Geochemical Distribution, Enrichment, and Potential Toxicity of Trace Metals in the Surface Sediments of Okinawa Mangrove, Southwest Japan

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Abstract

Surface sediments along the coast of Okinawa Island were analyzed to determine their geochemical compositions, and to assess sediment quality and the potential for ecological harm, based on comparison with established international sediment quality guidelines. The Contamination factor (CF) and the geoaccumualtion index (*Igeo*) of As, Pb, Zn, Cu, Ni, and Cr were computed to determine the pollution status of the mangrove and foreshore sediments. The lowest effect level (LEL) and the severe effect level (SEL) established by the New York State Department of Environmental Conservation (NYSDEC), and the threshold effect level (TEL) value and the probable effect level (PEL) developed by the Canadian Council of Ministers of the Environment (CCME) were the benchmarks applied to assess the potential for ecological harm. Among the sampling set, the highest average concentration of Pb (22 mg/kg), Zn (82 mg/kg), Ni (26 mg/kg), and Cr (81 mg/kg) occurred in the Ohura mangrove sediments, that of As (17 mg/kg) in the Gesashi mangrove, and that of Cu (22 mg/kg) in the suspended solids. The loss on ignition values of the foreshore sediments (20.32%) are nearly two and three times greater than that of the Gesashi mangrove and the Ohura mangrove, respectively. On average, the CFs of As, Pb, Zn, Cu, Ni, and Cr in the suspended solids, those of Pb, Zn, Cu, Ni, and Cr in the Gesashi mangrove and foreshore sediments, and those of Ni and Cr in the Ohura mangrove display low contamination (CF≤1), whereas the average CF of As in the three sampling areas show moderate enrichment (CF:1-3). The average *Igeo* values indicate that among the selected trace metals, only As in the Gesashi and Ohura mangroves show significant values, but even these are rated as unpolluted to moderately polluted (*Igeo*:0-1). The average concentrations of As in the Gesashi and Ohura mangroves and the foreshore sediments exceed both the LEL and TEL, but fall below the SEL and PEL, suggesting that this metal may moderately impact biota health. This is also the case for Cu and Ni in the Ohura mangrove and the suspended solids, and for Cr in the Ohura mangrove.

Keywords: Trace metals, geochemistry, mangrove sediments, foreshore sediments, sediment quality, Okinawa, southwest Japan

1. Introduction

Trace metals are among the most persistent pollutants in aquatic ecosystem because of their resistance to decomposition in natural conditions (Arnason & Fletcher, 2003). Trace metals in the aquatic environment originate from both natural processes (e.g., weathering and erosion of rocks and soils) and anthropogenic activity such as agricultural runoff and sewage disposal (Liaghati et al., 2003). Due to anthropogenic activities, the concentrations of some trace metals such As, Pb, Zn, Cu, Ni, and Cr are often enriched in sediments relative to the upper continental crust (Ismail et al., 1993; Glasby et al., 2004; Shazili et al., 2006). These enriched trace metals may become a potential source of pollution.

Okinawa Island, located in southern Japan, is the largest in the Ryūkyū Islands archipelago. The island is characterized by the production of sugar cane, pineapple, papaya, and other tropical fruit. Other economic activities include sugar refining, cattle rearing, tuna fishing, and pineapple canning. Higashi village in the north-eastern part of Okinawa produces about one third of all pineapples in Okinawa, and is the largest production site in Japan (Takemitsu et al., 2005). In addition, the north-eastern part of Okinawa is characterized by a variety of coastal ecosystem environments, including beaches and mangrove forests. These coastal

ecosystems play a major role in the mobilization and distribution of trace elements which may potentially pollute or contaminate the adjacent aquatic ecosystems.

Contamination of aquatic ecosystems by trace metals can be confirmed in water, organisms, and sediments (Albering et al., 1999; Sprenke et al., 2009). Sediments are the ultimate sink for anthropogenic chemical contaminants that may be contained in effluents originating from industrial, urban, recreational, and agricultural activities (Hatji et al., 2002; Apitz et al., 2005). The geochemical analysis of sediments thus provides an insight into the pollution status of the environment with respect to diverse chemical elements such as trace metals.

Over the last few decades, many geochemical studies of coastal sediments have been carried out to evaluate the extent of contamination from trace metals (e.g. Daskalakis et al., 1995; Long et al., 1998; Ishiga et al., 2000a). Okinawa Island has been of interest to many geochemical studies over the past years (eg. Ohde et al., 2004; Naumih & Oomori, 2006; Vuai & Tokuyama, 2011). Although most of these studies have focused on trace metal pollution, investigations in the north-eastern part of Okinawa have been limited, particularly in the foreshore and mangrove environments. However, mangrove forests capture land-derived nutrients, pollutants, and suspended matter before these contaminants reach deeper water (Rivera-Monroy & Twilley, 1996; Tam & Wong, 1999). In addition, mangrove sediments are typically anaerobic and reduced, and hence are rich in sulfides and organic matter. Consequently, retention of water-borne trace metals is favoured in such sediments (Tam & Wong, 2000), and subsequent oxidation of sulfides allows metal mobilization and bioavailability (Clark et al., 1998).

The present study aims to determine trace and major element concentrations in suspended solids, mangrove sediments and foreshore sediments of Okinawa, to examine their distribution patterns and sources. These data will be used to assess the pollution status of the sediments using contamination factors and the geoaccumulation index, and to evaluate potential toxic effects of the metal concentrations on aquatic biota, by reference to established international sediment quality guidelines.

2. Study Area

Okinawa is situated to the southwest of mainland Japan (Figure 1), in the southern-most Nansei-islands chain spreading southeast from Kyushu at a latitude of 24-27°N. The population of Okinawa is a little over one million (1 130 682), but with density varying widely, from 7500 persons km^{-2} in the south to 34 persons km^{-2} in the north (West & van Woesik, 2001).

Okinawa island has a subtropical climate, in which the annual average atmospheric temperature and precipitation reach 22.7°C and 2036.9 mm, respectively (National Astronomical Observatory, 2004).

The predominant soils in Okinawa Island are red and yellow in hue (locally called kunigami mahji). Such soils are distributed in the central to northern parts of the island, and cover about 55% of the total land area of 1,225 km^2; these soils are readily eroded due to the high annual rainfall (Onaga, 1986). The northern part of Okinawa is covered with dry, fine-grained red soils. Land development such as pineapple cultivation and construction of recreational facilities increases the amount of exposed soil. Every significant rainfall carries soil to the coast, coloring the ocean red (Takemitsu et al., 2005).

The Gesashi and Ohura mangroves are situated in the northeastern part of Okinawa. The Gesashi area is located in Higashi village of Nago City, and mangrove occurs in the estuary of the Gesashi River, which flows into Arume Bay (Figure 1). The Gesashi mangrove is developed along the estuary, and widens its distribution in the watershed, particularly on the southwestern side of the river. The Gesashi mangrove has an estimated area of about 10 ha, and is the largest mangrove on Okinawa island (Okinawa Prefecture, 2015). The mangrove vegetation in this area is mainly composed of *Rhizophora stylosa* in the frontal zone (around 2 m in height). In the middle to upper zones, *Kandelia candel* (around 1.5 m) and *Bruguiera gymnorrhiza* (maximum 5 m) dominate the forest. In this mangrove, the catchment of the Gesashi River has an area of 726.83 ha, and it has been widely developed for cultivation. In addition, the mangrove is endangered by red soil inflow, and urgent countermeasures have been planned. Forest occupies 67.8% of the total catchment area, and cultivated land forms about 25.6% (Okinawa Prefecture, 2015). The total inflow of red soil has been estimated at 9,390 t/year, and some 34,860 m^3 of sediment has accumulated since 2014. Observed thickness of the red soil at the frontal part of the mangrove was 50 cm, thinning to 20 cm upstream.

The red soil is composed of very fine particles, with more than 90% less than 64 μm (clay size) in diameter (Okinawa Prefecture, 2015). Accumulation of red soil continues at present, as observed in March 2015 (Ishiga et al., 2016), coating the breathing roots of the mangroves (Okinawa Prefecture, 2015).

The Ohura district in Nago City lies in the estuary of the Ohura River, which flows into Ohura Bay (Figure 1). The outer mouth of southern Ohura Bay comprises a wide coral reef known as the Henoko coast (NACS-J, 2010).

The area of the Ohura mangrove is estimated to be about 2.5 ha. The soil in this mangrove is dark, and is rich in organic matter. *Kandelia candel* (around 2m high) and *Bruguiera gymnorrhiza* (over 4m high) are the main species which dominate in the forest.

