

Environmental change of the fluvial-estuary system in relation to Arase Dam removal of the Yatsushiro tidal flat, SW Kyushu, Japan

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Abstract Chemical composition of 24 elements for sediments and suspended solids, water parameters and the diatoms from Yatsushiro Bay tidal flat, Kuma River, and Arase Dam have been determined to examine changes from 2002 to 2013. In 2002, sediment delivered to the bay by the Kuma River was restricted by the Arase Dam; however, in April 2010 two gates were opened and also repeated seasonal flushing allowing resumption of natural sediment transport. Bulk chemical composition of the tidal flat sediments has changed since 2002, with marked decreases in concentrations of As, Zn, Pb and total sulfur. Decreased heavy metal contents in 2012 and 2013 for bay sediments after opening the dam and repeated flushing is attributed to dilution by previously impounded quartz and feldspar. Restoration of natural sediment transport by opening the dam and repeated flushing has thus brought about a recovery in the Yatsushiro marine environment.

Keywords Marine environment · Kuma River · Tidal flat · Yatsushiro Bay · Arase Dam · Recovery

Introduction

Tidal flats constitute an important interface between land and sea and between marine and fresh water systems (Cardoso et al. 2008). They are enriched in nutrients by oceanic and continental inputs and are among the most productive aquatic ecosystems (Nixon 1988; Cardoso et al.

2008). Coastal lagoons, one typical estuarine environment, receive the by-products of inland human and natural activities. Human activity in coastal lagoons has increased considerably in recent years, and its impact on these productive and economically important environments has become a major concern (Cardoso et al. 2008; Morrison et al. 2001). Therefore, tidal flat sediments are among the most important indicators of changes in sediment accumulations and aquatic system geochemistry. Although lake sediments may be better indicators of nutrient and metal accumulation, marine bays and estuaries also record input changes, although they may be less effective in retaining nutrients (Nixon 1988; Carriquiry et al. 2011).

Marine sediments serve as the ultimate sink for heavy metals discharged into the aquatic environment (Luoma and Bryan 1981). The fate and flux of heavy metals discharged from continent to ocean is controlled by biogeochemical and sedimentological processes taking place in estuaries (Monbet 2006). Metals accumulate in bottom sediments through several types of reactions: flocculation, coagulation, formation of particulate Fe and Mn oxyhydroxides and absorption on suspended matter (Sholkovitz 1976; Zwolsman et al. 1997; Nair et al. 2013).

Sediments reflect the geochemical history of a given region including the local anthropogenic impact. Hence, sediments act as useful indicators of long- and medium-term metal flux. Higher concentrations of heavy metals in sediments may not necessarily indicate anthropogenic contamination, due to different background levels in parent materials and sediment properties. The natural occurrence and chemical speciation of metals can also complicate the evaluation of potentially polluted aquatic sediments (Schiff and Weisberg 1999).

Nutrients and suspended sediment concentration are higher in the inner areas of the coastal lagoon than in the

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tidal channel (Schwindt et al. 2004). Diatoms (Bacillariophyceae) are also an important and often dominant component of benthic microalgal assemblages in estuarine and shallow coastal environments (Sullivan 1999). Different environmental factors may influence the diatom distributions observed in each zone of the lagoon. Therefore, SEM images of diatoms in suspended solids can be very useful for understanding the prevailing estuarine environment.

Dam is a construction crossing the river and may change a fluvial system (Xiangbin et al. 2013). When a river is dammed, it may affect not only the decrease of water flow, but also the sediments deposited on the upstream side of the dam. And the dam's sustainability can be threatened by rapid sedimentation of detritus (Haregeweyn et al. 2012). The Arase Dam is the first dam to be removed in Japan and this will take ~ 7 years for completion. This action was implemented as a mitigation strategy based on sediment properties and sedimentation impacts (Haregeweyn et al. 2012; Trabelsi et al. 2012). The aim of this study is to identify and document the change of geoenvironmental conditions of the Yatsushiro Bay sediments after repeated flushing and removal of the Arase Dam gates.

