Environment and Pollution; Vol. 6, No. 1; 2017
ISSN 1927-0909 E-ISSN 1927-0917
Published by Canadian Center of Science and Education

Geochemical Assessment of Trace Metal Distribution and Contamination in the Surface Sediments of the Coast of Okinawa Island, Japan

Ibrahima M'Bemba Diallo[1] & Hiroaki Ishiga[1]

[1] Department of Geoscience, Shimane University, Matsue, Japan

Correspondence: Ibrahima M'Bemba Diallo, Department of Geoscience, Shimane University, 1060 Nishikawatsu, Matsue, Shimane, 690-8504, Japan. Tel: 81-80-3888-6598. E-mail: ibrahimacentral@yahoo.fr

Received: June 22, 2016 Accepted: July 28, 2016 Online Published: November 17, 2016

doi:10.5539/ep.v6n1p1 URL: http://dx.doi.org/10.5539/ep.v6n1p1

Abstract

In this study, sediment samples were collected from the Awase tidal, the Minamigusuku, and Nakagusuku areas located in the Okinawa Island. The collected samples were analyzed using X-ray fluorescence to determine their geochemical compositions and distribution, and to assess the sediment quality in the study areas. Contamination factor (CF), geoaccumulation index (Igeo), and the United State Environmental Protection Agency Sediment Quality Guideline (US EPA SQG) were used to assess the sediment quality. The results show that the highest average concentrations of As (11mg/kg), Pb (8 mg/kg), Zn (19 mg/kg), Cu (6 mg/kg), and Cr (14mg/kg) occurred in the Minamigusuku area. The CF and Igeo values, and the US EPA SQG indicate that among the selected trace metals only As displays significant values in the study areas. The CF values of As in the Minamigusuku area show moderate enrichment, and in this same area, the Igeo values of As present significant values, ranging from moderate to considerable contaminations, implying a possible effect on the biota in this location. Compared to the US EPA SQG, the Awase tidal flat and Nakagusuku areas are moderately polluted, whereas Minamigusuku is heavily polluted, suggesting that As may possible impact the biota in these areas. Consequently, for a better sustainable development of the coast of Okinawa Island , a regular monitoring and assessment of study areas, particularly the Minamigusuku area, is necessary to determine over time the concentration of As.

Keywords: Geochemistry, trace metals, Awase tidal flat, Minamigusuku, Nakagusuku, sediment quality, Okinawa Island, south west Japan

1. Introduction

Trace metals are among the most common environmental pollutants and their occurrences in

water and biota indicate the presence of natural or anthropogenic sources (Singh et al., 2005). Higher concentrations of trace metals are released into the aquatic environment as a result of leaching from bedrocks, atmospheric deposition, water drainage, runoff from riverbanks, and discharge of urban and industrial wastewaters (Soares et al. 1999; Yang & Rose, 2005).

Elevated concentrations of trace metals in aquatic environments may induce severe impacts, with likely loss of aquatic habitat, decrease in fishery and aquatic plant resources, fish migration, and overall human health concern (Young, 2007). Furthermore, in aquatic environments, many trace metals are transported predominantly in association with particulate matter, which makes coastal sediments the most important repository for metal pollutants that enter the seas (Ridgway & Shimmield, 2002).

Awase tidal flat, Minamanigusuku, and Nakagusuku areas are situated in the Island of Okinawa, where sugar cane, pineapple, papaya, and other tropical fruit are produced. In addition, the Okinawa Island is characterized by important sedimentary environments such as, bays and tidal flats, which may trap trace metals derived from natural and anthropogenic sources, and thereby endangering the biota which populates the coast of Okinawa Island. Therefore, assessing the sediment quality in these various sedimentary environments in the Island of Okinawa is of great importance for the sustainable management of the Island.

Over the past years, although some geochemical studies were conducted in the Island of Okinawa (Ramos et

al.,2004; Naumih & Tamotsu, 2006; Vuai &Tokuyama, 2011). However, to date, research on the sediment quality along this Island has been lacking, or very limited, particularly in the Awase tidal flat, Minamigusuku, and Nakagusuku areas. Consequently, the main objectives of the present study were to determine the geochemical compositions of the Awase tidal flat, the Minamigusuku, and the Nakagusuku surface sediments, and to assess the sediment quality in these three areas using the contamination factor, the geoaccumulation index, and the United States Environmental Protection Agency (US EPA) sediment quality guideline.

2. Study Area

The three study areas, Awase tidal flat, Minamigusuku, and Nakagusuku are located in the Okinawa Island, south west of Japan (Figure1). The population in the Island exceeds one million people (1 130 682), with a density varying widely from 7500 persons km^{-2} in the south to 34 persons km^{-2} in the north (West & van Woesik, 2001). The island has a subtropical climate where the annual average atmospheric temperature and precipitation reach 22.7°C and 2036.9mm, respectively (National Astronomical Observatory, 2004). In Okinawa, 45% of the annual rainfall is concentrated between the rainy season of May-June and the typhoon season of July-October.

