222	0.512**	−0.048	0.295	0.522**	0.396*	0.564**	−0.575**	−0.042	0.122
Cu			1	0.227	−0.134	0.633**	0.522*	0.565**	0.553**	−0.639**	0.000	0.234
Fe				1	0.075	0.171	0.405**	0.253	0.249	0.484**	0.111	0.039
Ni					1	−0.304	−0.246	−0.232	−0.253	0.403**	0.123	−0.207
Pb						1	0.290	0.695**	0.620**	−0.642**	0.113	0.310
Zn							1	0.611**	0.706**	−0.663**	−0.185	0.327
TOC (%)								1	0.674**	−0.611**	0.037	0.405*
pH									1	−0.706**	0.011	0.313
DO										1	−0.005	−0.183
Salinity											1	−0.045
Temperature												1

similarity in the sources of metals at the sites studied (Gao and Chen, 2012; Mahmoudabadi et al., 2015; Vicente-Martorell et al., 2009). The results obtained are displayed in terms of correlation matrix shown in Table 5 which shows that Cr and Zn showed positive correlation with Fe and further confirms the Kruskal-Wallis test for these metals.

In addition, Cd, Cu and Pb also showed positive correlations with each other and also with TOC (%) which is indicative of having major common sources of anthropogenic and terrigenous origins. The correlation matrix presented in Table 5, shows that Ni does not show a positive correlation with any of the metals analysed.

3.2.3. Principal component analysis

The relationships between the metals through the correlation matrix appear intricate and difficult to explain individually. Hence, in order to further justify the variance in their sources, the Principal Component Analysis (PCA) was carried out on the correlation matrix to compress the number of variables i.e. metals. (Loska and Wiechuła, 2003). Three principal components (PC) were extracted after conducting PCA using SPSS software as shown in Fig. 2 which is in three-dimensional rotated space. PC 1 and PC 2 displayed eigenvalues of 3.5 and 1.6, respectively and they contributed towards 72.5% of the cumulative variance. Although the third component has an eigenvalue of 0.9 which is < 1, it still explains about 13.1% of the total variance and contributed significantly towards the cumulative variance of 85.7%.

Fig. 2 clearly shows that PC 1 showed strong loadings for Cr, Fe and Zn and weak loading for Cd, Cu, Pb and Ni. This indicates that Cr, Fe and Zn present in Laucala Bay are of similar origin but different to the sources of Cd, Cu, Pb and Ni. PC 2 showed strong loadings of only Ni and a weak loading for the remaining 6 metals. This is an indication that the source of Ni in Laucala Bay could be different from the rest of the metals analysed. PC 3 showed strong positive loadings for Cd, Cu and Pb and weak loadings for the remaining metals. This confirms a common origin for Cd, Cu and Pb. Since PC 1 and PC 3 showed some correlation as seen in the correlation matrix, it was noted that the positive loadings of Cu and Pb did not differ much between PC 1 and PC 3. Thus, at certain sites the sources of Cu and Pb could be similar to the source of Cr, Fe and Zn. The strong positive loading of Cd for PC 3 suggested that the wastewater discharge is the main source for Cd in Laucala Bay.