The geology of Okinawa is characterized by Paleozoic sedimentary rocks with subordinate igneous dikes, the Shimajiri Formation of Miocene-Pliocene age, Ryukyu Limestone of upper Pliocene or lower Pleistocene age, and Recent coral reefs (Konishi et al., 1972).
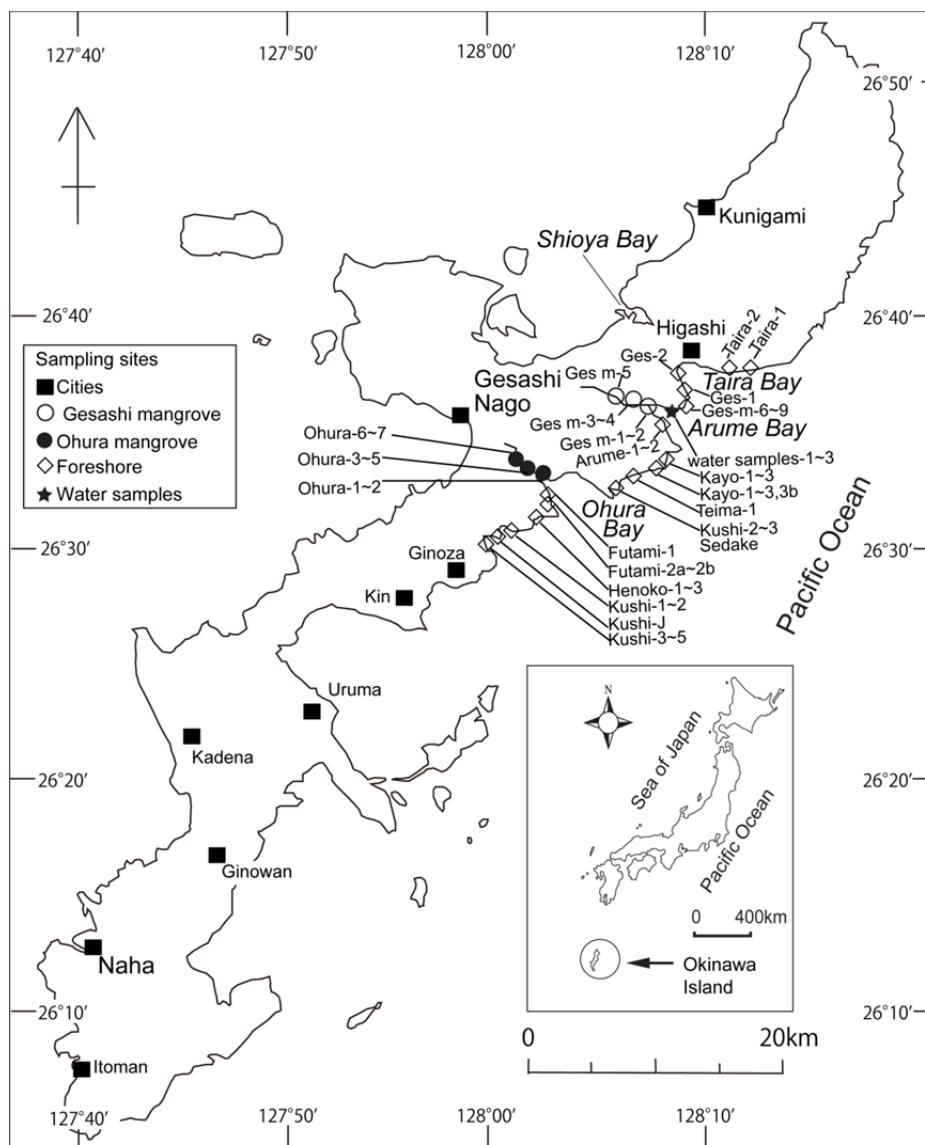
Figure 1. Locations of sampling sites of the Gesashi and Ohura mangrove sediments, and water samples from the coasts of the Okinawa Island. *Inset*, location in Japan

3. Materials and Methods

3.1 Sampling

Between March to May 2015, sediment samples were collected from the Gesashi mangrove (*n*=5), Ohura mangrove (*n*=7), and the foreshore (*n*=32) (Figure 1). In this same period, water samples were also collected at three sampling sites (five liters at each site) for determining the suspended solids. The sediment samples were collected at low tide. At each sampling site, about 200 g of the uppermost two cm of the surface sediment was collected with a plastic spatula.

3.2 Analytical procedures

Approximately 50 g of each sample were dried in an oven at 110°C for 48 h. The dried samples were then ground for 20 minutes in an automatic agate pestle and mortar grinder. The powdered samples were then compressed into briquettes using a force of 200 KN for 60 s, following the method of Ogasawara (1987). Abundances of selected major elements (TiO_2, Fe_2O_3, MnO, CaO and P_2O_5), trace elements (As, Pb, Zn, Cu, Ni, and Cr) and total sulfur (TS) in the sediments were determined by X-ray fluorescence (XRF) in the Department of Geoscience, Shimane University, using a Rigaku RIX-2000 spectrometer. Average errors for all elements are less than ±10% relative.

3.3 Statistical Analysis

Pearson's correlations were computed in Excel 2013 to determine the correlation coefficients between the elements. Hierarchical cluster analysis was performed in the PAST software package on the concentrations of As, Pb, Zn, Cu, Ni and Cr to determine the geochemical similarity between the sampling sites of the study areas.

3.4 Loss on Ignition

Loss on ignition (LOI) of the samples was determined by ignition of sub-samples in a muffle furnace at 1,050°C for 4 h. Gravimetric LOI was calculated from the net weight loss.

3.5 Suspended Solids

For the determination of the suspended solids in the coastal areas, the water samples were collected in plastic bottles and stored at 4ºC during transport to Shimane University. The water samples were then filtered using Whatman 45μm quartz filter paper. Prior to filtering, the filter papers were dried at 110ºC for at least 2h, and their weight then recorded. After filtration, the filter and retained filtrate was then oven dried at 110ºC for at least 2h, and the weight of the filtrate determined by difference. Finally, to determine the 12 selected trace and major elements, the filtered suspended solids were analyzed by X-ray fluorescence spectrometry using a Rigaku RIX-2000 spectrometer equipped with an Rh-anode tube at Shimane University.

3.6 Sediment Quality

In this study, the composition of Japan upper crust from Togashi et al. (2000) was used as reference values to evaluate the pollution status of the study areas with respect to As, Pb, Zn, Cu, Ni, and Cr. Contamination factors (CF) and the geoaccumulation index (*Igeo*) were used as sediment quality indicators.

A. Contamination factor (CF)

CFs of each sampling sites were computed according to Håkanson (1980):

$$CF = Cx/Cref \hspace{3cm} \text{Equation (1)}$$

where Cx is the concentration of the element of interest, and Cref is the reference background concentration. The classifications of the sediments for CF (Håkanson, 1980) are: ≤1=low contamination, 1–3=moderate contamination, 3–6=considerable contamination, and ≥6= extreme contamination.

B. Geoaccumulation index (Igeo)

To evaluate the pollution level in the study areas, geoaccumulation index values were determined based on Müller (1969), according to the following equation:

$$Igeo = \log_2[Cn/1.5Bn] \hspace{3cm} \text{Equation (2)}$$

where Cn is the measured concentration of the element n and Bn is the geochemical background value of element n in average crust (Togashi et al., 2000). The 1.5 factor is introduced to include possible variations of the background values due to lithogenic effects. Sediment quality based on the Igeo values (Müller 1969) is given as: <0=practically unpolluted, 0–1=unpolluted to moderately polluted, 1–2=moderately polluted, 2–3=moderately to strongly polluted, 3–4= strongly polluted, 4–5=strongly to extremely polluted, and >5=extremely polluted.

4. Results and Discussion

4.1 Sediment Characteristics

The sediment samples from the mangrove areas are essentially muddy, whereas those from the foreshore consist mainly of fine sand. The Gesashi mangrove sediments are red in hue, whereas dark sediments dominate in the Ohura mangrove. Among the foreshore sediments, the Kushi, Arume, Abu, and Henoko samples are characterized by red sands, and those at Futami by yellow sands. Loss on ignition values of the Gesashi mangrove, the Ohura mangrove, and the foreshore surface sediments average 7.54, 12.10, and 20.32 wt%,

respectively. Figure 2 shows the ranges, quartiles and median LOI values for the Gesashi and Ohura mangroves, and the foreshore sediments.
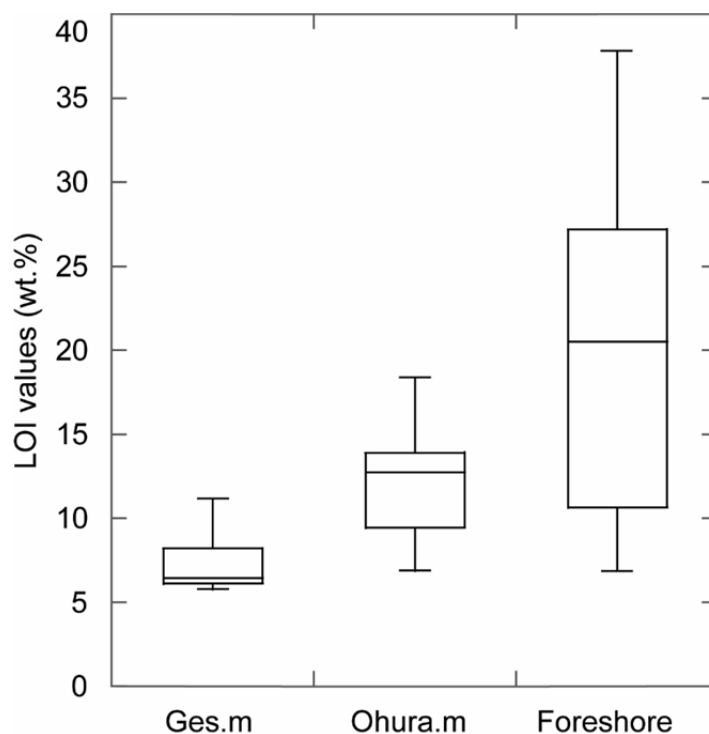

Figure 2. Loss on ignition (LOI) values for the Gesashi mangrove (Ges.m), Ohura mangrove (Ohura.m), and foreshore surface sediments

4.2 Concentrations of Elements

Elemental concentrations of the Gesashi and Ohura mangroves, the foreshore sediments, and the suspended solids are presented in Table 1. This table also includes the loss on ignition data, as along with the Japan upper crust (JUC) values from Togashi et al. (2000), and those of the upper continental crust (UCC) from Rudnick & Gao (2005).

