



Metal pollution in sediments and bivalves in Marovo Lagoon, Solomon Islands

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ABSTRACT

Heavy metal concentrations were determined in sediment and bivalve samples from Marovo Lagoon in the Solomon Islands. In the sediments, heavy metal levels ranged from 10 ± 3 – 47 ± 2 $\mu\text{g/g}$ Cd, 25 ± 4 – 351 ± 5 $\mu\text{g/g}$ Cr, 145 ± 3 – 418 ± 7 $\mu\text{g/g}$ Cu and 20 ± 3 – 371 ± 5 $\mu\text{g/g}$ Pb. When compared to the baseline values measured in a 1991 study of the same lagoon, a simple relative ratio in the range of 2–43 was noted for all metals in sediments as compared to baseline and confirms significant anthropogenic influence. The heavy metal contamination of bivalves showed level ranges of 2.00 ± 0.01 – 10 ± 1 $\mu\text{g/g}$ Cd, 9 ± 3 – 42 ± 2 $\mu\text{g/g}$ Cr, 47 ± 3 – 76 ± 3 $\mu\text{g/g}$ Cu and 24 ± 11 – 86 ± 14 $\mu\text{g/g}$ Pb. The higher levels of metals in the Marovo Lagoon sediments when compared to other Pacific studies are attributed to intense logging activities around the bay and other potential anthropogenic sources such as mining and discharge of waste into the lagoon.

The persistent character of heavy metals in the environment is of great concern, due to their long lasting toxic effects (Gangaiya et al., 2001). The marine sediment is an important scavenger of metals that acts as a reservoir in the aquatic systems, accumulating more metals than the surrounding water. The marine sediment regulates the mobility of a wide variety of contaminants to the water column in an unperturbed environment. The assessment of heavy metal pollution in sediments have been carried out using different pollution indices, however a more complete assessment of heavy metals can be undertaken when bio-indicators are included in the assessment. When dealing with heavy metals in bio-indicators in the marine environment, it is important to note that, the stationary sedentary organisms are more advantageous to use than the mobile organisms such as fish. Sedentary organisms such as bivalves are widely used as metal pollution indicators, due to a range of biological characteristics that can include, filter feeding behavior and potential to bioaccumulate chemical pollutants including metals (Zhou et al., 2008; Bonacci et al., 2006).

Contamination of coastal environments by heavy metals is a major worldwide concern. Besides Australia and New Zealand there have been studies on heavy metal contamination in marine sediments in the Pacific island countries such as Fiji (Collen et al., 2011; Denton et al., 2001; Morrison et al., 2001; Maata and Singh, 2008; Chand et al., 2011; Pratap et al., 2020), Solomon Islands (Naidu et al., 1991), Guam (Denton et al.,

2006; Denton et al., 1997), Samoa (Imo et al., 2014), Tonga (Morrison and Brown, 2003), French Polynesia (Besson et al., 2020) and Tahiti (Besson et al., 2020). Some of these studies conducted in the Pacific island countries established a baseline survey (Besson et al., 2020; Morrison & Morrison and Brown, 2003; Naidu et al., 1991). Most of the studies conducted in the South Pacific region engaged in spatial scale monitoring which showed enrichment of heavy metals due to anthropogenic activities around the local coastal marine environment (Denton et al., 2005; Collen et al., 2011; Pratap et al., 2020; Denton et al., 2006; Denton et al., 2007; Imo et al., 2014; Maata and Singh, 2008; Morrison et al., 2001) and suggested regular monitoring (Chand et al., 2011; Pratap et al., 2020; Denton et al., 2006; Imo et al., 2014).

The strong association of the South Pacific Island people to their surrounding waters is one important aspect of their life and culture that has been going on for many years. The people from local villages rely on the marine resources for their daily protein and income. Though, this strong relationship was established between people and their surrounding waters, the knowledge on heavy metal contamination in marine environment and the health risk they could pose to the local community through the bio-magnification process in the food chain are very limited (Boboria, 2020). Therefore, it is important to monitor and carry out assessments of the risk that heavy metal contamination may play, and improve the communication of these findings back to the local