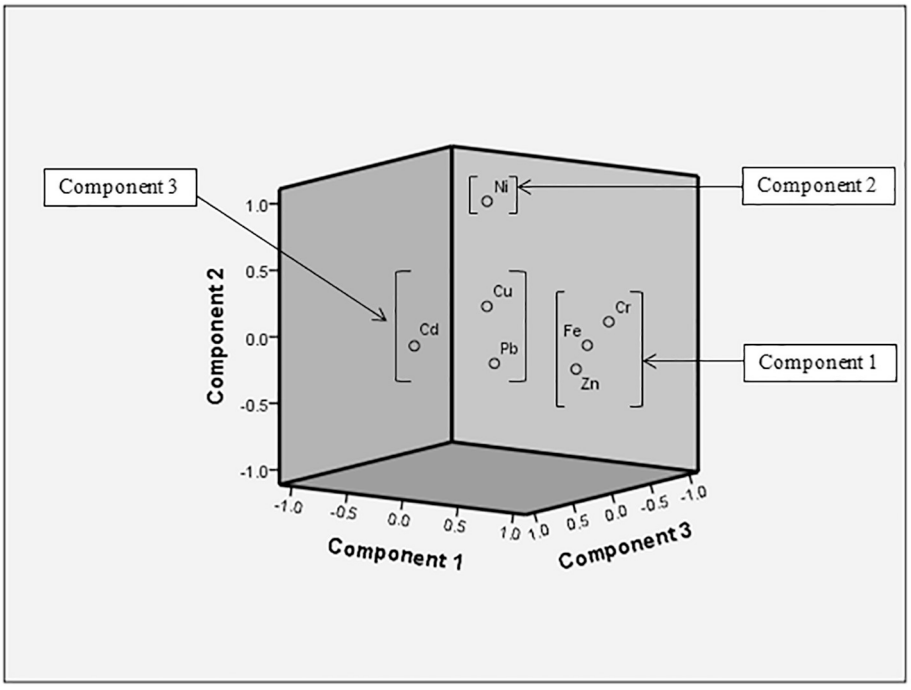

Fig. 2. Component plot in rotated space showing the loadings of the different metals in each of the 3 components as it has been described earlier on.

Table 6
Enrichment Factors for the studied metals at different sites in Laucala Bay. Sites 1–7 are sewage wastewater effluent discharge sites, sites 8, 11, 19, 20 are river mouths, sites 13–15 are sites close to the reef and the sites 9, 10, 12, 16–18 are coastal sites close to landmass.

Sites	Metals enrichment factor						
	Cd	Pb	Cu	Fe	Cr	Zn	Ni
1	7.92	6.37	9.58	2.62	3.13	2.51	0.20
2	6.60	6.34	9.85	2.61	3.85	2.92	0.20
3	6.28	6.03	8.32	2.41	3.20	2.43	0.15
4	6.71	6.21	8.67	2.30	3.18	2.28	0.14
5	9.12	6.28	9.49	2.89	3.80	3.08	0.16
6	6.14	6.90	8.80	2.90	3.54	3.44	0.15
7	6.36	6.09	9.12	2.94	3.67	2.61	0.16
8	6.16	6.73	5.73	2.65	3.49	2.52	0.28
9	5.71	5.59	7.43	4.69	3.02	3.75	0.43
10	5.46	5.19	8.70	3.46	3.08	3.73	0.61
11	5.68	5.25	7.35	4.69	2.60	4.22	0.47
12	5.44	5.80	3.59	3.81	2.23	2.89	0.25
13	4.27	3.81	4.35	1.04	2.00	0.38	0.20
14	4.83	3.84	3.32	0.47	2.24	0.49	0.22
15	3.73	3.80	3.60	1.89	1.83	1.54	0.3
16	5.79	4.00	5.02	2.18	2.78	2.25	0.21
17	5.77	4.98	5.54	4.01	2.40	3.95	0.35
18	5.35	3.58	5.45	4.08	2.91	2.56	0.60
19	5.29	5.55	3.38	4.45	2.90	2.87	0.87
20	5.58	3.39	3.83	3.76	2.83	2.34	0.48

3.3. Enrichment factor (EF)

The enrichment factors for the studied metals were determined to comment on the extent of contamination due to anthropogenic activities. Table 6 lists the EF values obtained in the present study using lithium (Li) as the normalizer (Aloupi and Angelidis, 2001; Loring, 1990). It is evident that the EF of Ni at all the sites studied is < 2. As per the criterion set by Han et al. (2006), EF values of < 2 indicate minimal enrichment and thus Ni mainly has natural origin (weathering of sandstone, sedimentary rocks, clay minerals, slate, basalt, etc.) which is different to other metals analysed.

The values of EF for Cd, Cu and Pb at sites 1, 2, 3,4,5,6 and 7 are in range of 5 to 10 indicating significant enrichment maybe from KWWTP effluent discharge. It is known that the KWWTP discharges sewage effluent which has not been treated for metals removal. Albeit HMs concentration in sewage effluent is low but due to the persistent nature of the metals they would have been accumulated in the sediments over time resulting in the high concentrations at these sites (Shamuyarira and Gumbo, 2014; Tytła et al., 2016; Zhang et al., 2017). In addition, it is important to point out that the high content of Pb at the discharge site could be possibly due to the corrosion of the outfall pipeline and or the internal corrosion in the wastewater distribution system at the KWWTP (Peng et al., 2010; Slavíčková et al., 2013).