4.2.1 Gesashi Mangrove

The average concentration of As in the Gesashi mangrove (15 mg/kg) is triple that of UCC and double the JUC value (Table 1). However, the average concentrations of Pb (12 mg/kg), Zn (39 mg/kg), Cu (16 mg/kg), Ni (10 mg/kg), and Cr (57 mg/kg) at Gesashi are slightly lower than in both UCC and JUC. On average the Gesashi mangrove sediments contain 0.65 w% TiO_2, 4.90 w% Fe_2O_3, 0.04 w% MnO, 2.57 w% CaO, and 0.09 w% P_2O_5. The average TS content is 2111 mg/kg. No TS data are available for UCC and JUC.

4.2.2 Ohura mangrove

The average concentration of As is considerably enriched with respect to UCC and JUC, and those of Pb and Zn are slightly enriched. The concentrations of Cu and Cr are almost identical to the UCC and JUC values, whereas Ni contents are lower than these two reference values. In the Ohura mangrove, TiO_2, Fe_2O_3, MnO, CaO, and P_2O_5 averaged 0.86, 5.75, 0.03, 1.45, and 0.12 w%, respectively. The average TS content is 2479 mg/kg.

4.2.3 Foreshore Sediments

Except the average concentration of CaO (25.43 wt%), which is almost seven times greater than that of JUC and UCC, and that of As (17 mg/kg), which is slightly above the JUC, but almost double that of UCC, the average concentrations of the other elements (7 mg/kg Pb, 11 mg/kg Zn, 6 mg/kg Cu, 9 mg/kg Ni, 14 mg/kg Cr, 0.12 wt% TiO_2, 0.80 wt% Fe_2O_3, 0.02 wt% MnO, and 0.05 wt% P_2O_5) are considerably lower than the two reference values (JUC and UCC). TS averages 2121 mg/kg.

Table 1. Elemental concentrations in the Gesashi and Ohura mangroves, foreshore surface sediments and suspended solids

Sampling no.	Trace elements (mg/kg)						Major elements (wt%)						LOI (%)
	As	Pb	Zn	Cu	Ni	Cr	TS	TiO_2	Fe_2O_3	MnO	CaO	P_2O_5	
Okinawa													
Gesashi mangrove sediments (n=5)													
Ges m-1	12	9	35	14	6	51	2481	0.55	3.77	0.02	7.55	0.08	11.12
Ges m-2	17	11	43	17	8	62	2526	0.68	5.35	0.05	3.48	0.11	8.20
Ges m-3	15	13	39	17	11	57	2452	0.70	5.35	0.05	0.70	0.09	6.45
Ges m-4	13	11	37	15	14	54	1407	0.63	4.56	0.03	0.51	0.07	6.10
Ges m-5	15	13	42	18	12	63	1688	0.68	5.47	0.07	0.60	0.09	5.82
Avg.	15	12	39	16	10	57	2111	0.65	4.90	0.04	2.57	0.09	7.54
Min.	12	9	35	14	6	51	1407	0.55	3.77	0.02	0.51	0.07	5.82
Max.	17	13	43	18	14	63	2526	0.70	5.47	0.07	7.55	0.11	11.12
Ohura mangrove sediments (n=7)													
Ohura 1	9	24	79	26	28	76	3516	0.82	4.95	0.02	0.67	0.12	13.80
Ohura 2	14	14	60	12	12	54	1397	0.61	4.65	0.03	6.35	0.09	9.36
Ohura 3	13	26	97	32	30	92	3361	0.97	6.66	0.03	0.72	0.16	18.34
Ohura 4	11	25	92	31	30	88	2590	0.92	6.73	0.04	0.66	0.14	14.01
Ohura 5	11	25	91	33	31	90	3061	0.90	5.84	0.04	0.63	0.14	12.73
Ohura 6	10	22	81	27	27	92	2051	0.91	6.06	0.04	0.58	0.12	9.54
Ohura 7	9	18	73	19	24	76	1375	0.86	5.35	0.03	0.51	0.08	6.89
Avg.	11	22	82	26	26	81	2479	0.86	5.75	0.03	1.45	0.12	12.10
Min.	9	14	60	12	12	54	1375	0.61	4.65	0.02	0.51	0.08	6.89
Max.	14	26	97	33	31	92	3516	0.97	6.73	0.04	6.35	0.16	18.34
Foreshore sediments (n=32)													
Ges m-6	6	6	6	5	nd	13	1962	0.09	0.63	0.01	27.41	0.04	22.22
Ges m-7	8	5	10	6	nd	17	1935	0.18	1.03	0.01	25.29	0.05	21.25
Ges m-8	4	4	0	5	nd	nd	3040	nd	nd	nd	51.29	0.05	37.72
Ges m-9	5	6	13	21	1	7	1672	0.05	0.47	nd	23.84	0.04	26.18
Ges 1	3	6	2	3	nd	1	3027	nd	0.07	nd	49.34	0.05	37.78
Ges 2	4	4	4	5	nd	nd	2726	nd	nd	nd	46.10	0.04	34.51
Taira 1	5	6	14	10	nd	7	2015	0.08	0.72	0.02	32.01	0.04	27.63
Taira 2	8	6	9	7	23	18	2052	0.04	0.51	0.01	28.45	0.05	24.64
Arume 1	12	10	32	7	5	38	1213	0.39	2.46	0.02	7.57	0.05	8.10
Arume 2	12	9	35	8	4	39	1014	0.36	2.52	0.02	9.54	0.05	9.13
Abu 1	4	6	6	5	nd	14	2363	0.09	0.29	nd	25.89	0.05	19.76
Abu 2	4	5	2	2	nd	4	3000	0.01	nd	nd	40.35	0.06	30.83
Abu 3	4	5	4	4	nd	10	2629	0.04	0.01	nd	37.28	0.05	28.44
Abu 3b	5	7	9	4	nd	26	1200	0.19	0.57	nd	14.73	0.04	11.04
Kayo 1a	5	7	9	3	nd	12	1682	0.14	0.52	nd	20.34	0.04	15.96
Kayo 2	5	6	6	1	nd	10	1820	0.11	0.46	nd	22.42	0.04	17.92
Kayo 3	5	7	7	4	nd	10	2004	0.11	0.38	nd	24.26	0.04	18.42
Henoko 1	19	11	21	5	nd	16	1457	0.14	1.04	0.01	10.51	0.05	9.23
Henoko 2	18	11	21	5	nd	10	1182	0.08	1.01	0.01	11.61	0.05	10.77
Henoko 3	19	13	24	6	nd	11	1168	0.09	1.15	0.02	11.07	0.05	10.54
Futami 1	17	19	61	19	13	59	2063	0.67	4.58	0.05	4.65	0.11	9.62
Futami 2a	7	6	6	4	nd	2	2639	0.02	0.11	nd	36.41	0.05	26.73
Futami 2b	7	6	2	4	nd	nd	2943	0.02	0.07	nd	38.25	0.05	28.39
Kushi J	7	5	1	4	nd	2	2785	0.01	nd	nd	36.43	0.04	24.90
Kushi 1	10	6	8	3	nd	7	2423	0.10	0.28	nd	20.57	0.04	17.06
Kushi 2	9	6	6	4	nd	10	2540	0.08	0.25	nd	18.82	0.04	15.53
Kushi 3	24	7	9	5	nd	11	1450	0.08	0.89	0.05	7.44	0.03	6.93
Kushi 4	21	5	5	3	nd	13	1542	0.07	0.60	0.02	10.15	0.03	8.24
Kushi 5	20	6	7	4	nd	11	1659	0.09	0.67	0.01	11.87	0.03	8.96
Kushi 2Sedake	9	6	10	12	nd	3	2979	0.04	0.13	nd	33.12	0.05	23.64
Kushi 3Sedake	8	6	5	3	nd	3	2936	0.05	0.11	0.01	33.89	0.05	24.89
Teima	4	5	1	4	nd	nd	2765	nd	nd	nd	42.87	0.05	33.14
Avg.	9	7	11	6	9	14	2121	0.12	0.80	0.02	25.43	0.05	20.32
Min.	3	4	0	1	1	1	1014	0.01	0.01	0.01	4.65	0.03	6.93
Max.	24	19	61	21	23	59	3040	0.67	4.58	0.05	51.29	0.11	37.78
Suspended solids (n=3)													
GW-1	3	10	24	22	19	26	3961	0.35	2.53	0.11	0.67	0.35	na
GW-2	5	8	34	24	26	25	1601	0.42	1.95	0.06	1.99	0.17	na
GW-3	2	9	17	18	19	2	1100	0.06	0.09	0.01	2.40	0.10	na
Avg.	3	9	25	22	21	18	2221	0.28	1.52	0.06	1.69	0.21	
Min.	2	8	17	18	19	2	1100	0.06	0.09	0.01	0.67	0.10	
Max.	5	10	34	24	26	26	3961	0.42	2.53	0.11	2.40	0.35	
JUC	7	17	74	25	38	84	na	0.62	5.39	0.11	3.39	0.12	
UCC	5	17	67	28	47	92	na	0.64	5.04	0.10	3.59	0.15	

Loss on ignition (LOI); Japan upper crust (JUC; Togashi et al.,2000); Upper continental crust (UCC; Rudnick and Gao, 2005); *nd* not detected ;*na* not analyzed

4.2.4 Suspended Solids

The elemental concentrations in the suspended solids are almost all significantly depleted relative to JUC and UCC values, except for P_2O_5 (0.22 wt %) which is slightly above these two references values (0.12 and 0.15 wt%, respectively; Table 1).