Study area

The Arase Dam is situated in Kumamoto Prefecture, southwest Kyushu, Japan. This dam is on the Kuma River, about 19.9 km from the Yatsushiro tidal flat. The process of removing the Arase Dam was begun in April, 2010. The Arase Dam consists of eight gates. Removal of two gates was initiated in April 2010 to first flush out the excess water and then sediments deposited above the dam. Repeated gate openings especially in the winter season have been used to control water levels (Kumamoto Prefecture Government from personal communication) for these several years.

The Yatsushiro Sea is an inlet of ca. 1,200 km², with a tidal range during the spring tide reaching ca. 4 m in the innermost part and ca. 3 m in the southern part of the inlet. Sandy and muddy tidal flats of ~ 46 km² develop in the northern region of the bay (Subiyanto et al. 1993). Yatsushiro Bay is a semi-closed estuary opening to the East China Sea. The Kuma River flows into Yatsushiro Bay forming a large tidal flat at the river mouth (Fig. 1). These tidal flats are formed in the southern region of the Uto Peninsula and along the western edge of the Yatsushiro Plain (Fig. 1). The Hikawa River flows along the Hikawa District and is open to the Yatsushiro Sea (Fig. 1). The drainage area of the Kuma River above the Arase Dam is 1,880 km². Typical sea level in the Yatsushiro Sea ranges between +4.67 and -0.16 m.

Geology of the study area

The basement geology of central Kyushu is composed of four geologic terranes. The Yatsushiro Bay basement is composed of the Higo terrane while that underlying the Kuma River is the muddy Jurassic melange of the Chichibu terrane. The Usuki–Yatsushiro tectonic line divides the Higo terrane and the Chichibu terrane (Fig. 1, Matsumoto and Kanmera 1964; Yamamoto 1992). The Higo terrane consists of three Jurassic metamorphic zones; the Ryuhozan, the Manotani and the Higo metamorphic rocks. The Ryuhozan metamorphics are similar to the Ryoke and consist of granitic gneiss and granites. The Manotani metamorphic rocks consist of pelitic schist and basic schist (Matsumoto and Kanmera 1964; Yamamoto 1992). The Higo metamorphics are psamite calcareous schist yielding the ultra mafic rocks and limestone of southwest Japan. The geology of the Yatsushiro Bay area is made up of Cretaceous–Paleogene sedimentary rocks, the Chichibu terrane, and the Higo terrane (Matsumoto and Kanmera 1964; Yamamoto 1992). The Kuma River primarily flows in the Chichibu terrain (Fig. 1). The Higo volcanic rocks (mainly andesite) are widely distributed south of the map area (Fig. 1, Matsumoto and Kanmera 1964; Yamamoto 1992). Thus, the provenance of Kuma River sediments is the Chichibu terrane and the Higo volcanics.

Methodology

Stream sediments and tidal flat sediments were collected at locations plotted in Fig. 1, also taking into account the previously studied locations of Dozen and Ishiga (2002). Sediment samples were collected from (a) the downstream area of the Kuma River; (b) the Yatsushiro tidal flat at the mouth of the Kuma River; (c) the southern area of the Uto Peninsula and (d) the western margin of the Yatsushiro Plain (Fig. 1). These locations cover the northeast region of the Yatsushiro Sea. Approximately 500 g of bulk sediment samples were collected using a hand pit method at 1–6 cm depths. Sample collection was done during low tides (27–35 cm) from 24th to 27th March 2012. The samples were stored at 4 °C in double sealed bags during transport for analysis.

Approximately 50 g of each sample was oven dried at 160 °C for 48 h before being crushed to fine powder in an automatic agate mortar and pestle grinder. Sediment samples were then compressed into briquettes, using a force of 200 KN for 60 s. Portions (7–10 g) of the crushed materials were transferred into glass vials and dried at 110 °C for at least 24 h prior to determination of loss on ignition (LOI). Gravimetric LOI determinations were calculated from the net weight loss after ignition in a muffle furnace at