The EF values for Cd, Cu and Pb at sites 8, 9, 10, 11, 12, 16, 17, 18, 19 and 20 were between 3 and 8 which confirmed these sites to be moderately-significantly enriched with Cd, Cu and Pb due to anthropogenic origin. However, the Post hoc test suggested that the concentrations of Cd, Cu and Pb at these sites were significantly different from the concentration obtained at KWWTP sites (Table 4). This shows that Cd, Cu and Pb at these sites originate from a totally different anthropogenic source. Since these sites are close to the river mouths and land mass, one probable source could be wastes disposed from the industries and households, fertilizer run-offs from plantations along the river, sewage discharge by stocks grazing and squatter settlements alongside the rivers which open into Laucala Bay eventually. As the sites 13, 14 and 15 were close to the reefs so the EF values for Cd, Cu and Pb were between 2 and 5 which indicated moderate enrichment. This meant that there was an average amount of enrichment from the anthropogenic sources. This result coincides with the result of the Post hoc test for Cd, Cu and Pb at the sites close to the reef which confirmed that indeed the wave action due to extreme weather conditions is responsible for dispersing the contaminants from near coastal sources to sites closer to the reefs.

Overall, it can be concluded that Cd, Cu and Pb were mainly of the anthropogenic origin at most of the sites studied. At the KWWTP effluent discharge sites, these metals could be from the industrial wastewater discharged where their concentrations were found to be significantly higher in comparison to the same set of metals at the other sites (WAF, 2016). The wastes released into the sewerage system from different industries in the form of wastewater maybe significantly concentrated with Cd, Cu and Pb. Inspite of being exposed to the anthropogenic sources, the concentrations of Cd, Cu and Pb at the reef sites were less than the concentrations at the other sites because of the dilution effect. It should be noted that the organic matter content at the reef sites were less than the organic matter content at the other sites. Organic matter has been an important complexing agent which negates the dilution effect of metals in waters; hence, leading to the accumulation of metals in sediments (Song et al., 2015). Therefore, a low content of organic matter could be a factor contributing to the observed low concentration of metals near the reefs.

The EF of Cr, Fe and Zn at all sites except 13,14 and 15 was between 2 and 5 which indicated moderate enrichment. This indicates that in addition to the contribution due to the natural sources of metals at these sites there is also an average level of contribution from the anthropogenic sources. Since the Post hoc test results for Cr, Fe and Zn showed no significant difference between the metals concentration between the KWWTP sites and the sites near the river mouths and close to the land (Table 4), it was concluded that the anthropogenic source of these metals is the wastewater effluent discharged from the KWWTP and also the terrigenous input. The probable reason for moderate enrichment could be occurrence of low levels of these metals in the wastewater effluents discharged as majority of it could have been removed in the sewage sludge. The values of EF for Cr, Fe and Zn for sites 13, 14 and 15 were < 2 which meant minimal if any enrichment from the anthropogenic input. The Post hoc test results also showed that the concentrations of these metals at sites 13, 14, 15 were significantly different from both the KWWTP sites and the river mouth sites (Table 4). Hence, the study confirmed that the anthropogenic sources

Table 7
Geo-accumulation index (I_{geo}) for the studied metals at different sites in Laucala Bay. Sites 1–7 are sewage wastewater effluent discharge sites, sites 8, 11, 19, 20 are river mouths, sites 13–15 are sites close to the reef and the sites 9, 10, 12, 16–18 are coastal sites close to landmass.