Located in the south east of Okinawa Island, Awase tidal flat, which extends about 200 ha, is one of the largest tidal flat areas in Okinawa, with an intertidal zone of approximately 265ha. Awase tidal flat is also home to diverse benthic species and the sediment in the area is of muddy and sandy components. Minamigusuku and Nakagusuku areas are famous for their agricultural activities such as rice cropping.

The geology of Okinawa Island is characterized by rock formations made up of Paleozoic sedimentary rocks with subordinate igneous dikes, the Shimajiri formation of the Miocene-Pliocene age, the Ryukyu limestone of the upper Pliocene or lower Pleistocene age, coral reefs of recent age (Konishi et al., 1972)

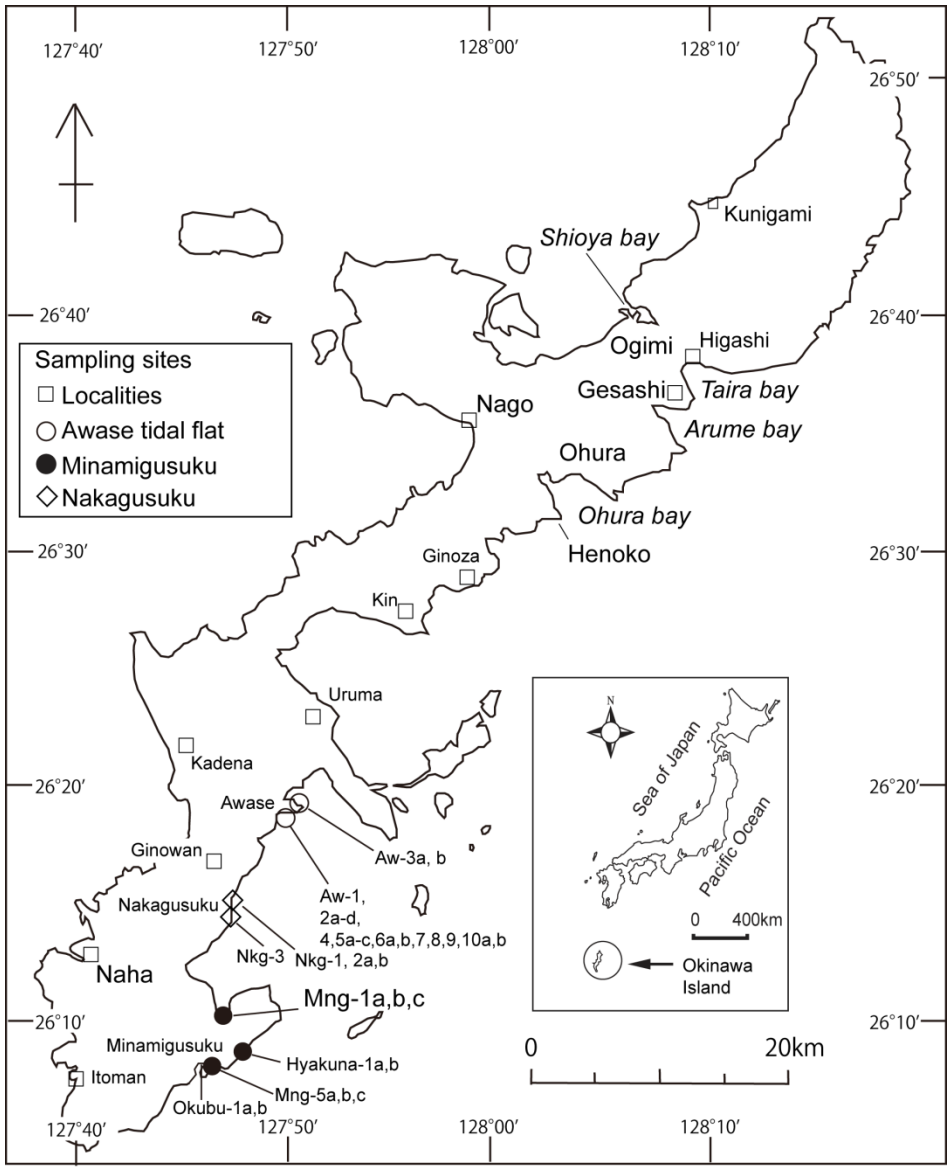

Figure1. Locations of sampling sites of the Awase (Aw) tidal, Minamigusuku (Mng), and Nakagusuku (Nkg) surface sediments from the Okinawa Island. Inset, location in Japan.

3. Materials and Methods

3.1 Sampling

In February 2016, seventeen, ten, and six sediment samples were collected from the Awase tidal flat, Minamigusuku, and Nakagusuku areas, respectively. About 200g of the uppermost 2 cm of each sampling site were packed in plastic bags and stored in a cooler box at 4ºC for transport to the laboratory.