On average the suspended solids contain 3 mg/kg As, ranging from 2 to 5 mg/kg, and Pb averaged 9 mg/kg, ranging from 8 to 10mg/kg. Zinc and Cu abundances ranged from 17 to 37 mg/kg and 18 to 24 mg/kg (average 25 and 22 mg/kg respectively), whereas Ni and Cr contents ranged between 19 and 26 mg/kg and 2 and 26 mg/kg, with averages of 21 and 18 mg/kg, respectively. Averages of all these trace elements are lower than in JUC and UCC (Table 1). The concentrations of the major oxides in the suspended solids averaged 0.28 wt% TiO_2, 1.52 wt% Fe_2O_3, 0.06 wt% MnO, 1.69 wt% CaO, and 0.21 wt% P_2O_5. All except P_2O_5 are lower than in JUC and UCC.

Figure 3 presents a graphical statistical summary of As, Pb, Zn, Cu, Ni, and Cr concentrations in the Gesashi and Ohura mangroves, foreshore, and suspended solids from Okinawa.
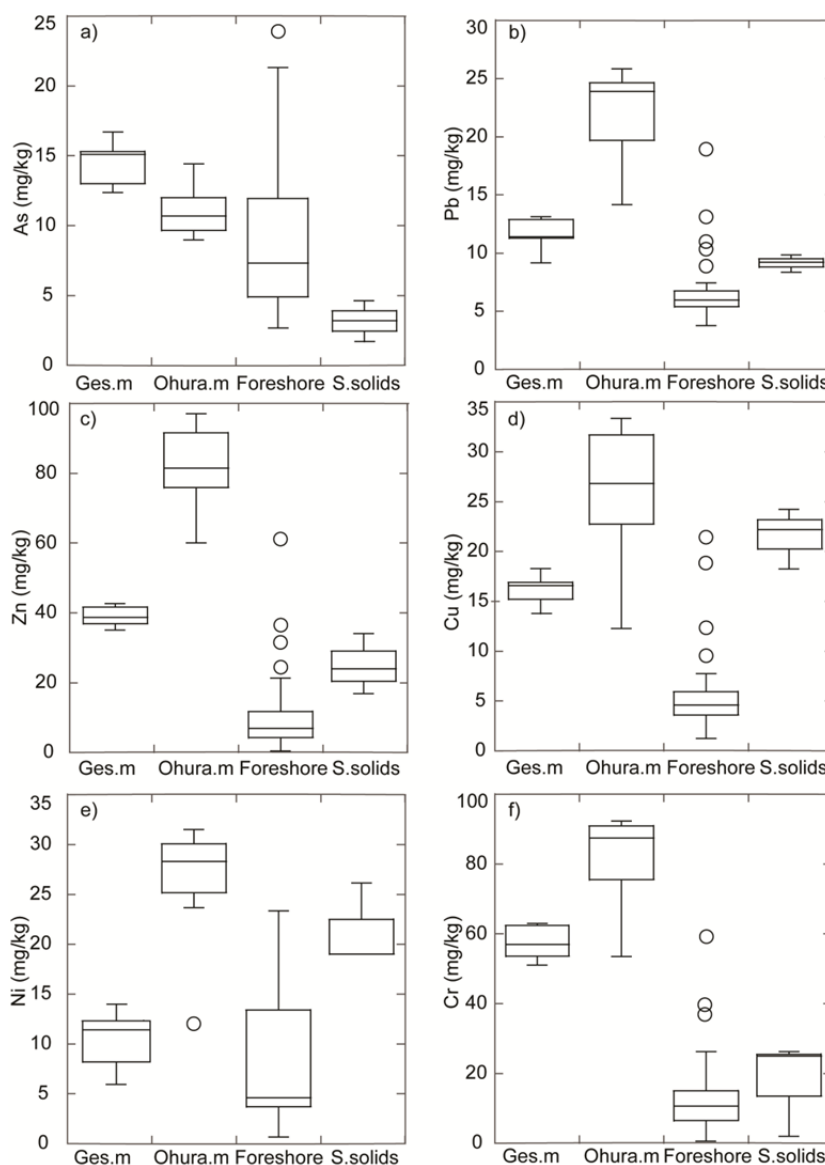

Figure 3.a-f Statistical summary of the concentratins of trace metals in the Gesashi mangrove (Ges.m), Ohura mangrove (Ohura.m), foreshore surface sediments,and suspended (S) solids from water samples. *Vertical lines* give the range (min. and max.), excluding outliers (circles); *boxes* show the first quartile (lower) and the third quartile (upper); *the horizontal lines* within the boxes indicate the median

4.3 Correlation matrices

Table 2 shows the correlations between the elements in the Gesashi and Ohura mangroves, foreshore sediments, and suspended solids. In addition, the correlations between the selected elements (As, Pb, Zn, Cu, Ni, and Cr) and TiO_2 (Figure 4), Fe_2O_3 (figure 5), and LOI (Figure 6) characterize the association between these trace elements, the two considered major elements, and the organic and carbonate contents.

Table 2. Correlations between the elements in the Gesashi and Ohura mangroves, foreshore surfacesediments, and suspended solids

	As	Pb	Zn	Cu	Ni	Cr	TS	TiO_2	Fe_2O_3	MnO	CaO	P_2O_5	LOI
Gesashi mangrove (n=5)													
As	1.00												
Pb	**0.60**	1.00											
Zn	**0.92**	0.59	1.00										
Cu	**0.80**	**0.84**	**0.90**	1.00									
Ni	0.05	**0.72**	0.17	0.45	1.00								
Cr	**0.89**	**0.63**	**0.99**	**0.94**	0.20	1.00							
TS	0.33	-0.27	0.04	-0.17	-0.81	0.00	1.00						
TiO_2	**0.85**	**0.91**	**0.79**	**0.88**	0.53	**0.79**	-0.05	1.00					
Fe_2O_3	**0.89**	**0.89**	**0.89**	**0.95**	0.46	**0.89**	-0.05	**0.98**	1.00				
MnO	**0.84**	**0.80**	**0.91**	**0.99**	0.31	**0.95**	-0.03	**0.86**	**0.94**	1.00			
CaO	-0.37	-0.89	-0.43	-0.68	-0.94	-0.46	**0.60**	-0.79	-0.73	-0.57	1.00		
P_2O_5	**0.87**	0.29	**0.78**	**0.61**	-0.40	**0.76**	**0.64**	0.54	**0.63**	**0.71**	0.08	1.00	
LOI	-0.37	-0.89	-0.47	-0.71	-0.94	-0.49	**0.64**	-0.78	-0.74	-0.60	**1.00**	0.06	1.00
Ohura mangrove (n=7)													
As	1.00												
Pb	-0.30	1.00											
Zn	-0.17	**0.94**	1.00										
Cu	-0.28	**0.97**	**0.97**	1.00									
Ni	-0.53	**0.95**	**0.92**	**0.96**	1.00								
Cr	-0.41	**0.84**	**0.90**	**0.91**	**0.90**	1.00							
TS	-0.15	**0.89**	**0.73**	**0.79**	**0.76**	0.54	1.00						
TiO_2	-0.47	**0.83**	**0.90**	**0.87**	**0.92**	**0.97**	0.53	1.00					
Fe_2O_3	-0.01	**0.71**	**0.88**	**0.79**	**0.70**	**0.85**	0.38	**0.85**	1.00				
MnO	0.25	-0.15	0.10	-0.04	-0.11	0.15	-0.49	0.16	0.54	1.00			
CaO	**0.78**	-0.76	-0.73	-0.77	-0.91	-0.85	-0.51	-0.91	-0.59	0.05	1.00		
P_2O_5	0.13	**0.89**	**0.93**	**0.90**	**0.76**	**0.77**	**0.80**	**0.71**	**0.79**	0.00	-0.47	1.00	
LOI	0.26	**0.79**	**0.76**	**0.70**	0.59	0.48	**0.87**	0.49	0.57	-0.15	-0.29	**0.89**	1.00
Foreshore sediments (n=32)													
As	1.00												
Pb	0.51	1.00											
Zn	0.46	**0.91**	1.00										
Cu	0.08	0.44	0.59	1.00									
Ni	0.10	0.10	-0.07	-0.29	1.00								
Cr	0.28	**0.73**	**0.87**	0.41	0.09	1.00							
TS	-0.58	-0.49	-0.52	-0.16	0.72	-0.50	1.00						
TiO_2	0.25	**0.77**	**0.90**	0.44	-0.07	**0.96**	-0.40	1.00					
Fe_2O_3	0.41	**0.84**	**0.96**	0.50	-0.03	**0.93**	-0.46	**0.95**	1.00				
MnO	0.47	0.50	0.54	**0.60**	-0.16	0.52	-0.18	0.54	**0.61**	1.00			
CaO	-0.76	-0.65	-0.65	-0.22	0.34	-0.64	**0.86**	-0.62	-0.64	-0.57	1.00		
P_2O_5	-0.02	**0.60**	**0.61**	0.43	0.39	0.57	0.25	**0.62**	**0.68**	0.42	0.05	1.00	
LOI	-0.78	-0.59	-0.56	-0.07	0.25	-0.57	**0.82**	-0.54	-0.54	-0.47	**0.98**	0.11	1.00
Suspended solids (n=3)													
As	1.00												
Pb	-0.55	1.00											
Zn	**0.99**	-0.65	1.00										
Cu	**0.99**	-0.41	**0.96**	1.00									
Ni	**0.86**	-0.90	**0.91**	**0.76**	1.00								
Cr	**0.86**	-0.05	**0.79**	**0.93**	0.47	1.00							
TS	0.18	**0.72**	0.06	0.34	-0.35	**0.66**	1.00						
TiO_2	**0.95**	-0.26	**0.90**	**0.99**	0.65	**0.98**	0.48	1.00					
Fe_2O_3	**0.74**	0.15	**0.66**	**0.84**	0.29	**0.98**	**0.79**	**0.92**	1.00				
MnO	0.52	0.43	0.41	**0.65**	0.00	**0.88**	**0.94**	**0.76**	**0.96**	1.00			
CaO	-0.25	-0.67	-0.13	-0.40	0.29	-0.71	-1.00	-0.54	-0.83	-0.96	1.00		
P_2O_5	0.29	**0.64**	0.17	0.44	-0.25	**0.74**	**0.99**	0.58	**0.86**	**0.97**	-1.00	1.00	