Sites	Metals I_{geo} values						
	Cd	Cr	Cu	Fe	Ni	Pb	Zn
1	3.53	1.26	3.26	1.94	−1.81	2.42	1.87
2	3.26	1.68	3.29	1.92	−1.81	2.40	2.08
3	3.26	1.49	2.86	1.88	−2.09	2.40	1.89
4	3.56	1.68	3.19	2.02	−2.07	2.42	2.00
5	3.74	1.67	3.25	2.08	−2.08	2.40	2.17
6	3.22	1.63	3.20	2.14	−2.15	2.36	2.39
7	3.22	1.63	3.20	2.11	−2.12	2.36	1.94
8	3.29	1.26	3.50	1.67	−1.58	2.41	1.60
9	3.29	1.49	3.24	1.86	−1.58	1.58	1.54
10	2.08	1.49	2.75	1.81	−1.81	2.42	1.36
11	1.92	1.71	3.01	1.64	−1.69	1.77	1.49
12	2.08	1.68	2.02	1.71	−1.56	1.74	1.52
13	2.87	0.68	2.79	−4.02	−1.87	1.75	−0.93
14	2.92	0.68	1.05	−0.44	−1.55	1.76	−0.38
15	3.19	0.68	2.32	−3.99	−1.73	1.74	−0.95
16	3.29	1.00	1.62	1.28	−2.09	2.07	1.32
17	2.96	1.26	2.18	1.71	−1.81	2.05	1.69
18	3.26	1.45	2.28	1.72	−1.63	1.55	1.99
19	1.92	1.05	2.12	1.01	−2.09	2.3	1.26
20	2.08	0.74	2.34	0.18	−2.79	1.62	0.81

contributing towards the concentrations of Cr, Fe and Zn at the KWWTP sites and the river mouth sites are not significantly affecting the concentrations of the same metals near the reefs.

3.4. Geo-accumulation index (I_{geo})

In order to justify the EF obtained, I_{geo} values were also calculated for all the metals studied and reported in Table 7. A similar trend was observed in terms of the contamination category associated with the I_{geo} of studied metals and the enrichment category associated with the EF values of the same metals. The I_{geo} values for Cd, Cu and Pb were mostly between 1-2 and 2-3 which were assigned to be moderately contaminated and moderately to strongly contaminated(Muller, 1969). This indicated that Laucala Bay was mostly contaminated with Cd, Cu and Pb and as per the EF values the same metals that are mostly enriched by anthropogenic means. Similarly, the I_{geo} values for Cr, Ni and Zn were mostly between 1 and 2 which corresponded to a moderately contaminated category and the same metals showed to have moderate enrichment as per the EF category as well. This finding indicated that along with the anthropogenic enrichment, there were some natural sources which were significantly contributing towards the moderately contaminated status of Laucala Bay in terms of Cr, Fe and Zn contents. However, the I_{geo} of Ni was below zero at all the sites which clearly indicated Laucala Bay is uncontaminated with Ni. This was further justified by the fact that the concentration of Ni analysed for all the studied sites of Laucala Bay was much lower (Table 1) than the average shale value of 68 mg/kg (Turekian and Wedepohl, 1961). The EF for Ni was much < 2 which confirmed no major anthropogenic enrichment and the observed concentrations were primarily of natural origin.

3.5. Risk assessment of metals at Laucala Bay

3.5.1. Sediment quality guidelines

Sediments Quality Guideline (SQG) was used to rank and prioritize the sites the show metal toxicity. There are two values used as guideline for each heavy metal given as effects range low (ERL) and effects range median (ERM). It is noteworthy that the two values generated are not toxicity thresholds meaning that there is no assurance that a value less than ERL will have no toxicity and a value more than ERM will be toxic. In order to do this, toxicity tests need to be performed. The ERL and ERM values are used just to provide an estimate concentration of heavy metal below which toxicity is least likely or most likely to occur. However, ERM values for metals are better indicators for sites of concern in regards to the metals toxicity than the ERL values (Hübner et al., 2009; Praveena et al., 2008; Zheng et al., 2008). Long et al. (1995) has proposed ERL and ERM guideline values for heavy metals which are stated in Table 8.

It was noted that the concentration of Cr and Zn was well below their respective ERL guideline at all the sites studied. This meant that Cr and Zn toxicity is least likely to occur in Laucala bay as of now. None of the metals studied showed concentrations above the ERM guideline but 90% of the sites showed Pb and Cu concentrations between the ERL and ERM guideline value and similar observations were made for Ni (95% of the sites) and Cd (75% of the sites). Hence, these sites can be said to have an intermediate or moderate rank of toxicity for living things.