Approximately 50 g of each sample were oven-dried at 110ºC for 24 h. The dried samples were then ground for 20 min in an automatic agate mortar and pestle grinder.

3.2 Analytical Procedures

Selected major and trace element (TiO_2, Fe_2O_3, MnO, CaO, P_2O_5, As, Pb, Zn, Cu, Ni, Cr) and total sulfur (TS) concentrations were determined by XRF at Shimane University, using a RIX-2000 spectrometer (Rigaku Denki Co. Ltd) equipped with a Rh-anode X-ray tube. All analyses were made on pressed powder briquettes (a force of 200 kN for 60 s), following the method of Ogasawara (1987). Average errors for all elements are less than ±10% relative. Analytical results for USGS standard SCo-1 (Cody Shale) were acceptable compared to the proposed

values of Potts et al. (1992).

3.3 Sediment Quality

Contamination factor (CF), geoaccumulation index (Igeo), and United States Environmental Protection Agency Sediment Quality Guidelines (US EPA SQG) were used to assess the sediment quality. Japan upper crust from Togashi et al.(2000) was used as reference values to compute the CF and Igeo. For the US EPA SQG, values provided by Perin et al. (1997) were considered as the reference values

A. Contamination factor (CF)

The CF for each sampling site was obtained according to Håkanson (1980):

$$\mathrm{CF} = \mathrm{C}x/\mathrm{C}ref \quad (1)$$

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)

Geoaccumulation index values were determined based on Müller (1969) following this equation:

$$\mathrm{Igeo} = \log_2[\mathrm{C}n/1.5\mathrm{B}n] \quad (2)$$

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. Resuts and Discussion

4.1 Sediment Characteristics

The sediments in the study areas are composite, including mud and sand. Awase tidal flat sediments contain more sand than mud, whereas Minamigusuku and Nakagusuku show mudy sediments, essentially composed of silt and clay. Both the sandy and the muddy sediments in these areas are influenced by the red soil erosion, which is frequent in the Okinawa Island, and therefore imparts red color to the coastal sediments (Takemitsu et al., 2005)

Table 1 Elemental concentrations, contamination factor (CF), and Geoaccumulation index (Igeo) in the Awase tidal flat, Minamigusuku, and Nakagusuku surface sediments