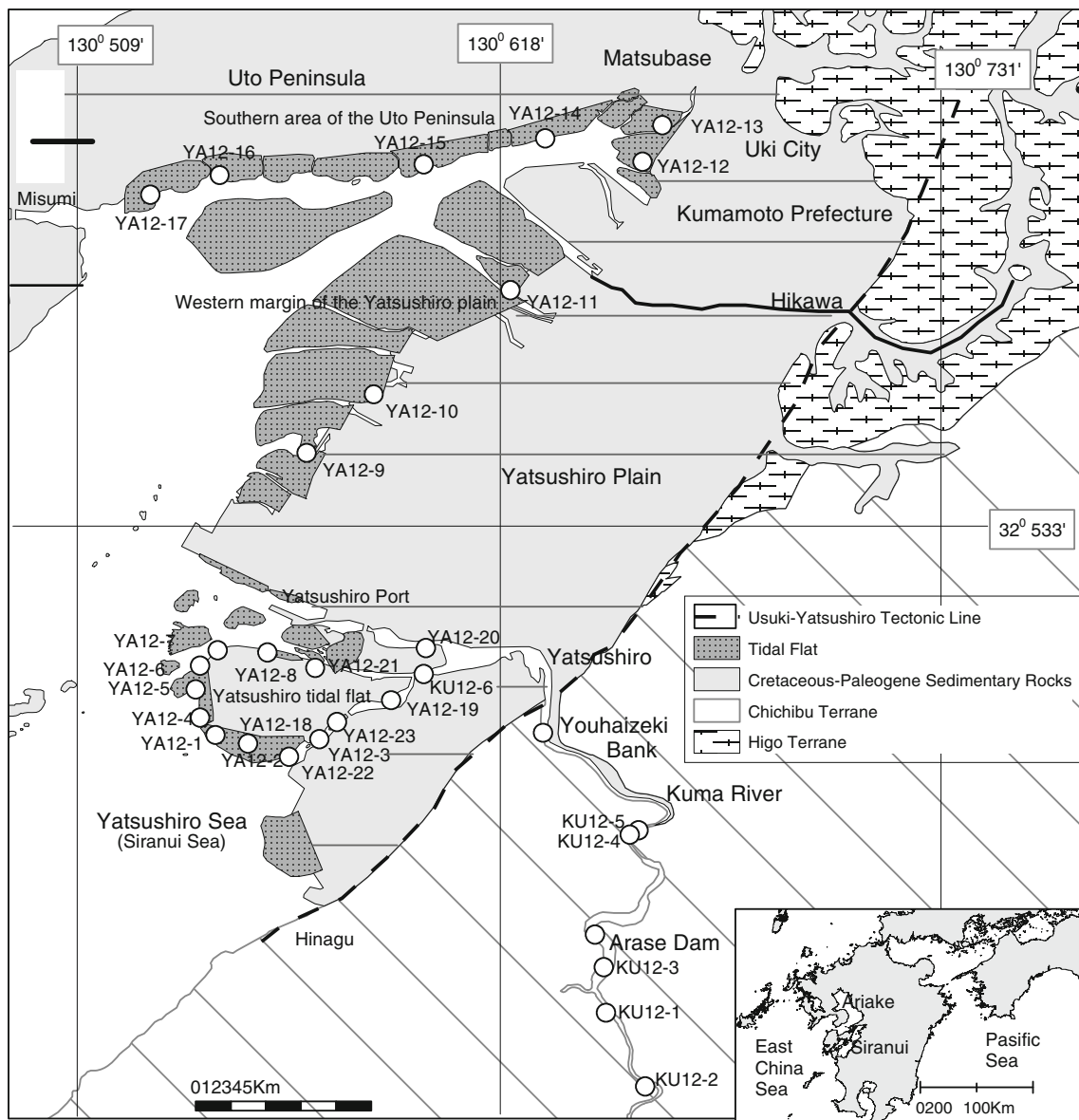


Fig. 1 Simplified geological map and sediment sampling locations of the Kuma River and the Yatsushiro tidal flat in South West Kyushu, Japan

1,020 °C for at least 2 h. Glass fusion beads for X-ray fluorescence analysis (anhydrous basis) were prepared with an alkali flux (80 % lithium tetraborate, 20 % lithium metaborate), using a sample to flux ratio of 1:2. Analysis of 24 major and trace elements were then determined by X-ray fluorescence spectrometry using a Rigaku RIX-2000 spectrometer equipped with an Rh-anode tube at Shimane University. Analytical methods, instrumental conditions, and calibration followed those described by Ogasawara (1987) for press discs and Kimura and Yamada (1996) for glass fusion beads.