3.5.2. Risk assessment code (RAC)

There have been numerous risk assessment methods available such as geo-accumulation index and SQGs but none of these methods take into account chemical speciation. However, RAC provides a better interpretation of the relationship between the bioavailable fraction and metal mobility (Liang et al., 2018). RAC is the assessment based on the strength of the bond between the metals and other components of the sediment and their ability to be released from the sediments and enter the food chain. RAC assesses this by applying a scale to the percentage of the metals in the carbonate and exchangeable fractions i.e. F1 (See Supplementary Material 1). It is the only fraction that consisted of metals to be weakly bound in the exchangeable and carbonate form and can easily equilibrate with the overlying water and rapidly become bioavailable. On the other hand, F2 consisted of the metals which will only remobilize under reducing conditions and then become bioavailable. F3 consisted of the metals which can only remobilize under oxidizing conditions while F4 consisted of the metals which cannot remobilize at all (Budiyanto and Hindarti, 2018; Nemati et al., 2011).

The RAC criteria given in Table 8 were used to assess the potential risk of the metals in Laucala Bay. The study confirmed that there is no risk of bioavailability of Cd and Pb from the sediments of Laucala Bay. According to data in Supplementary Material 1 except for site 8, the remaining sites had Cu in the range of 11–30% in F1. Hence, Cu is considered to be at a medium risk of bioavailability. However, there is a high probability of Cu becoming at high risk of bioavailability since these sites are subject to increased anthropogenic sources of pollution. Site 8 had about 45% of Cu present in F1 which indicated a very high risk of bioavailability because this site (Nasinu River mouth) has been much polluted as well.

For Cr, except for sites 12, 18 and 19 which showed a low risk of bioavailability, the rest of the sites had Cr between 11 and 30% which is indicative of a medium risk of bioavailability (Table 9), although as per SQGs the concentration of Cr was below ERL. Similarly, Fe was seen to be between 1 and 10% in F1 at sites 1, 2, 3, 4, 5, 6, 7, 8, 10 and 19 which confirmed low risk of bioavailability of Fe. However, Fe was between 11 and 30% at the remaining sites studied which indicated a medium risk of Fe bioavailability. Ni and Zn was present between 11 and 30% in F1 at all the sites studied which indicated a medium risk of bioavailability.

4. Conclusions

Analyses of metals in sediment samples from Laucala Bay indicate a significant increase in metal contamination. At some sites, an increase by $> 500\%$ were noted as compared to the baseline study carried out almost 25 years ago. Rapid urbanization in approximately last three decades have led to an increase in formal and informal settlements that have dire consequences on solid waste management and wastewater treatment. The multivariate analyses showed that Cu, Cd and Pb were clustered into one component confirming the discharge of wastewater effluent into Laucala Bay as a major point source. This was further supported by EF, I_{geo} and correlation matrix values that the source of Cd, Cu and Pb was mainly anthropogenic and moderately polluted. Due to the persistence nature of metals it tends to accumulate in the sediments and therefore it can be concluded that long term discharge of

Table 8
Guideline values of ERL and ERM.

Heavy metal	ERL (mg/kg)	ERM (mg/kg)
Cd	1.2	9.6
Cr	81	370
Cu	34	270
Pb	46.7	218
Ni	20.9	51.6
Zn	150	410

Table 9
Criteria for risk assessment code (Rodríguez et al., 2009).

Criteria (%)	RAC
< 1	No risk
1–10	Low risk
11–30	Medium risk
31–50	High risk
> 50	Very High risk

wastewater without any treatment method for removal of metals has led to contamination of Cu, Cd and Pb. Also, the concentration of Cr, Fe and Zn did not differ significantly between the KWWTP and the river mouths and close to the land sites but it differed significantly between KWWTP and sites away from the land-close to the reef. However as per RAC, Cd and Pb does not pose any risk to environment, however Cr, Cu, Ni, Fe and Zn had certain concentrations bound to the exchangeable and carbonate fractions and were classified as medium risk of toxicity. This study suggested that constant monitoring of the metals concentration in Laucala Bay is extremely important and stringent mitigation options needs to be implemented to strengthen the solid waste management and waste-water treatment practices in Suva.

CRediT authorship contribution statement

Arsita Pratap: Conceptualization, Methodology, Investigation, Writing - original draft, Visualization. **Francis S. Mani**: Conceptualization, Methodology, Investigation, Writing - review & editing, Supervision, Funding acquisition, Project administration. **Surendra Prasad**: Writing - review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

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