sample	Trace elements (mg/kg)							Major elements (wt.%)					Contamination factor (CF)					Geoaccumulation index (Igeo)				
	As	Pb	Zn	Cu	Ni	Cr	TS	TiO_2	Fe_2O_3	MnO	CaO	P_2O_5	As	Pb	Zn	Cu	Cr	As	Pb	Zn	Cu	Cr
Awase tidal flat (Aw)																						
Awase 1	5	6	15	7	nd	nd	4710	nd	nd	nd	53.12	0.11	0.69	0.35	0.20	0.26		-1.12	-2.11	-2.92	-2.53	
Awase 2a	5	5	4	5	nd	2	4305	nd	nd	nd	52.11	0.08	0.76	0.29	0.05	0.19	0.02	-0.97	-2.38	-4.88	-2.97	-6.13
Awase 2b	5	5	3	3	nd	nd	4328	nd	nd	nd	52.74	0.07	0.75	0.32	0.04	0.13		-1.00	-2.24	-5.20	-3.55	
Awase 2c	5	6	nd	5	nd	nd	4161	nd	nd	nd	53.11	0.06	0.66	0.32		0.21		-1.18	-2.21		-2.82	
Awase 2d	5	4	nd	6	nd	nd	4152	nd	nd	nd	52.87	0.06	0.75	0.24		0.22		-1.00	-2.64		-2.77	
Awase 3a	7	6	7	4	nd	nd	4324	nd	nd	nd	51.22	0.08	0.97	0.35	0.09	0.17		-0.63	-2.11	-3.99	-3.16	
Awase 3b	6	6	5	7	nd	nd	4361	nd	nd	nd	52.46	0.07	0.88	0.32	0.06	0.27		-0.77	-2.21	-4.53	-2.48	
Awase 4	6	6	31	6	nd	nd	5010	nd	nd	nd	52.11	0.09	0.82	0.36	0.41	0.24		-0.87	-2.06	-1.86	-2.64	
Awase 5a	9	9	31	5	nd	nd	2161	0.25	1.29	nd	24.04	0.08	1.32	0.52	0.42	0.18		-0.18	-1.52	-1.84	-3.06	
Awase 5b	9	8	31	5	nd	nd	2182	0.28	1.32	0.02	26.53	0.08	1.32	0.45	0.42	0.18		-0.18	-1.73	-1.84	-3.06	
Awase 5c	8	7	23	5	nd	16	2570	0.19	0.78	0.01	35.62	0.08	1.12	0.42	0.31	0.21	0.19	-0.42	-1.82	-2.29	-2.82	-3.01
Awase 6a	8	6	8	6	nd	1	3698	0.02	0.20	nd	50.66	0.07	1.18	0.34	0.11	0.24	0.01	-0.35	-2.16	-3.79	-2.67	-7.13
Awase 6b	8	7	10	5	nd	2	3419	0.03	0.26	nd	48.59	0.07	1.19	0.39	0.13	0.18	0.03	-0.33	-1.95	-3.51	-3.03	-5.71
Awase 8	4	5	nd	5	nd	nd	3923	nd	nd	nd	54.11	0.05	0.54	0.28		0.20		-1.46	-2.44		-2.91	
Awase 9	4	4	nd	5	nd	nd	4000	nd	nd	nd	52.31	0.05	0.57	0.24		0.18		-1.39	-2.67		-3.06	
Awase 10a	7	5	nd	7	nd	5	4234	nd	nd	nd	53.11	0.08	1.06	0.28		0.29	0.06	-0.50	-2.41		-2.36	-4.71
Awase 10b	5	5	nd	5	nd	nd	4048	nd	nd	nd	52.11	0.07	0.78	0.29		0.20		-0.94	-2.38		-2.88	
Average	6	6	15	5		5	3858	0.15	0.77	0.01	48.05	0.07	0.90	0.34	0.20	0.21	0.06	-0.78	-2.18	-3.33	-2.87	-5.34
Minamigusuku (Mng)																						
MNG 1a	22	9	31	4	nd	18	2542		1.67	0.04	5.00	0.12	3.28	0.55	0.42	0.17	0.22	1.13	-1.46	-1.83	-3.12	-2.79
MNG 1b	21	10	35	5	nd	14	2345	0.23	1.86	0.04	32.85	0.12	3.12	0.61	0.47	0.21	0.16	1.06	-1.29	-1.67	-2.82	-3.20
MNG 1c	23	12	29	7	nd	23	2286	0.23	1.83	0.04	31.67	0.11	3.35	0.69	0.39	0.28	0.27	1.16	-1.11	-1.96	-2.42	-2.46
MNG 2a	11	8	24	8	nd	8	3123	0.12	1.05	106.00	43.95	0.11	1.57	0.45	0.32	0.32	0.09	0.07	-1.73	-2.23	-2.21	-4.03
MNG 2b	11	9	25	7	nd	19	3150	0.19	1.45	0.03	39.17	0.12	1.62	0.53	0.34	0.26	0.22	0.11	-1.50	-2.15	-2.53	-2.74
MNG 5a	6	6	6	4	nd		3831	nd	nd	nd	52.78	0.10	0.91	0.38	0.08	0.16		-0.72	-1.99	-4.16	-3.19	
MNG 5b	7	6	7	6	nd		3682	nd	0.04	nd	53.11	0.11	0.99	0.34	0.10	0.23		-0.61	-2.16	-3.95	-2.72	
MNG 5c	5	5	7	6	nd		3591	nd	nd	nd	52.21	0.11	0.74	0.29	0.10	0.24		-1.03	-2.38	-3.92	-2.67	
Okubu 1a	4	5		6	nd		4314	nd	nd	nd	52.11	0.07	0.57	0.27	0.00	0.24		-1.39	-2.47		-2.62	
Okubu 1b	3	6	2	6	nd	2	4460	nd	nd	nd	52.11	0.08	0.40	0.36	0.03	0.22	0.02	-1.92	-2.04	-5.50	-2.77	-6.05
Average	11	8	19	6		14	3332	0.19	1.32	21.23	41.50	0.10	1.65	0.45	0.23	0.23	0.17	-0.21	-1.81	-3.04	-2.71	-3.55
Nakagusuku (Nkg)																						
Nakagusuku-1	11	7	11	8	nd	nd	3530	nd	0.70	nd	53.50	0.07	1.66	0.39	0.15	0.30		0.15	-1.93	-3.28	-2.32	
Nakagusuku-2a	4	5		7	nd	nd	3974	nd	nd	nd	51.96	0.08	0.60	0.31		0.27		-1.31	-2.27		-2.46	
Nakagusuku-2b	4	5	4	5	nd	nd	3928	nd	nd	nd	52.78	0.07	0.57	0.31	0.06	0.20		-1.39	-2.27	-4.72	-2.91	
Nakagusuku-3	7	7	5	4	nd	nd	3848	nd	nd	0.01	53.11	0.08	0.99	0.39	0.07	0.17		-0.61	-1.95	-4.39	-3.16	
Hyakuna 1a	3	4		4	nd	nd	3634	nd	nd	nd	52.11	0.07	0.43	0.23		0.16		-1.81	-2.71		-3.27	
Hyakuna 1b	3	4		4	nd	nd	3634	nd	nd	nd	53.44	0.07	0.49	0.23		0.18		-1.63	-2.71		-3.09	
Average	5	5	7	5			3758				52.82	0.07	0.79	0.31	0.09	0.21		-1.10	-2.30	-4.13	-2.87	
JUC average	7	17	74	25	38	84	*na*	0.62	5.39	0.11	3.39	0.12										
UCC average	5	17	67	28	47	92	*na*	0.64	5.04	0.10	3.59	0.15										