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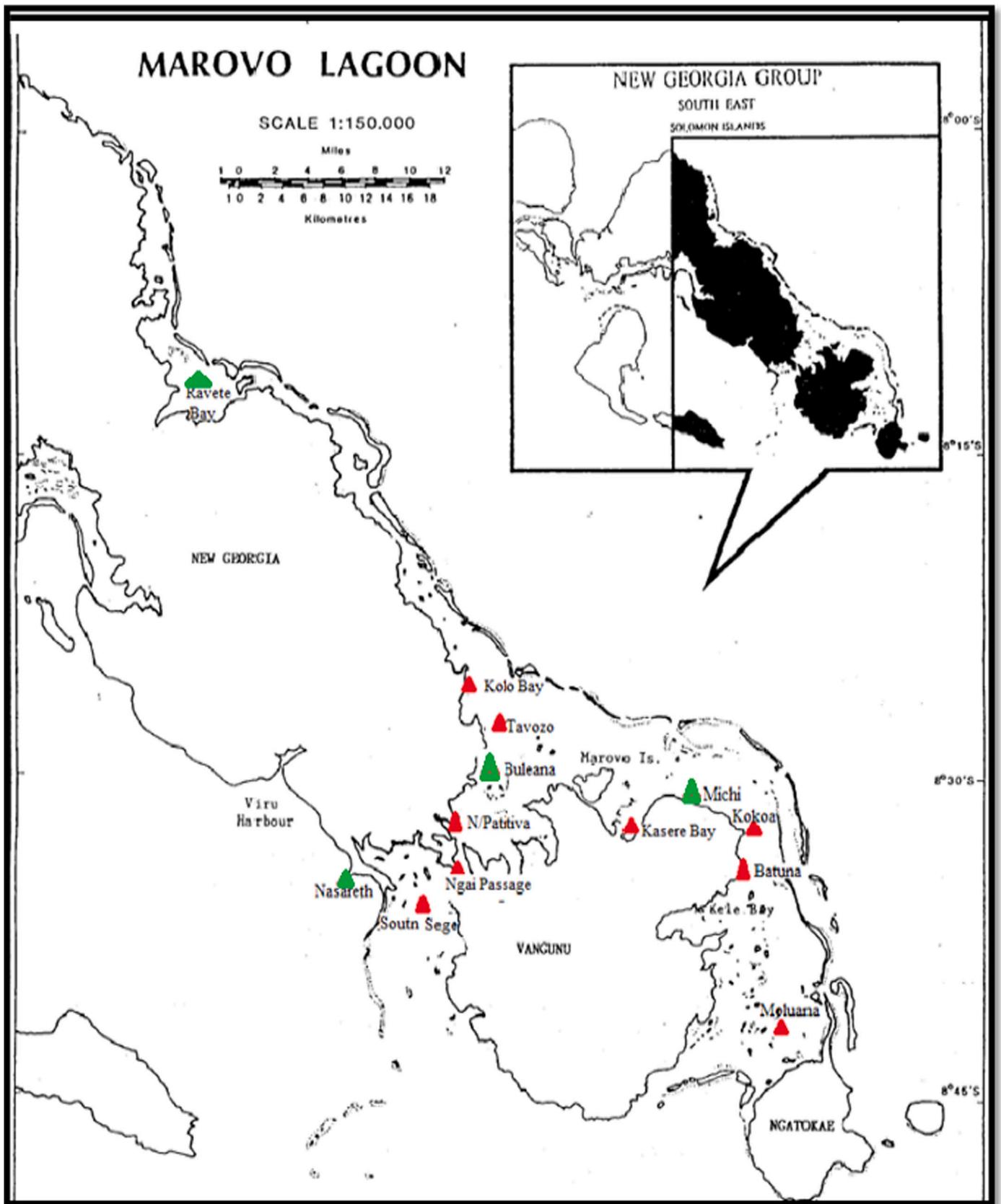


Fig. 1. Showing sampling sites for sediments and bivalves in the lagoon. Green triangles are bivalve sampling sites and red triangles are sediment sampling sites. Sediment and bivalves were both collected from Kokoa. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

community and environmental managers.

Marovo Lagoon is the largest lagoon in the world that is located in the western part of Solomon Islands and connects the coast of three large volcanic Islands: New Georgia, Vangunu and Gatokai (Fig. 1). The islands around the lagoon are rich in inland rivers that feed the lagoons with freshwater. It was once a world heritage site with an area of 700 km² (Stoddard, 1999). The scattered 200 islets and a raised well-defined double barrier reef which indicated that the islands have been undergoing two subduction processes over the different geological times in the past, highlighting the fascinating feature of the lagoon (Ministry of Environment Conservation and Meteorology, 2008; Hviding, 2008; SPREP, 2011). It was reported by Duke et al. (2007) that most of the hydrological systems in the lagoon including chemical, biological and physical set up in the lagoon are being highly influenced by this great double barrier reef. Due to the double barrier reef systems and its inability to allow the proper mixing and flushing of contaminants in the lagoon; the area has experience great stress from the increasing population and human activities around the lagoon (Love and Ross, 2005).

The lagoon lost its heritage status due to degradation of the tropical rain forest by intense logging activities in the area and depletion of marine stocks resources by intense fishing activities (Pauku, 2009). The anthropogenic activities and mostly logging by foreign companies around the lagoon exhausted about 80% of the forest which led to reduction of biodiversity both inland and the marine environments. The activity caused some of the main freshwater streams in the area to dry up as well as causing deterioration of natural flora and fauna in the lagoon by altering the physical and chemical parameters of rivers and streams with high eroded soil (Khatri and Tyagi, 2015). High sedimentation and soil erosion are also reported commonly around the lagoon (Kinch et al., 2006). Besides logging, mining activities for gold and copper was also conducted in the area with a lot of drilling activities in the surrounding hills. The Copper (Cu) and gold (Au) deposits are reported high in the area (Lum, 1993). Following the extensive logging activity around 800 ha of logged areas were planted with palm oil trees. This oil palm plantation required high amount of fertilizers especially in steep slope areas. The coral bleaching reported around the lagoon are expected from waste from oil palm plantation and soil erosion entering the lagoon (Duke et al., 2007). In addition to coral bleaching the lagoon has witnessed fish kill events linked to algal blooms including the growth of the toxic dinoflagellate and diatom pseudo-nitzschia species (Albert et al., 2013).

A baseline study of heavy metal contamination at Marovo Lagoon was conducted by Naidu et al. (1991) as part of a regional study which recommended further work was required to monitor the increasing levels of anthropogenic activity. This current paper reports the results of investigations carried out in 2014 at Marovo Lagoon on the extent of contamination of sediments by heavy metals such as chromium (Cr), lead (Pb), copper (Cu), and cadmium (Cd) and the data compared to the previous values obtained by Naidu et al. (1991). To complement the sediment work, accumulation in the bivalve *Tridacna crocea* (*T. crocea*) was conducted, a species previously shown to be a useful indicator in studies monitoring metal accumulation (Astudillo et al., 2005). This study also focused on the major sources of heavy metal contamination in the lagoon with the aid of cluster and PCA analysis as well as correlation of metals in the sediments and bivalves to see whether there is any significant relationships exist between the two parameters.