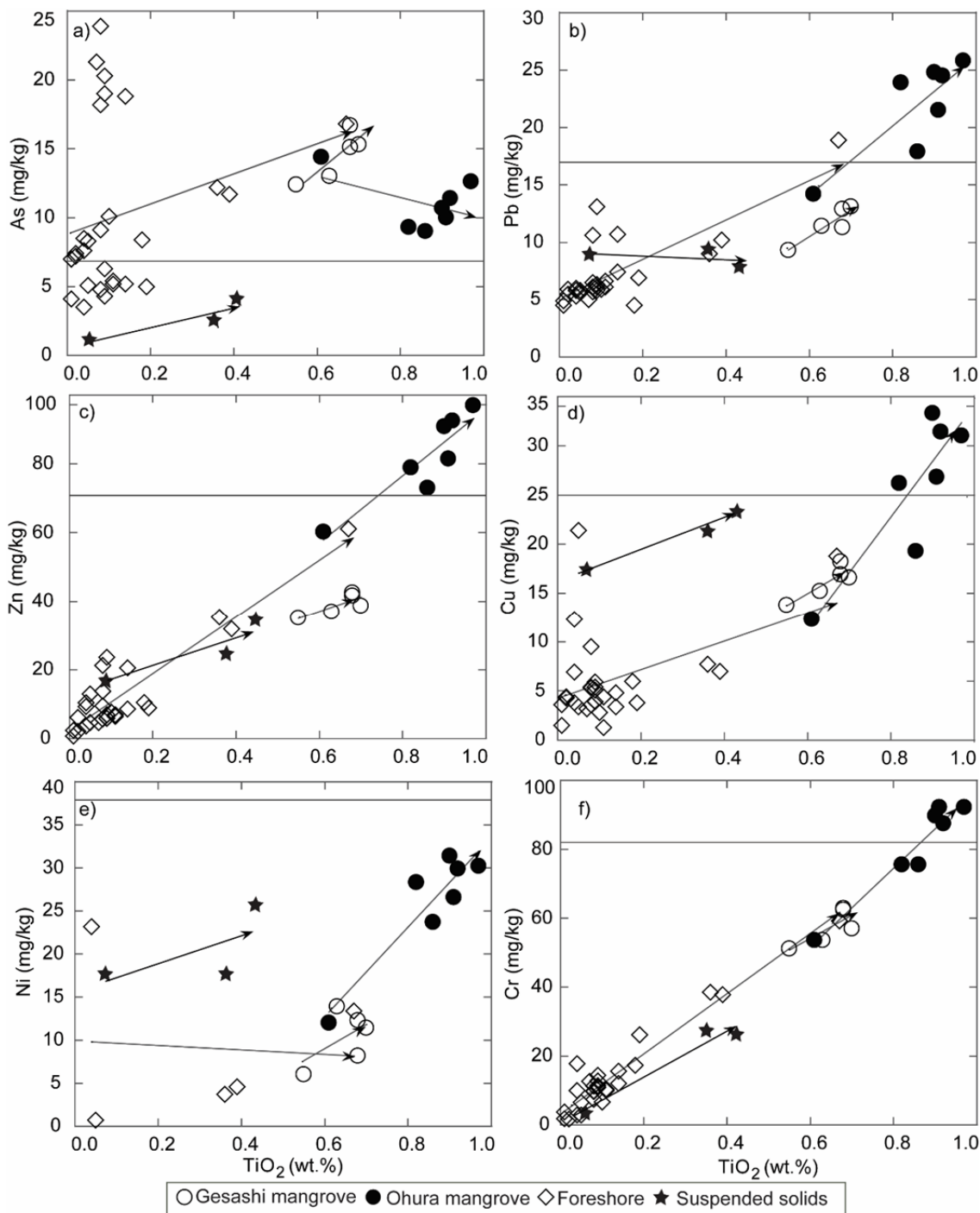Bold values highlight strong correlations (≥60)