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

4.2 Major and Trace Element Abundances

The elemental concentrations, the average value of the trace and major element analyses in the Awase tidal flat, Minamnigusuku, and Nakagusuku surface sediments are shown in Table1. In addition, this table includes the contamination factor and geoaccumulation index values, as well as the average values of Japan upper crust (JUC; Togashi et al.,2000) and upper continental crust (UCC; Rudnick & Gao, 2005).

4.2.1 Awase Tidal Flat

In the Awase tidal flat sediments, the As, Pb, Zn, Cu, and TS average 6, 6, 15, 5, 5, and 3858 mg/kg, respectively. The average abundances of the major elements were 0.15 wt.% for TiO_2, 0.77 wt.% for Fe_2O_3, 0.01 wt.% for MnO, 48.05 wt.% for CaO, and 0.07 wt.% for P_2O_5.

4.2.2 Minamigusuku

On average the trace elements showed 11mg/kg As, 8mg/kg Pb, 19mg/kg Zn, 6mg/kg Cu, 14mg/kg Cr, and 3332mg/kg TS. TiO_2, Fe_2O_3, MnO, CaO, P_2O_5 averaged 0.19, 1.32, 21.23, 41.50, and 0.10 wt%, respectively.

4.2.3 Nakagusuku

In the Nakagusuku area, the average concentrations of trace elements indicated 5mg/kgAs, 5mg/kg Pb, 7mg/kg Zn, 5mg/kg Cu, and 3758mg/kg TS. CaO 52,82wt.% and P_2O_5 0.07 wt.% were essentially the two major elements reported in this area because TiO_2 was not detected in the entire samples, and Fe_2O_3 and MnO were only detected in two sediment samples, particularly in the sediment sample nakagusuku-1 (Fe_2O_3 0.70wt%) and sample nakagusuku-3 (MnO 0.10w%).

Overall, Ni was not detected in all the study areas, as were Cr and TiO_2 in the Nakagusuku area. The highest average concentrations of As (11mg/kg), Pb (8mg/kg), Zn (19mg/kg), Cu (6mg/kg), and Cr (14mg/kg) were reported in the Minamigusuku area. The statistical summary of the concentrations of As, Pb, Zn, Cu, and Cr in the three study areas are shown in Figure 2.