Composite sediment samples were collected from the same 9 sites as in the 1991 study. However, the bivalve samples (n = 35) were collected randomly at 5 newly selected sites where the *T. crocea* species can be found in the vicinity of the sediment sampling sites. These reefs were about 30–50 m away from the sediment sampling stations and it is assumed that both sites share similar physicochemical characteristics. The sampling sites were mainly around the areas where a fish kill occurred in 2011 (Fig. 1). The sediment and bivalve samples (200 g) were collected in polyethylene bags by divers and the samples were exported in ice box to the lab for preservation at –20 °C (Sultan et al.,

Table 1

Mean metal concentrations in sediments for all investigated sites obtained in this study and the baseline study (Naidu et al., 1991). The metal concentrations in both studies are expressed as µg/g dry weight.

Names of sites	This study				Baseline data (Naidu et al., 1991)			
	Cu	Pb	Cr	Cd	Cu	Pb	Cr	Cd
Tavoza	358 ± 7	300 ± 3	218 ± 5	28 ± 3	NS	–	–	–
Batuna	228 ± 4	107 ± 5	75 ± 5	34 ± 3	6.21	79.5	1.1	2.11
Kasere Bay	405 ± 4	121 ± 6	163 ± 4	27 ± 3	164	18.7	18.7	0.81
Kokoa	226 ± 4	159 ± 6	94 ± 5	34 ± 3	NS	–	–	–
North Patutiva	197 ± 4	371 ± 5	151 ± 2	38 ± 3	11.2	53.1	61.2	1.6
South Sege	145 ± 3	75 ± 4	75 ± 4	47 ± 2	16.5	56.2	29.3	1.1
Ngai Passage	326 ± 7	89 ± 6	351 ± 5	31 ± 3	29.4	53.1	86.2	1.1
Kolo Bay	418 ± 7	133 ± 3	294 ± 5	25 ± 3	49.4	34.3	78.7	0.8
Moluana	162 ± 3	20 ± 3	25 ± 4	10 ± 3	NS	–	–	–
Average	274	153	161	30	46	49	46	1
% increase since 1991	494	211	251	2340				

NS: New site.

2011; Maata and Singh, 2008; Young and Ross, 2001; Morrison et al., 2001). Prior to exportation of bivalve samples for preservation, the soft tissues were removed with plastic knives and packed into polyethylene bags. Some physical parameters and chemical parameters of water including pH, dissolved oxygen, turbidity, temperature and salinity were also measured at each site with their respective calibrated meters.

A 1.0 g air-dried sediment were disaggregated and 10.0 g freeze-dried bivalve samples were homogenized separately with a mortar and pestle in the laboratory. The sediment samples were sieved using a 63 µm mesh and was pulverized prior to acid digestions (HNO₃/HCl, 1:3 v/v). The bivalve digestions used 1:1 perchloric acid (HClO₄) and nitric acid (HNO₃) (Csuros and Csuros, 2002). Both digestions were cooled to room temperature and filtered prior to AAS analysis. Samples were acid digested and analysed in triplicates.

The consistency and accuracy of the results were checked by analyzing the marine sediment certified reference material for trace metals (PACS-1), and also performing the percentage recovery for the metals. Sample blanks were repetitively analysed after the analysis of every batch of 20 samples. The LOD was also calculated for each metal analysed. The results obtained shows an excellent agreement with the certified reference material and the percentage recoveries were between 90 and 110%. The variation in triplicate sample analysis was usually within ±5%.

The concentrations of the metals in Marovo Lagoon sediments appeared in the following ranges: 10 ± 3–47 ± 2 µg/g Cd, 25 ± 4–351 ± 5 µg/g Cr, 145 ± 3–418 ± 7 µg/g Cu and 20 ± 3–371 ± 5 µg/g Pb dry weight (see Table 1). The metal abundance for all sites in the lagoon was: Cu > Cr > Pb > Cd indicating Cu as the most abundant metal in the lagoon. The highest levels of the Cr and Cu were found at Kolo and Kasere Bays with the respective concentrations of 294 ± 5 and 163 ± 4 µg/g for Cr, 418 ± 7 and 405 ± 4 µg/g for Cu and moderate levels of other metals were observed. The two sites are located right within the vicinity of a copper mine prospecting area, which is a potential suggested source of heavy metals at both sites (Bedner, 2007).

Based on the total metal assessment the order of the contaminated sites is as follows: Tavoza > Kolo Bay > Ngai Passage > North Patutiva > Kasere > Kokoa > Batuna > South Sege, indicating Tavoza to be the

most contaminated site in the area. Based on the total metal trend above, it was suggested that the metal contamination at Ngai Passage also came from the mining activities just like Kolo and Kasere Bay that lies within the same vicinity of the mining zone (see Fig. 1). The distribution of metals between these sites was aided through a huge river (Kele River) that runs through the mining areas. This river is rich in minerals deposits and a lot of gold panning and drilling activities took place in the upper and downstream of the river by different prospecting companies and the local villages around the lagoon (Bedner, 2007). Tavozo is far away from mining activities and its highest levels of metal contamination could probably suggest another highly influential anthropogenic source like logging activities.

An interesting finding is the high level of Pb ($371 \pm 5 \mu\text{g/g}$) and Cd ($38 \pm 3 \mu\text{g/g}$) with the moderate levels of Cu ($197 \pm 4 \mu\text{g/g}$) and Cr ($151 \pm 2 \mu\text{g/g}$) at North Patutiva, an isolated site without much anthropogenic perturbation. This site was chosen as a reference site by Naidu et al. (1991) due to its remoteness from human influences. Although the primary source of Pb is unknown at this site, its close proximity to the active Kavachi submarine volcano could be a contributory factor. A weak hydrodynamic environment that does not actively disperse contaminants that were being deposited by the current turbulences in the area could attribute to heavy metal accumulation of metals at this site (Duke et al., 2007; Nagajyoti et al., 2010). In addition to submarine volcanoes, high levels of Pb and Cd at this site could be attributed to three possible sources such as: (1) mining activities (2) high sedimentation from the degraded logged soil and (3) fishing and boating activities. Boating activities may be another relatively minor source of metals into these receiving environments as previously suggested by Cuong et al. (2011) and Zahir et al. (2012).