Figure 4. a-f Correlations between Ti0$_2$ (wt.%) and As, Pb, Zn, Cu, Ni, and Cr in the Gesahsi and Ohura mangroves, foreshore surface sediments, and suspend solids from Okinawa Island. *Arrows* show the detrital trend lines. *Horizontal lines* indicates Japan upper crust average values from Togashi et a1. (2000)
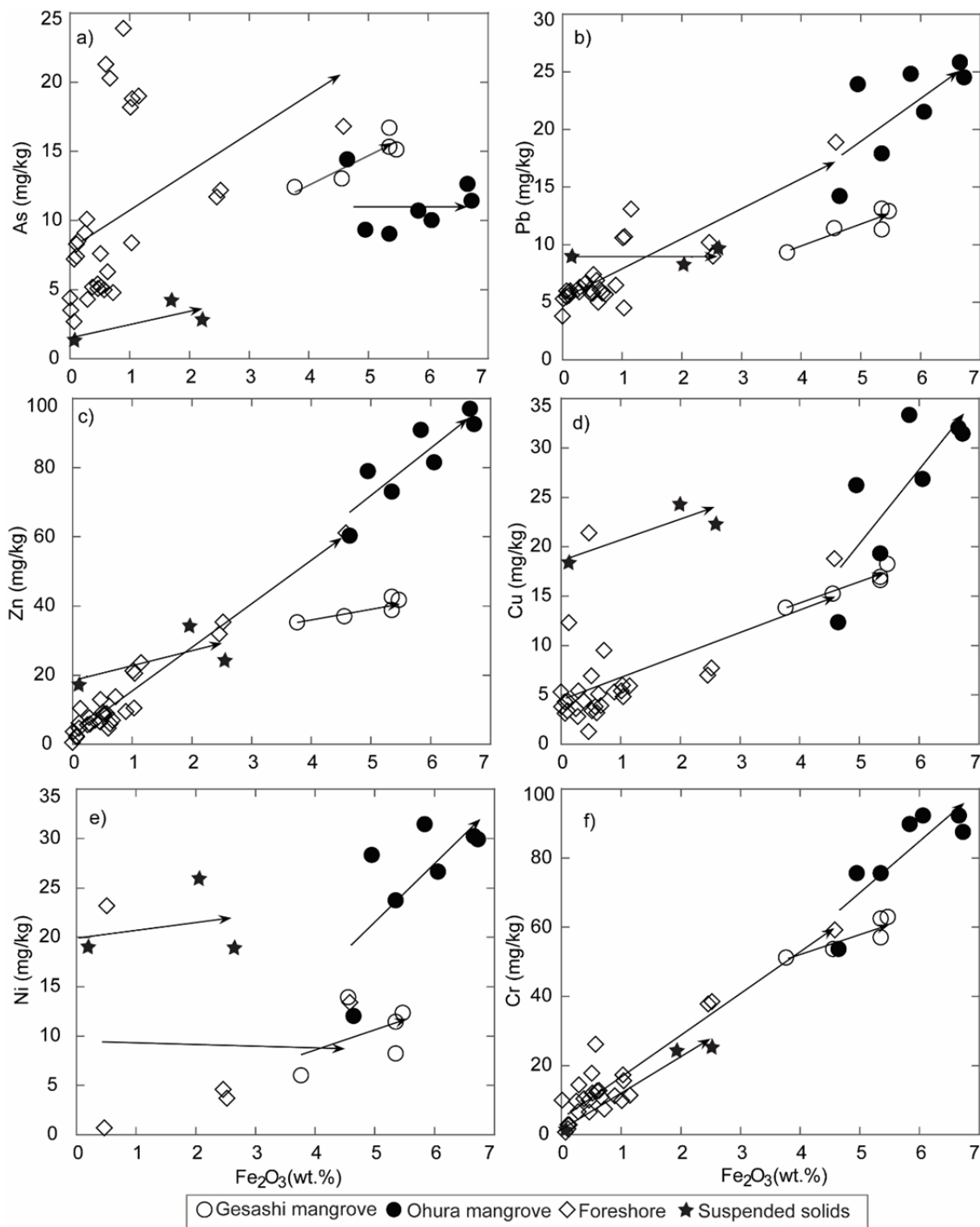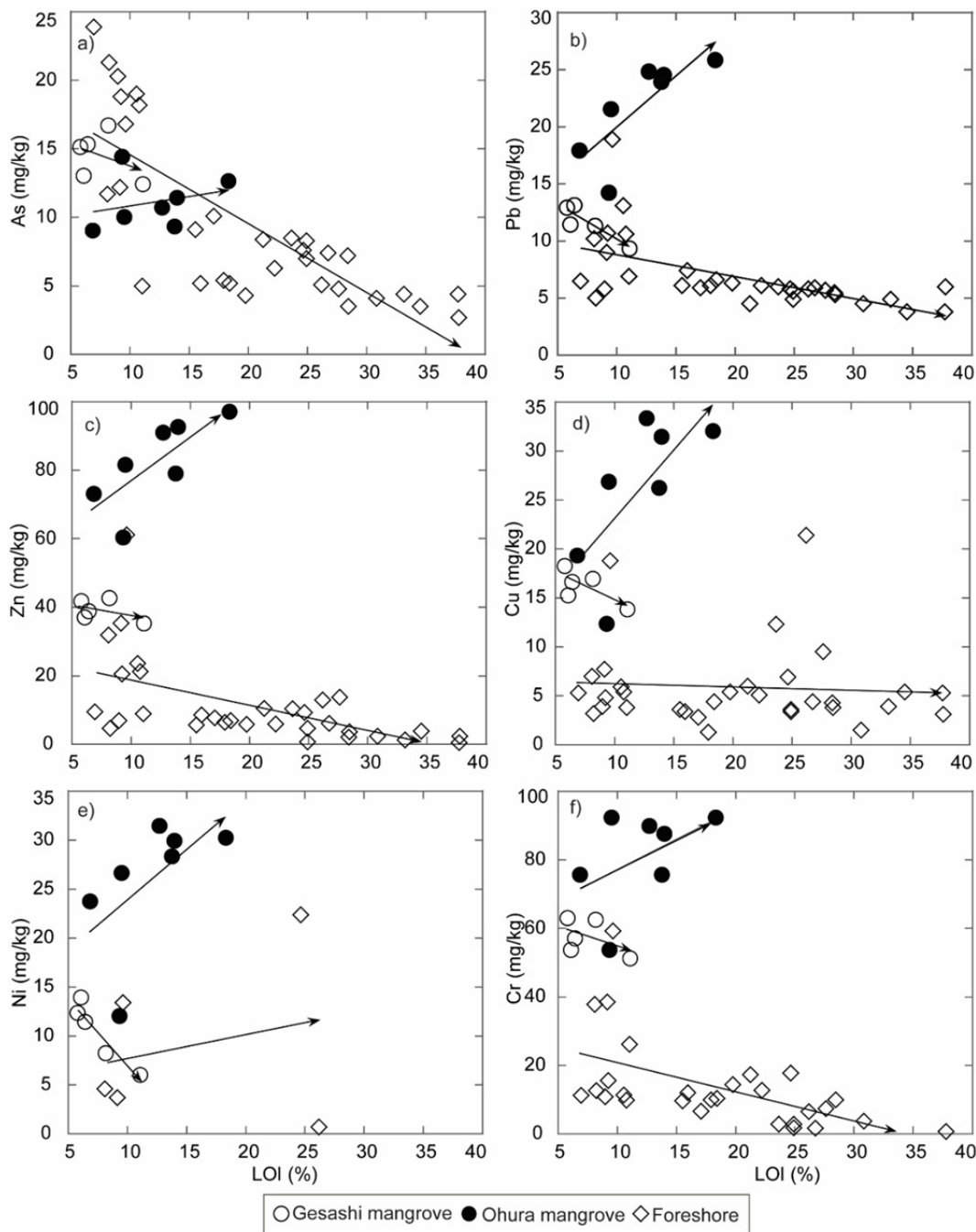
Figure 5. a-f Correlations between Fe_2O_3 (wt.%) and As, Pb, Zn, Cu, Ni, and Cr in the Gesashi and Ohura mangroves, foreshore surface sediments, and suspended solids from Okinawa Island

Figure 6. a-f Correlations between Loss on ignition (LOI) and As, Pb, Zn, Cu , Ni, and Cr in the Gesahsi and Ohura mangroves, and foreshore surface sediments from Okinawa Island

4.3.1 Gesashi Mangrove Sediments

TiO_2, Fe_2O_3, and MnO show strong positive correlation with As, Pb, Zn, and Cu, but these three major elements display weak and negative correlations with Ni and TS, respectively. The LOI values are negatively or weakly correlation with all elements except TS and CaO which are positively correlated with LOI.

4.3.2 Ohura Mangrove Sediments

Lead, Zn, Cu, Ni, and Cr are strongly associated with TiO_2 and Fe_2O_3, whereas As is negatively correlated with these two major oxides. In addition, the LOI values show strong positive association with Pb, Zn, and Cu, but weak or negative association with all other elements.
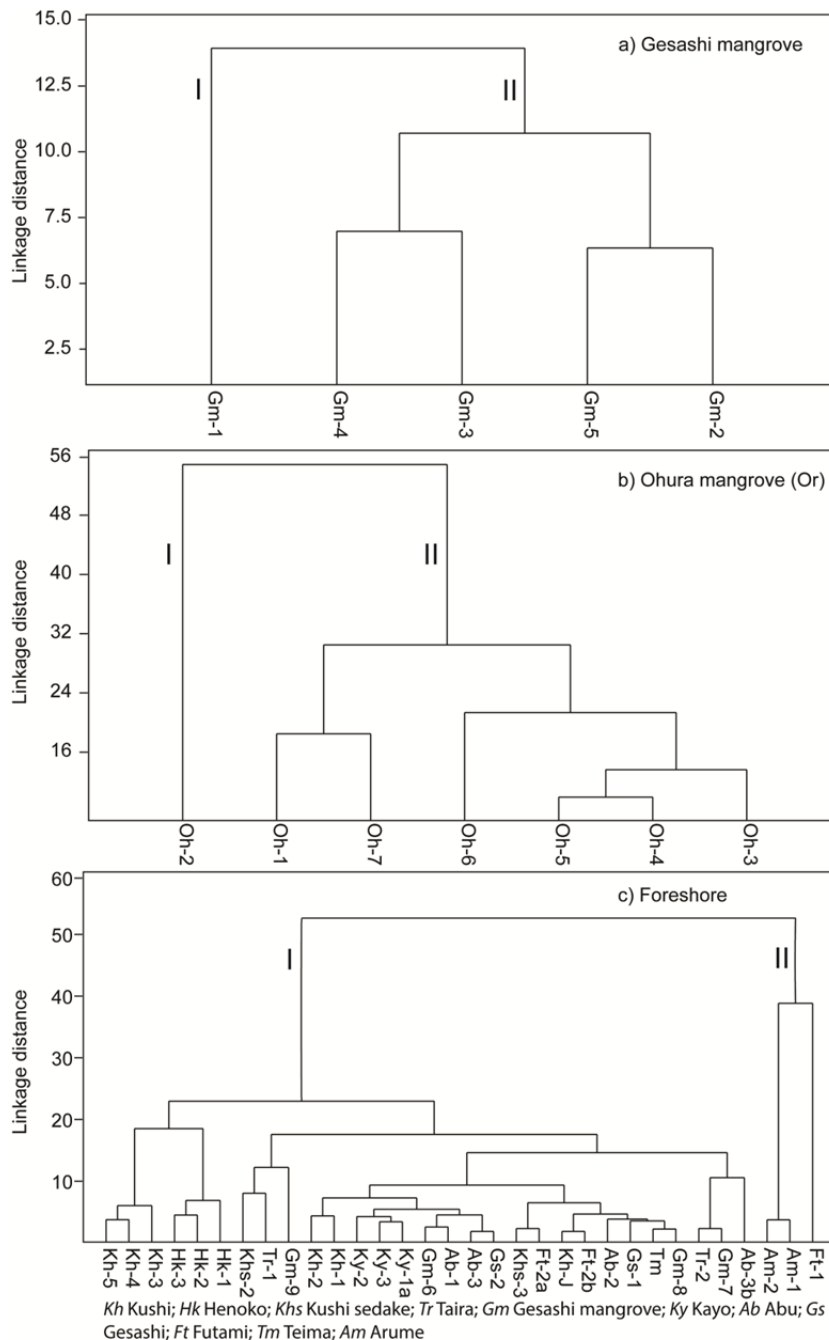
Figure 7. a-c Hierarchical cluster analysis of the Gesashi mangrove(a), Ohura mangrove(b), and foreshore(c) surface sediments based on the concentrations of As, Pb, Zn, Cu, Ni, and Cr

4.3.3 Foreshore Sediments

Fe_2O_3 is strongly associated with Pb, Zn, Cr, TiO_2, MnO, and P_2O_5, but weakly correlated with As and Cu, and negatively correlated with Ni and TS. TiO_2 shows strong correlation with Pb, Zn, and Cr, weak correlation with As and Cu, negative correlation with Ni and TS. MnO is strongly correlated with Cu. The LOI is negatively or weakly correlated with all elements, except TS and CaO, which show strong positive correlation with LOI.