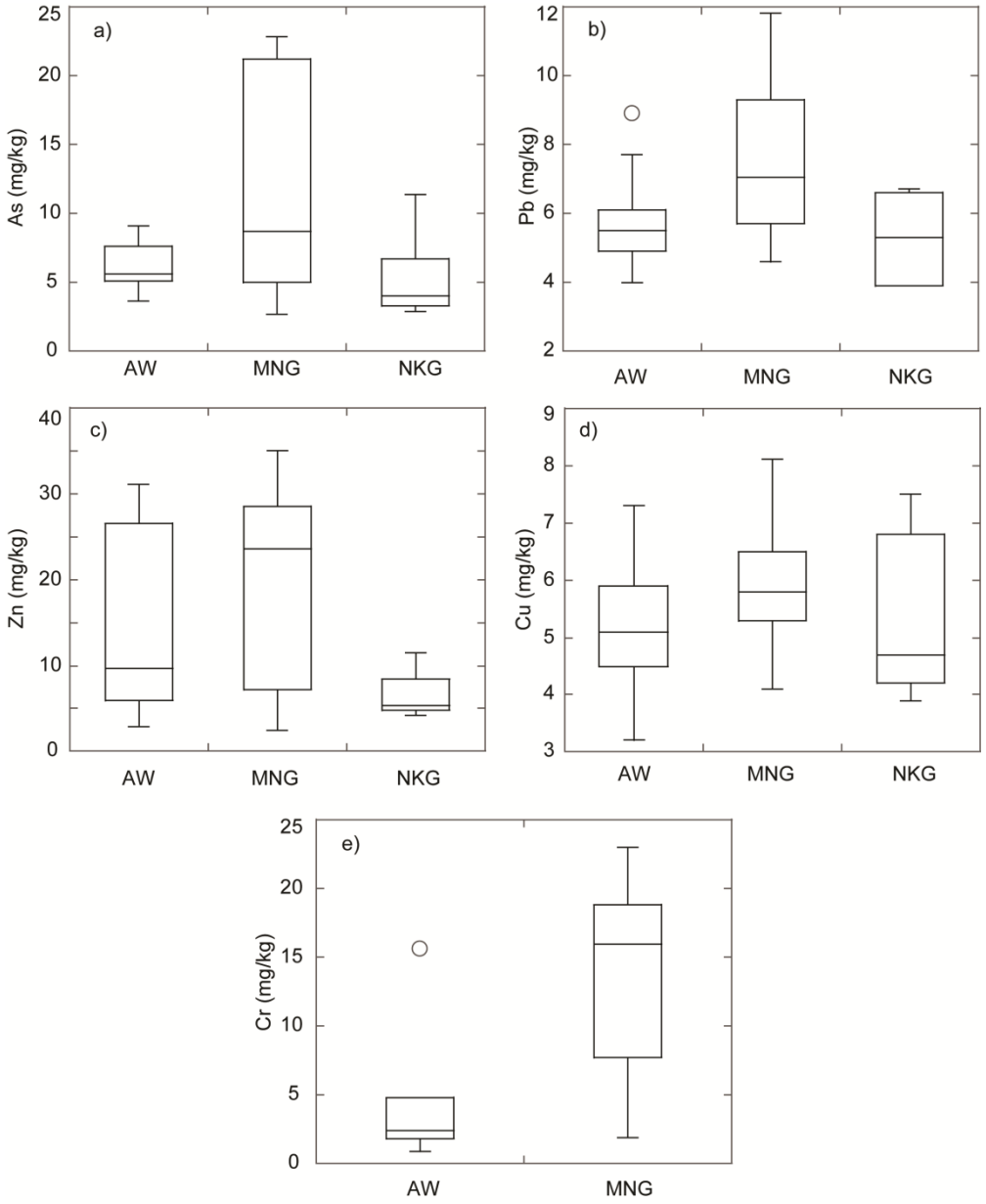

Figure 2 a-e Statistical summary of the trace metal concentrations in the Awase tidal flat (AW), Minamigusuku (MNG), and Nakagusuka (NKG) sediments .Vertical lines give the range (min. and max.), excluding outliers(circles); boxes show the first quartile (lower) and the third quartile (upper); the horizontal lines within the boxes indicate the median.

4.3 Normalization to JUC and UCC

The average concentrations of As, Pb, Zn, Cu, and Cr in the study areas were normalized to JUC and UCC to evaluate the enrichment level of the trace metals. This normalization is displayed in figure3. This figure indicates that in the Awase tidal flat, Minamigusuku, and Nakagusuku surface sediments, Pb, Zn, Cu, and Cr are depleted with respect to both JUC and UCC, as is As in the Awase tidal flat and Nakagusuku areas with respect to JUC. However, the concentrations of As in the Awase tidal flat and the Nakagusuku areas are slightly enriched

relative to UCC, whereas in the Minamigusuku area As is considerably enriched with respect to the reference values, particularly with UCC, suggesting a potential risk to biota in this area.

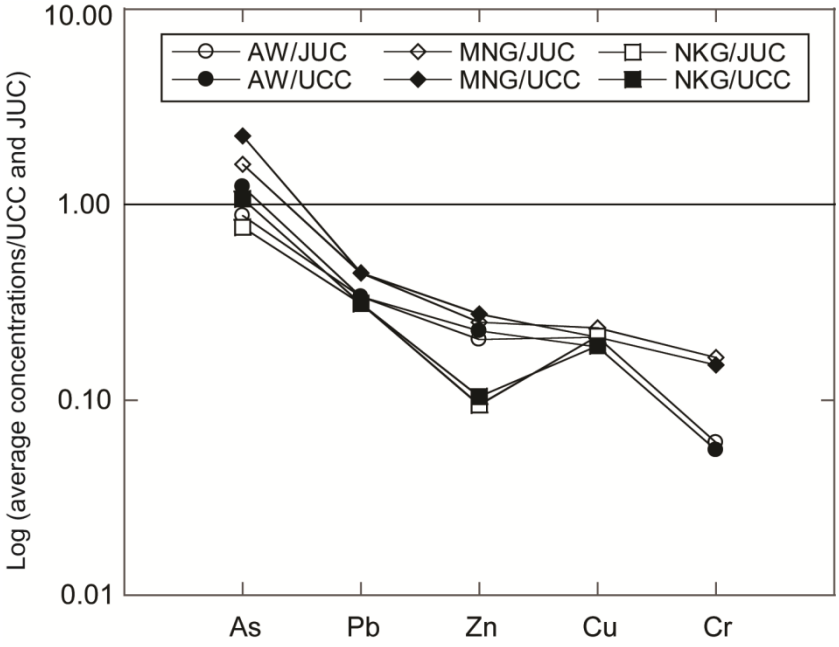

Figure3. average metal concentrations in the Awase tidal flat (AW), Minamigusuku (MNG), and Nakagusuku (NKG) normalized to UCC upper contimental crust (Rudnick & Gao,2005) and to JUC Japan upper crust (Togashi et al.2000)