The difference in Pb and Cd concentrations for Kasere Bay, Kolo Bay and North Patutiva can be further explained by the hydrogeology features for each site such as high organic content and the sediment composition (Abdallah, 2007). Kolo Bay and North Patutiva have a great depth factor that experienced a poor mixing rate by wind, hence leaving more metals trapped and locked in the bottom sediments (Duke et al., 2007). Marovo Lagoon is typically an enclosed wind-influenced lagoon rather than current mixing (Duke et al., 2007), hence deep profile sites like, that of North Patutiva and Kolo Bay are expected to show elevated metal levels due to the insufficient wind mixing and addition by hydrological means to the already elevated metal levels at these sites (Duke et al., 2007). The redistribution or resuspension of pollutants in the sediments or dispersal in deep waters are not affected by waves that are influenced by strong winds. In addition, the differences in levels of metals at the two sites (North Patutiva and Kasere) could be also explained in terms of the differences in depth profiles and organic content between the two sites. North Patutiva has a great depth profile and probably high organic content than Kasere that experienced a poor mixing rate by wind and consequently less dispersal rate. Hence leaving more metals trapped and locked in the high organic content in the bottom sediments whereas Kasere Bay that is lower in depth profile and has less organic content that tend to release more metals into water shed (Abdallah, 2007). Similarly, most sites in the lagoon such as Batuna have high levels of clay content that can trap more metals in the sediment (Abdallah, 2007; Olade, 1987).

The high levels of total metal at Tavozo and the moderate levels at Kokoa and Batuna were indication of high sedimentation from the logged soils as they lie within the heavily impacted logging sites where high sedimentations were commonly reported. A study conducted by Wall and Hansell (1973) on some soils around some parts in Solomon Islands, including the investigated area, had revealed the Cu and Pb content in soils to have ranged from 103 to 162 $\mu\text{g/g}$ and 360 $\mu\text{g/g}$ respectively. The existence of such high levels of Cu and Pb in the soil and the high sedimentation from the 80% logged forestry around the lagoon was the possible major source of these metals in the lagoon. Therefore, it is concluded that the sediment enriched with Cu and Pb found in this study is from the high weathering of logged forest soil and mining activities

Table 2

Comparing the current data with previous studies in Pacific Islands.

Sources/location	Cr ($\mu\text{g/g}$)	Cu ($\mu\text{g/g}$)	Pb ($\mu\text{g/g}$)	Cd ($\mu\text{g/g}$)
Present study (2014), Marovo Lagoon, Solomon Islands	25 ± 4 –351 ± 5	145 ± 3 –418 ± 7	20 ± 3 –371 ± 5	10 ± 3 –47 ± 2
Naidu et al., 1991, Marovo Lagoon, Solomon Islands	1.10–86.22	6.21–164	18.7–79.52	0.800–2.11
Denton et al., 2006, Agana Boat Basin (site 2), Guam	26.4–31.21	48.0–96.11	54.6–113	< 0.13–0.27
Collen et al., 2011, Suva Harbour, Fiji	26.2	37.21	13.43	NM
Imo et al., 2014, Samoa	NM	0.97–3.82	1.23–2.82	NM
Denton et al., 2006, Saipan Lagoon (site 14)	8.84–11.0	25.61–32.52	36.1–43.42	0.54–0.61
Maata and Singh, 2008, Suva Harbour, Fiji	NM	21.41–143	22.1–93.52	NM
Denton et al., 2001, Suva Harbour, Fiji (near battery factory)	16–106	64–1151	19.3–272	0.8–198
Morrison et al., 2001, Lami estuary, Fiji	NM	5–1980	4.20–516	NM
Collen et al., 2011, Carbonate reef flat, Fiji	63.44	46.9	13.43	NM
Morrison and Brown, 2003, Fanga'uta Lagoon, Tonga	NM	15	8	<0.11
Denton et al., 1997, outer/inner Apra Harbour, Guam	64.0–129	320–1435	142–395	3.50–6

NM – Not Measured.

around the lagoon.

Enclosed lagoons, such as Marovo are typically subject to enhanced human influences due to the restricted transport, and dispersal of contaminants (Maata and Singh, 2008). The other sites, Tavozo ($358 \pm 7 \mu\text{g/g}$ Cu, $300 \pm 3 \mu\text{g/g}$ Pb, $218 \pm 5 \mu\text{g/g}$ Cr and $28 \pm 3 \mu\text{g/g}$ Cd) and Kokoa ($226 \pm 4 \mu\text{g/g}$ Cu; $159 \pm 6 \mu\text{g/g}$ Pb; $94 \pm 5 \mu\text{g/g}$ Cr; $34 \pm 3 \mu\text{g/g}$ Cd) were experiencing low hydrodynamic and poor mixing rate of contaminants between the open sea and the lagoonal water due to the restriction of the double barrier reef that surrounds the area thus enhancing metal deposition in sediments (Duke et al., 2007). Batuna and South Sege (Table 1) are the only urban sites in the lagoon with numerous human based activities. A report by Wall and Hansell (1973) indicated high level of Cd in soil around the lagoon (40 $\mu\text{g/g}$). Sedimentation was often reported by local people around South Sege, Batuna, Kokoa, Tavozo and Ngai Passage in the lagoon (Nagajyoti et al., 2010; Talley, 2002). The high sedimentation of these degraded soils is the possible major source of high Cd and other heavy metals at these sites.