4.3.4 Suspended Solids

TiO_2 and Fe_2O_3 are both strongly correlated with As, Zn, Cu, Cr, and MnO. In addition, TiO_2 shows strong association with Ni, as does Fe_2O_3 with TS. MnO is strongly associated with Cu, Cr, TS, and P_2O_5.

Overall, TiO$_2$ shows strong correlations with As, Pb, Zn, Cu, and Cr in the Gesashi mangrove, with Pb, Zn, Cu, Ni and Cr in the Ohura mangrove, with Pb, Zn, and Cr in the foreshore sediments (Figure 4), and finally with As, Zn, Cu, Ni, and Cr in the suspended solids (Table 2). Titanium is considered to be immobile in most geological processes, and hence is transferred quantitatively from source to sediment; TiO$_2$ content has often been used as a proxy to define sediment sources (Roser, 2000). In addition, linear correlations exist between TiO$_2$ and other lithogenic elements in soils and sediments. Consequently strong correlations with TiO$_2$ should only reflect natural detrital origin, whereas lack of correlation between TiO$_2$ and a given metallic element suggest additional natural or anthropogenic enrichment of that element (Dalai & Ishiga, 2013). The strong associations between TiO$_2$ and the metals in this study suggest that the detrital fraction has contributed significantly to the enrichments of the analyzed metals (Figure 4).

In all the study areas, Fe$_2$O$_3$ displays almost identical correlation trends relative to the trace metals as TiO$_2$ (Table 2 and Figure 5), also implying primary detrital control. However, iron is an excellent scavenger of trace metals, and this process may also have influenced the enrichment of these trace elements.

Finally, the LOI does not exhibit any strong correlation with As, Pb, Zn, Cu, Ni, and Cr in the Gesashi mangrove and foreshore area. However, in the Ohura mangrove strong associations are recorded between the LOI and Pb, Zn, and Cu (Table 2), suggesting that the organic matter may have contributed to the enrichment of these trace metals.

4.4 Cluster Analysis

Based on the As, Pb, Zn, Cu, Ni, and Cr contents, the sampling sites in each study area fall into two clusters. In the Gesashi mangrove, cluster(I) is comprised of sampling site Gm-1 and cluster (II) is composed of the sampling sites Gm-4 to Gm-2, from left to right (Figure 7a). Cluster (I) in the Ohura mangrove consists of the sampling site Oh-2, whereas cluster (II) contains the sampling sites Oh-1 to Oh-3, from left to right (Figure7.b). The sampling set in the foreshore splits into Cluster (I), which includes sampling sites from Kh-5 to Ab-3b, and cluster (II) consists of the sampling sites from Am-2to Ft-1, from left to right (Figure7.c).

4.5 Contamination Factor (CF) and Geoaccumulation Index (Igeo)

The CF and Igeo values for the Gesashi and Ohura mangroves, the foreshore sediments, and the suspended solids are shown on Table 3. These values were obtained using equation (1) for the CF and equation (2) for Igeo.

4.5.1 Contamination Factor (CF)

Almost all the sampling sites from the Ohura mangrove show moderate enrichment with respect to As, Pb, Zn, and Cu, with CF values of 1-3 (Table 3). In addition, in this same area, more than half of the sampling sites are moderately enriched with respect to Cr, whereas in the Gesashi mangrove, only As displays moderate enrichment. In the foreshore sediments, almost all the sampling sites are moderately enriched in As. Furthermore, in the foreshore sediments, the Futami1 sampling site show moderate enrichment in Pb, and the Kushi3 and Kushi4 sites are considerably contaminated with As (CF:3-6).

On average, As, Pb, Zn, and Cu in the Ohura mangrove show moderate contamination (CF:1-3), as does As in the Gesashi mangrove and the foreshore sediments. Finally, in the suspended solids, all the selected trace metals show low contamination (CF<1).

4.5.2 Geoaccumulation Index (*Igeo*)

All the sampling sites in the Gesashi mangrove and more than half of the sampling sites in the Ohura mangrove are considered as unpolluted to moderately polluted (*Igeo*:0-1) (Table3) with respect to As, whereas the other trace elements (Pb, Zn, Cu, Ni, and Cr) show no sign of pollution, as do As, Pb, Zn, Cu, Ni, and Cr in the suspended solids(*Igeo*:< 0).

In all the sampling sites of the foreshore, no sign of pollution was recorded for Pb, Zn, Cu, Ni, and Cr (*Igeo*:< 0). Furthermore, most of the sampling sites show no contamination with respect to As. However, two foreshore sampling sites (Kushi3 and Kushi4) are rated as moderately polluted (*Igeo*:1-2) with respect to As, and seven sampling sites (Arume1, Arume2, Henoko1, Henoko2, Henoko3, Futami1, and Kushi5) are classified as unpolluted to moderately polluted (As *Igeo*:0-1).

Table 3. Contamination factor (CF) and Geoaccumulation index (*Igeo*) for the Gesashi and mangroves, foreshore surface sediments, and suspended solids