4.4 Contamination Factor

The contamination factor values derived from the Equation (1) are presented in Table 1. The CF values indicate that the concentrations of Pb, Zn, and Cu in all the study areas, as well as those of Cr in the Awase tidal flat and Minamigusuku surface sediments represent background values (CF $\leq$1). In contrast, relative to As, low contamination (CF1-3) are reported in six sediment samples of Awase tidal flat (Awase5a, Awase5b, Awase5c, Awase6a, Awase6b, and Awase10a), in two sediment samples of Minamigusuku (Mng 2a and Mng 2b), and in one sediment sample of Nakagusuku (Nakagusuku-1). Furthermore, with respect to As, considerable contamination (CF3-6) was observed in three sediment samples of Minamigusuku (Mng1a, Mng1b, and Mng1c).

On average, the contamination factor values indicate that only the Minamigusuku area shows moderate enrichment relative to As, which represents the most significant values among the selected trace elements in the study areas.

4.5 Geoaccumulation Index

Computed from Equation (2), the geoaccumulation index values are shown in Table1. These values indicate that all the Awase tidal flat sediment samples are practically unpolluted with respect to As, Pb, Zn, Cu, and Cr (Igeo<0). Similarly, the geoaccumulation index values of Pb, Zn , Cu, and Cr in the Minamigusuku and Nakagusuku areas show no sign of pollution (Igeo<0). However, the geoaccumulation index values of As indicate that half of the sediment samples in the Minamigusuku are unpolluted, whereas two samples (mng2a, mng2b) are rated unpolluted to moderately polluted, and three samples (mng1a,mng1b, and mng1c) are moderately polluted. Finally, in the Nakagusuku area, almost all sediment samples are practically unpolluted with respect to As, except the sampling site Nakagusuku-1, which is rated as unpolluted to moderately polluted (Igeo 1-2).

4.6 Sediment Quality Guidelines

The average concentrations of As, Pb, Zn Cu, and Cr (Table2) were compared to US EPA sediment quality guidelines (Perin et al.,1997) to evaluate the sediment quality in the Awase tidal flat, Minamigusuku, and Nakagusuku areas. Among the considered trace metals, only As presents significant values in the study areas.

Relative to As, Awase tidal flat and Nakagusuku areas are moderately polluted, whereas Minamigusuku area is heavily polluted, implying that As may possibly impact the biota in these three areas.

Table2 Average trace metal concentrations in the Awase tidal flat, Minamigusuku, and Nakagusuku surface sediments compared to the US EPA SQG

Element	Sampling area			US EPA SQG		
	Aw	Mng	Nkg	Nonpolluted	Moderately polluted	Heavily polluted
As	6	11	5	<3	3-8	>8
Pb	6	8	5	<40	40-60	>60
Zn	15	19	7	<90	90-200	>200
Cu	5	6	5	<25	25-50	>50
Ni	nd	nd	nd	<20	20-50	>50
Cr	5	14	nd	<25	25-75	>75

Aw Awase tidal flat; *Mng* Minamigusuku; *Nkg* Nakagusuku; *US EPA SQG* United States environmetal protection agency sediment quality guideline (Perin et al., 1997)

5. Conclusions

Geochemical analysis of the Awase tidal flat, Minamigusuku, and Nakagusuku surface sediments was performed in this study. The results show that among the selected trace elements (As, Pb, Zn, Cu, Ni, and Cr), Ni was not detected in any of the study areas, as was Cr in the Nakagusuku area. The highest average concentrations of As (11mg/kg), Pb (8 mg/kg), Zn (19 mg/kg), Cu (6 mg/kg), and Cr (14mg/kg) occurred in the Minamigusuku area.

Normalized values to JUC and UCC show that As is slightly enriched relative to UCC in the Awase tidal flat and Nakagusuku areas, whereas in the Minamigusuku area As is considerably enriched with respect to JUC and UCC, suggesting a potential risk to biota in this area.

On average, the contamination factor values indicate that only the Minamigusuku area shows moderate enrichment relative to As, which represents the most significant values among the selected trace elements in the study areas.

The geoaccumulation index values of all the selected trace elements show that the Awase tidal flat and Nakagusuku areas are practically unpolluted. Similarly, the geaccumulation index values of Pb, Zn, Cu, and Cr show no sign of pollution in the Minamigusuku area, whereas relative to As in this same area half of the sediment samples display significant values, ranging from moderate to considerable contaminations.

In comparison to the US EPA sediment quality guidelines, the most important values were observed with As, which shows moderate pollution in the Awase tidal flat and Nakagusuku areas, and heavy pollution in the Minamigusuku area, implying that As may possibly impact the biota in these locations.

Overall, based on the results of the study, regular monitoring of As need to be carried out in the three study areas, particularly in the Minamigusuku area where heavy pollution occurred in some of the sediments samples.

Aknowledgements

We thank the Japanese Government for financial support, through a Japanese Government (Monbukagakusho) Scholarship to the senior author.