The total metal average data obtained in this study was compared with the previous baseline study by Naidu et al. (1991) as given in Table 1. To understand the changes in metal concentration better, a simple ratio was calculated (metal concentration in this study at a particular site n was divided by metal concentration in the same site in Naidu et al., 1991). A ratio of 1 would indicate no change at all. However, the calculated ratios were in range of 2–43 indicating insignificant increases of metals over the years. The comparison between the Naidu et al. (1991) and the current study has some limitations due to different

Table 3

Igeo values for metals at different sites in the Marovo Lagoon. Class 0 = (no contamination; Igeo < 0); 1 = (none to medium contamination; 0 < Igeo < 1); 2 = (Moderate contamination; 1 < Igeo < 2); 3 = (moderately to strongly polluted; 2 < Igeo < 3); 4 = (heavily contaminated; 3 < Igeo < 4); 5 = (heavily to extremely contaminated; 4 < Igeo < 5); 6 = (extremely contaminated; Igeo > 5).

Sampling sites								
Metal	Batuna	Kasere	N. Patutiva	South Sege	Kokoa	Kolo Bay	Ngai Passage	Tavozo
Igeo values								
Cr	3.65	3.99	3.96	3.65	3.75	4.25	4.32	4.12
Pb	3.15	3.21	3.69	3	3.33	3.25	3.07	3.60
Cu	3.83	4.08	3.77	3.64	3.83	4.10	3.99	4.03
Cd	0.83	0.73	0.88	0.97	0.83	0.70	0.79	0.75

methodologies but it is assumed that the differences are not significant to introduce any artefacts in the interpretation. While Naidu et al. (1991) used hydrofluoric (HF) acid for digestion whereas the current study used aqua regia for digestion. Das and Ting (2017) showed that using HF alone did not result in complete metal solubilisation but combined with a strong acid or using aqua regia resulted in higher recoveries of 10–21% for heavy metals. In addition to that, HF is not often used in metal extractions due to its most corrosive behavior but aqua regia is commonly used in such studies (Turek et al., 2019; Hseu et al., 2002).

A Wilcoxon test of the two datasets and the result obtained has shown that the increase noticed in current dataset shows statistically significant increase for all heavy metals in Marovo Lagoon over the years with p value < 0.05. It is evident that the anthropogenic activities such as mining and deforestation have impacted the total heavy metal concentration of the lagoon. The high levels of heavy metals in the lagoon should further be considered seriously by the Government as they could threaten the economic zones such as reefs and intertidal zones (Duke et al., 2007).

The data obtained in this study was compared with various other aquatic heavy metal contamination studies in the Pacific. It was highlighted that the lower limit of heavy metal measured is higher than the lower limit of heavy metal observed in other studies except for Cu and Pb in Apra Harbour, Guam (see Table 2). However, it should be noted that Cr was highest in the present study while Pb and Cu were highest in Lami Estuary, Suva, Fiji (Morrison et al., 2001) which is located in one of the industrialized areas in Suva with possible industrial discharge directly into the river due to absence of any legislations in 1990s. The significant enrichment of several metals at Apra Harbour particularly Pb and Cu is due to the close vicinity of the sites to the hotel, wharf, commercial port and Dry Dock Island (Denton et al., 2005).

Igeo analysis was carried out to assess the contamination of lagoon by heavy metals with respect to the background natural levels of the elements. It is difficult to make an ideal comparison for natural levels when there is lack of data on natural background values of heavy metals in the lagoon and the background values stated by Naidu et al. (1991) is based on a very small dataset. To compensate for this, the average shale values were used. In the Igeo expression, the 1.5 constant was considered by Muller (1969) to account for the possible variations in the background values due to natural processes (Groot et al., 1982).

Muller's expression was used in the Igeo calculation;

$$I_{geo} = \log_2 (C_n / 1.5 B_n)$$

where C_n is the concentration of the investigated metal (n) in sediment and B_n is the background level of the same metal. For the purpose of this work, the average shale values (Cr = 90 µg/g, Cd = 0.3 µg/g, Cu = 45 µg/g and Pb = 20 µg/g) were used as the background values in the Igeo calculation (Turekian and Wedepohl, 1961) in the absence of the site specific background natural levels.

From the Igeo results (see Table 3), Cu, Cr and Pb were shown to be the most contaminated metal in the Marovo Lagoon. The Igeo values for the above three metals were in the following ranges 3 < Igeo < 4 and 4 < Igeo < 5 for all sites in the lagoon, corresponding to strongly polluted

Table 4

Showing the USA and Canadian Sediment Guidelines.

Guideline values for heavy metals of USA Guideline and Canadian Sediment Quality Guidelines				
Metal	USA Guidelines		Canadian Guidelines	
	ERL	ERM	TEL	PEL
Cd	1.2	9.6	0.7	4.2
Cr	81	370	52.3	160
Cu	34	270	18.7	108
Pb	46.7	218	30.2	112

Effects range–low: ERL; Effects range– median. ERM; Threshold effect level: TEL; probable effects level: PEL (µg/g wet).

and strong to very strong contamination on Igeo scale. The increased Igeo results in the present study does point to contamination of the lagoon due to anthropogenic sources. While Cd in the sediment appeared as the least contaminated metal for all sites in the harbour with 0 < Igeo < 1 (uncontaminated to moderately contaminated). The Igeo values exceeding 3 and 4 for the Marovo sediments showed that the lagoon is not healthy, a typical reflection of a very strong contamination. Generally, Igeo values showed that the lagoon needs more attention and should be given special consideration once the increased heavy metal contamination in the lagoon is to be overcome in the future.