Sampling sites	Contamination factor (CF)						Geoaccumulation index (Igeo)					
	As	Pb	Zn	Cu	Ni	Cr	As	Pb	Zn	Cu	Ni	Cr
Gesashi mangrove (n=5)												
Ges m-1	1.82	0.55	0.47	0.55	0.16	0.61	0.28	-1.46	-1.66	-1.44	-3.25	-1.30
Ges m-2	2.46	0.66	0.58	0.68	0.22	0.74	0.71	-1.17	-1.38	-1.15	-2.80	-1.01
Ges m-3	2.25	0.77	0.52	0.66	0.30	0.68	0.58	-0.96	-1.52	-1.18	-2.32	-1.14
Ges m-4	1.91	0.67	0.50	0.61	0.37	0.64	0.35	-1.16	-1.59	-1.30	-2.04	-1.23
Ges m-5	2.22	0.76	0.56	0.73	0.32	0.75	0.57	-0.98	-1.41	-1.04	-2.21	-1.00
Ohura mangrove (n=7)												
Oura 1	1.37	1.41	1.07	1.05	0.74	0.90	-0.13	-0.09	-0.49	-0.52	-1.01	-0.74
Oura 2	2.12	0.84	0.81	0.49	0.32	0.64	0.50	-0.84	-0.88	-1.61	-2.25	-1.23
Oura 3	1.85	1.52	1.31	1.28	0.79	1.10	0.30	0.02	-0.19	-0.23	-0.92	-0.45
Oura 4	1.68	1.44	1.25	1.26	0.79	1.04	0.16	-0.06	-0.26	-0.26	-0.93	-0.53
Oura 5	1.57	1.46	1.23	1.33	0.83	1.07	0.07	-0.04	-0.29	-0.17	-0.86	-0.49
Oura 6	1.47	1.26	1.10	1.07	0.70	1.10	-0.03	-0.25	-0.45	-0.48	-1.10	-0.45
Oura 7	1.32	1.05	0.99	0.77	0.62	0.90	-0.18	-0.51	-0.61	-0.96	-1.27	-0.74
Foreshore (n=32)												
Ges m-6	0.93	0.36	0.08	0.20	0.00	0.15	-0.70	-2.06	-4.23	-2.88		-3.30
Ges m-7	1.24	0.26	0.14	0.24	0.00	0.21	-0.28	-2.50	-3.41	-2.64		-2.86
Ges m-8	0.65	0.22	0.01	0.21	0.00	0.00	-1.21	-2.75	-7.91	-2.82		
Ges m-9	0.75	0.34	0.18	0.86	0.02	0.08	-1.00	-2.14	-3.10	-0.81	-6.35	-4.28
Ges 1	0.40	0.35	0.03	0.12	0.00	0.01	-1.92	-2.09	-5.52	-3.60		-7.49
Ges 2	0.51	0.22	0.05	0.22	0.00	0.00	-1.54	-2.75	-4.84	-2.80		
Taira 1	0.71	0.34	0.19	0.38	0.00	0.09	-1.09	-2.16	-3.01	-1.98		-4.09
Taira 2	1.12	0.34	0.13	0.28	0.61	0.21	-0.42	-2.14	-3.57	-2.44	-1.30	-2.82
Arume 1	1.72	0.60	0.43	0.28	0.12	0.45	0.20	-1.32	-1.80	-2.42	-3.63	-1.74
Arume 2	1.79	0.53	0.48	0.31	0.10	0.46	0.26	-1.50	-1.65	-2.28	-3.95	-1.71
Abu 1	0.63	0.37	0.08	0.22	0.00	0.17	-1.25	-2.02	-4.24	-2.80		-3.13
Abu 2	0.60	0.26	0.03	0.06	0.00	0.05	-1.31	-2.50	-5.54	-4.64		-5.05
Abu 3	0.51	0.31	0.05	0.15	0.00	0.12	-1.54	-2.27	-4.91	-3.30		-3.66
Abu 3b	0.74	0.41	0.12	0.15	0.00	0.31	-1.03	-1.89	-3.64	-3.30		-2.27
Kayo 1a	0.76	0.44	0.12	0.14	0.00	0.14	-0.97	-1.78	-3.70	-3.46		-3.38
Kayo 2	0.79	0.36	0.09	0.05	0.00	0.12	-0.92	-2.06	-4.10	-4.85		-3.66
Kayo 3	0.76	0.39	0.09	0.18	0.00	0.12	-0.97	-1.95	-4.00	-3.09		-3.60
Henoko 1	2.76	0.63	0.28	0.19	0.00	0.19	0.88	-1.25	-2.43	-2.97		-3.01
Henoko 2	2.68	0.62	0.29	0.22	0.00	0.12	0.84	-1.27	-2.39	-2.80		-3.67
Henoko 3	2.79	0.77	0.32	0.24	0.00	0.14	0.90	-0.96	-2.23	-2.67		-3.47
Futami 1	2.47	1.11	0.83	0.75	0.00	0.70	0.72	-0.43	-0.86	-1.00	-2.09	-1.09
Futami 2a	1.09	0.35	0.08	0.18	0.00	0.02	-0.46	-2.11	-4.18	-3.09		-6.21
Futami 2b	1.06	0.32	0.03	0.17	0.00	0.00	-0.50	-2.21	-5.74	-3.12		
Kushi J	1.03	0.29	0.01	0.14	0.00	0.02	-0.54	-2.38	-7.35	-3.38		-6.13
Kushi 1	1.49	0.35	0.10	0.11	0.00	0.08	-0.01	-2.11	-3.84	-3.74		-4.25
Kushi 2	1.34	0.36	0.08	0.14	0.00	0.12	-0.16	-2.06	-4.30	-3.38		-3.70
Kushi 3	3.51	0.38	0.13	0.21	0.00	0.13	1.23	-1.97	-3.55	-2.82		-3.48
Kushi 4	3.13	0.29	0.06	0.13	0.00	0.15	1.06	-2.35	-4.58	-3.55		-3.32
Kushi 5	2.99	0.34	0.09	0.16	0.00	0.13	0.99	-2.14	-3.99	-3.27		-3.53
Kushi 2Sedake	1.25	0.35	0.14	0.49	0.00	0.03	-0.26	-2.09	-3.42	-1.61		-5.44
Kushi 3Sedake	1.22	0.33	0.06	0.14	0.00	0.03	-0.30	-2.19	-4.59	-3.46		-5.44
Teima	0.65	0.29	0.02	0.16	0.00	0.00	-1.21	-2.38	-6.43	-3.27		
Suspended solids (n=3)												
GW-1	0.47	0.58	0.32	0.89	0.50	0.31	-1.67	-1.38	-2.21	-0.76	-1.58	-2.28
GW-2	0.68	0.49	0.46	0.97	0.68	0.30	-1.15	-1.60	-1.71	-0.63	-1.13	-2.33
GW-3	0.25	0.54	0.23	0.73	0.50	0.03	-2.58	-1.47	-2.71	-1.04	-1.58	-5.91

CF≤1 low contamination; CF1-3 moderate contamination; CF3-6 considerable contamination; CF≥6 extreme contamination (Håkanson 1980). Igeo < 0 practically unpolluted; Igeo 0-1 unpolluted to moderately polluted; Igeo 1-2 moderately polluted; Igeo 2-3 moderately to strongly polluted; Igeo 3-4 strongly polluted; Igeo 4-5 strongly to extremely polluted, and Igeo>5 extremely polluted (Müller 1969)

4.6 Comparison of Metal Concentrations with Sediment Quality Guidelines

To evaluate potential toxic effects of the metal concentrations on aquatic biota health, the average concentrations of As, Pb, Zn, Cu, Ni, and Cr in the study areas were compared with sediment benchmarks established by the New York State Department of Environmental Conservation (NYSDEC, 1999) and the Canadian Council of Ministers of the Environment (CCME, 1998) (Table 4).

Table 4. Sediment quality criteria and average metal concentrations (mg/kg) in the Gesashi and Ohura mangroves, foreshore surface sediments, and suspended solids

Metals	LEL[1]	SEL[2]	ISQG[3]	PEL[4]	Gesashi M.	Ohura M.	Foreshore	S. Solids
As	6	33	7	42	15	11	9	3
Pb	31	110	30	112	12	22	7	9
Zn	120	270	124	271	39	82	11	25
Cu	16	110	19	108	16	26	6	22
Ni	16	50	na	na	10	26	9	21
Cr	26	110	52	160	57	81	14	18

M Mangrove, *S* Suspended, *na* not analyzed

[1] Lowest effect level (LEL; NYSDEC 1999)

[2] Severe effect level (SEL; NYSDEC 1999)

[3] Threshold effect level *(TEL)* =Interim Sediment Quality Guideline (ISQG; SAIC 2002)

[4] Probable effect limit (PEL; SAIC 2002)

The NYSDEC (1999) proposed the lowest effect level (LEL) and the severe effect level (SEL), and considered that if the SEL criteria was exceeded, the metal may severely impact biota health, whereas if only the LEL criterion was exceeded, the metal may moderately impact biota health. The CCME (1998) guidelines identified two numerical levels: the lower level is termed the interim sediment quality guideline (ISQG) or threshold effect level (TEL) value, and the upper level is called the probable effect level (PEL). Sediment chemical concentrations below ISQG values are not expected to be associated with any adverse biological effects, while concentrations above the PEL are expected to be frequently associated with adverse biological effects.

The average concentration of As in the suspended solids is lower than the LEL and ISQG guidelines, suggesting no adverse biological effects. However, the average concentration of As in the Gesashi and Ohura mangrove sediments, and the foreshore exceeds both the LEL and ISQG, but fall below the SEL and PEL, suggesting that these metals may moderately impact biota health.

Lead and Zn at all sampling areas are all below LEL and ISQG, suggesting no adverse biological effects. This is also the case for Ni in the Gesashi mangrove and foreshore sediments, and Cr in the foreshore and suspended solids. In addition, the average concentration of Cu in the foreshore sediments is lower than the LEL and ISQG, as is Ni in the Ohura mangrove and foreshore, reflecting no impact on the living organisms. In contrast, Cu concentration in the Gesashi mangrove is equal to the LEL, whereas in the Ohura mangrove and suspended solids, Cu exceeds both the LEL and ISQS, but is lower than SEL and PEL, implying moderate impact on biota health. This is also the case for Ni in the Ohura mangrove and suspended solids, and Cr in the Gesashi and Ohura mangroves,

5. Conclusions

In this study, the highest average concentrations of Pb (22 mg/kg), Ni (26 mg/kg), and Zn (82 mg/kg), and Cr (81mg/kg) were observed in the Ohura mangrove sediments, that of As (17 mg/kg) in the Gesashi mangrove, and that of Cu (22 mg/kg) in the suspended solids.

In the Gesashi and Ohura mangroves, all trace elements except Ni and As, show strong positive correlation with TiO_2. In addition, Pb, Zn, and Cr in the foreshore sediments, and all elements except Pb in the suspended solids, are also strongly associated with TiO_2, suggesting that these elements are mainly detrital in origin, and hence are primarily derived from natural sources. In contrast, TiO_2 displays negative correlation with Ni in the Gesashi mangrove, with As in the Ohura mangrove, weak or negative correlation with As, Cu, and Ni in the foreshore sediments, and negative correlation with Pb in the suspended solids, implying that these elements may have been partially derived from anthropogenic sources.

The contamination factor indicates that As, Pb, Zn, and Cu in the Ohura mangrove show moderate enrichment at almost all the sampling sites. Similarly, more than half of the Ohura sampling sites display moderate enrichment with respect to Cr. The contamination factor for arsenic at almost all Gesashi mangrove and foreshore sampling sites indicate moderate enrichment. In the suspended solids, relative to all the selected trace metals, the contamination factor and the geoaccumulation index indicate low contamination and no sign of pollution, respectively.

Among all the study areas, the geoaccumulation index shows that only the mangrove sediments display significant values for arsenic, which is rated as unpolluted to moderately polluted at all the sampling sites in the Gesashi mangrove and at more than half of the sampling sites in the Ohura mangrove. The sediment quality guidelines show that As concentrations in the Gesashi and Ohura mangroves and foreshore sediments may moderately impact biota health, because levels exceed the LEL and ISQG, but fall below the SEL and PEL. This is also the case for Cu in the Ohura mangrove and in the suspended solids, for Ni in the Ohura mangrove and suspended solids, and also for Cr in the Gesashi and Ohura mangroves.

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