References

Håkanson, L. (1980). An ecological risk index for aquatic pollution control—a Sedimentological approach. *Water Research, 14*, 975–1001. http://dx.doi.org/10.1016/0043-1354(80)90143-8

Konishi, K., Kaneshima, K., Nakagawa, K., & Sakai, H. (1972). Pleistone dolomite and associated carbonates in south Okinawa. *the Ryukyu Islands. Geochemical Journal, 6*, 17-36. http://dx.doi.org/10.2343/geochemj.6.17

Müller, G. (1969). Index of geoaccumulation in sediments of the Rhine River. *Geological Journal, 2*, 108–118.

National Astronomical Observatory. (2004). *Chronological Scienctific Tables 2005.* Maruzen Co., Ltd., Japan, p.173,183 (in Japanese).

Naumih M. N., & Tamotsu, O. (2006). Evaluation of Heavy Metal Pollution on the Coastal Marine Environments of Okinawa Island, Japan. Bulletin Faculty Science, University. Ryukyushu, No.81: 93-104.

Ogasawara, M. (1987). Trace element analysis of rock samples by X-ray fluorescence spectrometry, using Rh anode tube. *Bulletin Geological Survey Japan, 38*(2), 57–68.

Perin, G., Bonardi, M., Fabris, R., Simoncini, B., Manente, S., & Tosi, L. (1997). Heavy metal pollution in central Venice Lagoon bottom sediments: evaluation of the metal bioavailability by geochemical speciation procedure. *Environmental Technology, 18*, 593–604. http://dx.doi.org/10.1080/09593331808616577

Potts, P. J., Tindle, A. G., & Webb, P. C. (1992). *Geochemical reference material compositions*. Whittles, Caithness.

Ramos, A. A., Inoue, Y., & Ohde, S. (2004). Metal contents in Porites corals: Anthropogenic input of river run-off into a coral reef from an urbanized area, Okinawa. *Marine Pollution Bulletin, 48*, 281–294. http://dx.doi.org/10.1016/j.marpolbul.2003.08.003

Ridgway, J., & Shimmield, G. (2002). Estuaries as repositories of historical contamination and their impact on shelf seas. *Estuarine, Coastal and Shelf Science, 55*, 903–928. http://dx.doi.org/10.1006/ecss.2002.1035

Rudnick, R. L., & Gao, S. (2005). The crust. In H. D. Holland & K. K. Turekian (Eds.), *Treatise on geochemistry, 3* (p. 537). Oxford: Elsevier Science.

Signh, K. P., Mohan, D., Signh, V. K., & Malik, A. (2005). Studies on distribution and Fractionation of Heavy metals in Gomti river sediment- a tributary of the Ganges, India. *Journal of Hydrology, xx*, 1-14.

Soares, H. M. V. M., Boaventura, R. A. R., Machado, A. A. S. C., & Esteves da Silva, J. C. G. (1999). Sediments as monitors of heavy metal contamination in the Ave river basin (Portugal): multivariate analysis of data. *Environmental Pollution, 105*, 311–323. http://dx.doi.org/10.1016/S0269-7491(99)00048-2

Takemitsu, A., Hiroyuki, F., Asha, M. H., Kouichirou, O., Hiroaki, K., Tamotsu, O., Akira, T. & Hatsuo, T.(2005). Simultaneous Measurement of Hydrogen Peroxide and Fe Species (Fe(II) and Fe(tot)) in Okinawa Island Seawater: Impacts of Red Soil Pollution. *Journal of Oceanography, 61*, 561-568.

Togashi, S., Imai, N., Okuyama-Kusunose, Y., Tanaka, T., Okai,T., Koma, T., & Murata, Y. (2000). Young upper crustal chemical composition of the orogenic Japan Arc. *Geochemical Geophysic Geosystem, 1*.

Vuai, S. A., & Tokuyama, A. (2011). Trend of trace metals in precipitation around Okinawa Island, Japan. *Atmospheric Research, 99*, 80–84. http://dx.doi.org/10.1016/j.atmosres.2010.09.010

West, K., & Van Woesik, R. (2001). Spatial and temporal variance of river discharge on Okinawa (Japan): inferring the temporal impact on adjacent coral reefs. *Marine Pollution Bulletin, 42*, 864–872. http://dx.doi.org/10.1016/S0025-326X(01)00040-6

Yang, H., & Rose, N. (2005). Trace element pollution records in some UK lake sediments, their history, influence factors and regional differences. *Environmental International, 31*, 63–75. http://dx.doi.org/10.1016/j.envint.2004.06.010

Young, E. (2007). Can 'fertilizing' the ocean combat climate change? Companies are planning to boost the ocean's plankton, hoping they will harvest more CO2 from the air. But will it work? *New Science, 15*, 42–45. http://dx.doi.org/10.1016/S0262-4079(07)62348-3