Sediment Quality Guidelines (SQGs) were employed to assess the ecological risk of the lagoon by comparison of the present mean values to the USA and Canadian marine SQGs (MacDonald and Ingersoll, 2002) (see Table 4). The SQGs is used as a screening tool in this study as there are limitations associated with this risk assessment technique. The guidelines do not consider the spatial and temporal variability, grain

Table 5

Levels of metals measured in soft tissues of *T. crocea* and expressed as µg/g dry weight. The mean values obtained for metals in bivalves were compared to WHO and EPA standards which are the maximum concentrations permissible in bivalves for safe human consumption.

Names of site	Cu	Pb	Cr	Cd	Total mean metals
Buleana	76 ± 3	86 ± 14	38 ± 3	10 ± 1	53
Nazareth	68 ± 2	28 ± 14	9 ± 3	7 ± 2	28
Kokoa	64 ± 3	28 ± 13	24 ± 3	8 ± 2	31
Michi	47 ± 3	24 ± 11	42 ± 2	3 ± 1	29
Ravete (ref)	47 ± 3	43 ± 14	28 ± 3	2 ± 0.6	30
Mean	60	42	28	6	
WHO, 1982 (µg/g)	10	–	–	2	
European Union (EU), 2006 ^a (µg/g)	–	1.5	1	1	
FAO, 1983 (µg/g)	–	–	1	2	

^a European Commission Regulation (EC No 188/2006) (2006) concerning setting maximum levels for certain contaminants in bivalves (µg/g wet weight).

Table 6

Comparison of heavy metal in bivalves obtained in this study with data from other Pacific Islands and developed countries. The data is presented as $\mu\text{g/g}$ dry weight.

Country	Cr ($\mu\text{g/g}$)	Pb ($\mu\text{g/g}$)	Cu ($\mu\text{g/g}$)	Cd ($\mu\text{g/g}$)	Reference
Tanapag Lagoon, Saipan	11.9–12.2	68.1–102	26.5–73.3	0.62–0.70	Denton et al., 2006
Fiji	–	0.450–0.900	–	–	Mosley and Aalbersberg, 2003
New Caledonia (Grande Rade)	5.8–12.1	–	77.3–146	0.21–1.30	Hedouin et al., 2009
Pango Bay (Guam) (Quidnipagus palatum)	<0.12–0.6	<0.20–0.89	4.26–68.5	<0.13	Denton et al., 2007
Croatia	1–2.9	2–7	3.7–11.1	–	Otchere, 2003
Italy	–	2.0–9.0	17.9–156	0.16–1.0	
Slovenia	2.7–10.3	0.79–11.5	6.5–49	0.44–1.09	
Turkey	1–3	5–21	90–260	2–4	
Morocco	1.9–28.9	0.1–26.45	4.1–43.1	2.12–34.71	
Spain	–	0.52–8.22	4.42–9.65	4.42–9.65	
Spain	2.2–45.7	0.3–6.1	6.8–29.9	0.2–0.77	
Present study (Marovo Lagoon)	9 ± 3 – 42 ± 2	24 ± 11 – 86 ± 14	47 ± 3 – 76 ± 3	2 ± 0.6 – 10 ± 1	

size dependency, sediment chemistry and bioavailability of contaminants to provide an accurate assessment of risk (Burton Jr., 2002).

The guidelines provide two values, Effects Range Low (ERL) and Effects Range Medium (ERM), which are an estimate concentration of heavy metals below which toxicity is least likely to occur or most like to occur (Long et al., 1995). Similar analogy is applied in Canadian SQGs with two values Threshold Effect level (TEL) and Probable Effect level (PEL). The contamination levels for each metal in the lagoon sediments with respect to the SQGs (ERL, ERM, TEL and PEL) were as follows: Cd showed 100% above all the SQGs; Pb showed 100% above ERL and TEL, 38% and 22% above PEL and ERM respectively; Cr showed 100% above TEL, 38% above ERL and 44% above PEL; Cu exhibited 100% above ERL, TEL and PEL, 44% above ERM. The data reaffirms the contaminated nature of sediments in the lagoon which reflect the influence of anthropogenic activities in the area.

This is a novel study utilizing *T. crocea* as bio-indicator of metal pollution in the area. It is commonly consumed by the local people of Marovo Lagoon. Only 35 samples were collected from 5 sites for analysis (see Fig. 1) however, we recommended that additional samples should be collected from other sites in the lagoon to provide a more comprehensive study. The data showed heavy metals in soft tissues in the order of: $\text{Cu} > \text{Pb} > \text{Cr} > \text{Cd}$ and with an average of $60 \mu\text{g/g}$ Cu, $42 \mu\text{g/g}$ Pb, $28 \mu\text{g/g}$ Cr and $6 \mu\text{g/g}$ Cd (see Table 5) with Cu claiming the highest mean level in the tissue, probably from natural Cu deposit and the fast uptake of Cu by organisms as an essential nutrient for their metabolic functions (Sultan et al., 2011; Duke et al., 2007). The uptake of metals by organisms is affected by certain factors such as salinity, seasonal factors, food intake, temperature, spawning and individual variations (Rouse et al., 1999). The uptake of metals in organisms is usually higher in low saline and high turbidity waters according to Chapman (1992). This maybe the probable explanation for the highest mean total for metals recorded for Buleana ($76 \pm 3 \mu\text{g/g}$ Cu; $86 \pm 14 \mu\text{g/g}$ Pb, $38 \pm 2 \mu\text{g/g}$ Cr and $10 \pm 1 \mu\text{g/g}$ Cd) with low salinity and high turbidity recorded for the site (data not shown here). Ravete is in the northern arms of the lagoon and is far from the research conducted area which is in the southern arms of the lagoon. Ravete which serves as the reference site for the bivalve study shows the lowest level of metals in the bivalve soft tissues except for Pb and Cr metals (see Table 5). It is rare for Ravete, a reference and remote site to claim the second highest Pb level in the study; however, conclusion was made that Ravete is located very close to any unidentified contaminated sources such as World War 2 burial grounds within the area (Muller, 1969). The heavy metal levels at Kokoa and Michi do indicate a high deposition of Cu and Cd enriched soils (103 – $162 \mu\text{g/g}$ Cu and $40 \mu\text{g/g}$ Cd) in reefs as reported by Wall and Hansell (1973) but the speculation needs to be confirmed with further research.

The mean values obtained for metals in bivalves in this study were compared to some standard guidelines set by EU, FAO and WHO which are the maximum concentrations legally permitted in bivalves for safe human consumption. Generally, the levels of heavy metals observed in

bivalves in this study exceeded the levels of standard guidelines that were given in Table 5. This suggests that the heavy metals concentrations in bivalves from Marovo Lagoon are of concern from a human health viewpoint and more detailed work is required. However, more studies need to replicated to cover more sites and should be used to inform decision makers. The heavy metal concentration in bivalves reaffirms that Marovo Lagoon is polluted from anthropogenic activities, high sedimentation and bay mineralogy leading to potential ecological impact in the area (Gangaiya et al., 2001).

Comparing the Marovo data with some Pacific data, the present study shows reasonably higher values for Cu, Cr, Pb and Cd in bivalve tissue than all the other neighboring lagoons except for Pb and Cu levels in Tanapag Lagoon and New Caledonia respectively as shown in Table 6. This indicated the ecological threat of heavy metal pollution in the Marovo Lagoon. This study also shows comparable or higher values to heavy metal pollution as recorded by bivalves for other developed countries (see Table 6). An interesting revelation is that the levels of Pb found in bivalves at Marovo Lagoon are extremely high. Although these developed countries have more industrial effluents and more population but the levels of heavy metal pollution are similar or even better than Marovo Lagoon. This points out to an important message for the developing island countries that sustainable development needs to be implemented to safeguard our aquatic environment. These high levels of metals in Marovo Lagoon are a reflection of ecological threat in the lagoon. Seeing the ecological importance of the lagoon, it is crucial to consider the well-being of the lagoon from further deterioration in the future.

Despite, the variations of metal concentrations between the investigated sites, the major influential activities that contribute metals in the lagoon were: (1) logging (2) mining, (3) agriculture and (4) boating and fishing. The logging history in the lagoon started way back in 1980s up until now. There were several logging companies that operated around the lagoon. About 80% of the rainforest across the entire Islands has been logged with deforestation becoming the major issue in the area (Pauku, 2009). These logging activities exposed more than half of the top soils in the island and caused deterioration to natural flora and fauna in the lagoon by polluting rivers and streams with contaminants from the degraded soil.

Evidences of huge sediment plumes and high sedimentation have been reported by the people of Marovo Lagoon in the vicinity of the logging areas (Lafranchi and Green Peace, 1999). Wall and Hansell (1973) had reported high levels of clay and organic matter that were rich in oxides of aluminum and iron in soils around the lagoon. The presence of high clay and organic content of soil around the lagoon tend to retain more metals in the sediments. It was reported by Wall and Hansell (1973) and Wall et al. (1979) that the soil around the lagoon is classified as an Acrorthox (a highly weathered soil). While there may be some Acrorthox in the area, it is unlikely to be dominant, given the range of soil parent materials identified around Marovo. When logging or other land vegetation disturbance occurs over a large area, the soil surface is

Table 7
Correlation matrix for metals in sediment and bivalve samples.

	Cu_sed	Pb_sed	Cr_sed	Cd_sed	Cu_bivalve	Pb_bivalve	Cr_bivalve	Cd_bivalve
Cu_sed	1.000							
Pb_sed	0.367	1.000						
Cr_sed	0.733*	0.400	1.000					
Cd_sed	-0.517	0.167	-0.200	1.000				
Cu_bivalve	0.700	0.400	0.300	-0.500	1.000			
Pb_bivalve	0.000	0.500	0.500	0.200	0.400	1.000		
Cr_bivalve	-0.300	0.600	0.300	-0.100	0.200	-0.100	1.000	
Cd_bivalve	0.900*	-0.300	0.600	-0.800	0.900*	0.300	-0.100	1.000

* Correlation is significant at the 0.05 level (2 tailed).

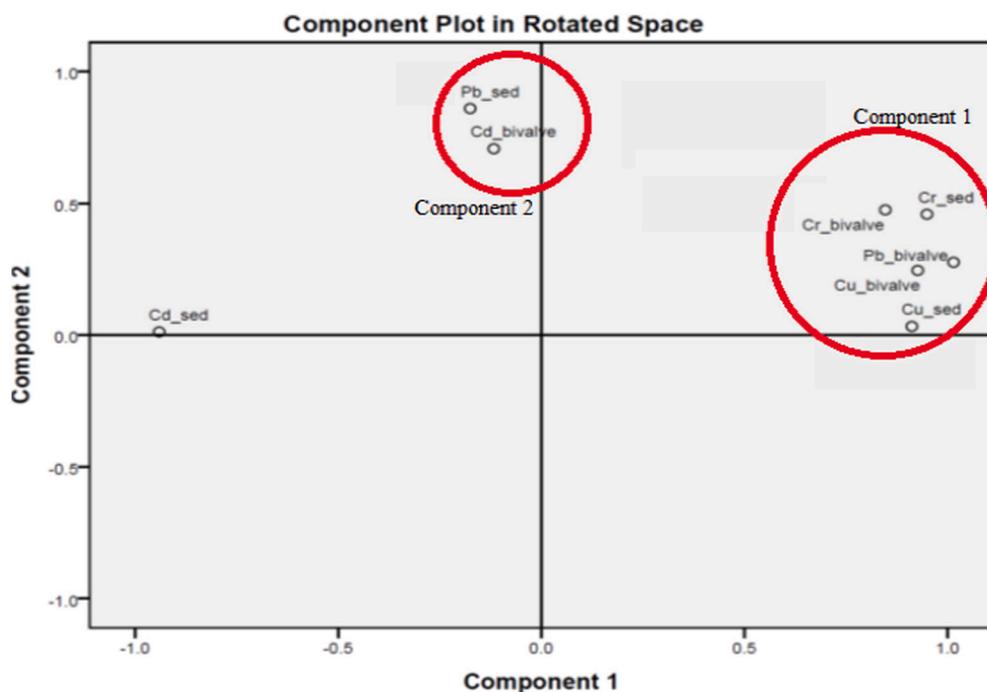


Fig. 2. The Principal Component Analysis showing the loading effect of two components, PC-1 and PC-2, on the samples.

often bare for some time and the surface soil substantially disturbed but not sufficient to produce extensive soil erosion. A soil transport medium is required and at Marovo, this is provided by rainwater and runoff. The Marovo area has annual rainfall of 3800–4000 mm per year (Maru et al., 2018), often falling as heavy rain or in thunderstorms. The water driven erosion will be enhanced by tropical cyclones and the work of Maru et al. (2018) indicates that the Western Solomon Islands region has an average of 2.5 cyclones per year. This reaffirmed a possibility that the lagoon could have received its metal loads from the erosion of soil and high sedimentation in lagoon caused by intense logging activities.

Following the complaints from the local people around the lagoon regarding the high soil erosion and the resultant siltation around the lagoon, the logging companies made agreement with the people and the Government to plant oil palm trees on these waste logged areas. The introduced oil palm station covers an area of 10,299 ha which occupies both the steeply and flat land areas (Mosley and Aalbersberg, 2003). The use of modern fertilizers was regularly applied on these plantations to promote growth on these waste lands. There is no clear indication of the quantity of fertilizers applied in the plantation yard and their associated impacts in the area. There were also evidences of algal bloom and coral bleaching near areas of these agricultural plots (Duke et al., 2007). The use of fertilizers in the lagoon would have elevated the levels of metals in the lagoon after being washed down from the forest.

The correlation patterns between the investigated elements are shown in Table 7. There is strong positive correlation between Cu, Pb

and Cr in sediment. Cadmium showed a very weak positive correlation with Pb and negative correlation with Cu and Cr. This tend to suggest that the sources of Cr and Cu are distinctively different from Cd and only similarity in source between Pb and Cd exist. This observation re-affirms the argument that high sedimentation rate of soil enriched in Pb and Cd due to logging activities could be the potential source of heavy metal contamination in Marovo lagoon.

The principal component analysis was applied on the total concentration data and only the two components with Eigen values above 1 after rotation were retained. The two principal components accounted for 83.2% of the total variance. Most of the commonalities showed 0.8 and 0.9, indicating that the extraction of the two components is satisfactory for this study.

To show the load of various variables and samples to particular principal components, the distribution of load and component matrix in the PC1-PC2 axis is indicated in Fig. 2. Fig. 2 showed component 1 which accounted for 54% of the variance among the metals in sediment and is characterized by a positive high contribution (loads) for Cr and Cu in sediments and bivalves with the respective commonalities of 0.9 and 1.0. Similarly, 29.2% of variance is accounted for the second component which represented high positive loads for Pb and Cd in sediment showing the relative commonalities of 0.9 and 0.9. In Fig. 2, PCA-1-PCA-2 grouped the variables in the geochemically reasonable interpretable manner. The loadings between the principle components and their position of variables in the coordinates showed that Cu and Cr in

sediment and bivalves have a common source of contamination which most likely to be from mining and agricultural activities around the lagoon. The second group of variables on the axis is Pb and Cd in sediment which also indicated a common source of origin probably from high sedimentation of logged damaged soil around the lagoon.

The mean levels of Cu, Pb, Cr and Cd in sediments exceeded the concentrations measured in the baseline study in 1991. This study shows a drastic increase in of heavy metal contamination over the years in contrast to the baseline study. The heavy metal pollution observed at Marovo Lagoon is serious than the other Pacific neighboring countries but are comparable to a few. The distribution of metals at all sites was variable and seems to be dispersed away from the coast. The poor hydrodynamic and geological structure such as double barrier reef affects the fate of these pollutants. The information supplied by *T. crocea* biomonitors also reaffirms the contamination of metals in sediments and is proved suitable for future studies in the lagoon and in Pacific Islands as a bio-indicator. The present study of bivalves from the lagoon was contaminated by heavy metals (Cu, Pb, Cd and Cr) in their soft tissues and exceeded the FAO, WHO and EC guidelines. Both correlation and PCA analysis conducted for heavy metals in sediments showed that Cu and Cr contamination originated from similar anthropogenic sources such as waste from mining and fertilizer applications whereas Cd and Pb resulted from high sedimentation rate from degraded soil resulting from deforestation. The heavy metal pollution of Marovo Lagoon is very critical and calls for greater public awareness and mitigation strategies should be implemented by the relevant authorities.

CRedit authorship contribution statement

Dickson Boboria: Conceptualization, Methodology, Investigation, Writing – original draft, Visualization. **Matakite Maata:** Writing – review & editing. **Francis S. Mani:** Conceptualization, Methodology, Investigation, Writing – review & editing, Supervision, Project administration.

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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