The grain size analysis was done using a laser diffraction particle size analyzer SALD-3000 and a settling tube grain size analyzer at Shimane University after treating the

oven-dried sediments with 30 % H₂O₂ for at least 24 h to digest the organic matter. Sediments consisting of grains <3 mm have been measured using the SALD-3000S grains >3 mm were measured using a settling tube. Calculations of grain size and sorting were made following Folk and Ward (1957).

Water physical parameters, temperature, pH, electrical conductivity (EC), dissolved oxygen (DO) and oxygen reduction potential (ORP) were determined in the field using a Horiba D-24 pH/conductivity meter. Water samples were collected with the aim of obtaining suspended solids. Water samples were collected in plastic bottles and stored at 4 °C during transport to Shimane University. The water samples were filtered to obtain the suspended solids using

Whatman 45 μm quartz filter paper. New filter papers were dried at 110 °C for at least 2 h and the weight was recorded prior to filtering. The filtrate was then oven dried at 110 °C for at least 2 h and the weight of filtrate was recorded. The filtered suspended solids were then analyzed for 19 major and trace elements by X-ray fluorescence spectrometry using a Rigaku RIX-2000 spectrometer equipped with an Rh-anode tube at Shimane University. The Mn content of water samples was determined with an on-site chemical analysis kit for trace components in water using a color development method for the filtrate. The average errors for all elements are less than $\pm 10\%$ relative.

The suspended solids were examined using a scanning electron microscope (SEM) to determine the diatoms in the sediments. SEM images of all the detected species of the Yatsushiro Bay and the Kuma River were recorded.

Results

Grain size and texture

Kuma River sediments are fine to medium grain with a mean grain size (MGS, phi scale) of 1.45–2.56 and very poorly sorted (Table 1). Except four samples that consist of very coarse sand to granules (MGS -1.83 to -0.19) rest of the Yatsushiro tidal flat at the mouth of the Kuma River range within coarse sand to fine sand with MGS of 0.45–2.87. Western margin of the Yatsushiro Plain are fine sand to very fine sand with MGS of 2.55–3.64. Sediment grain size in the southern region of the Uto Peninsula is mostly medium sand to very fine sands with MGS of 0.95–3.84 (Table 1). The Yatsushiro tidal flat, Western margin of the Yatsushiro Plain, southern region of the Uto Peninsula sediments are very well sorted to very poorly sorted.

Major elements

Most trace and major elemental concentrations are higher in Yatsushiro tidal flat sediments and lower in the Kuma River sediments (Table 1). The highest SiO_2 content are relatively uniform in composition in the Kuma River sediments (72.27–75.35 wt%). The Lowest SiO_2 content are seen in the southern region of the Uto Peninsula sediments (45.61–59.79 wt%). The Al_2O_3 content is highest in the Western margin of the Yatsushiro Plain (16.79–21.44) and southern region of the Uto Peninsula sediments (14.91–21.30 wt%) which is higher than UCC 15.2 wt% (Taylor and McLennan 1985). This higher Al_2O_3 content is due to higher clay content in these two areas. The distribution of SiO_2 and the Al_2O_3 content in the sediments shows that the river and the river mouth

areas consist of the sand fractions while the distal ends consist of more clay fractions. The Fe_2O_3 and MnO content is highest (6.72–26.57; 0.20–0.29 wt%, respectively) in the southern region of the Uto Peninsula sediments (Table 1) and indicates that these sediments are the most anoxic of all. The other locations Fe_2O_3 contents are close to UCC (Taylor and McLennan 1985) which is 4.5 wt% and MnO in <0.11 for most locations. The LOI shows a clear difference between the Kuma River, Western margin of the Yatsushiro Plain and the southern region of the Uto Peninsula (Table 1). These high values of LOI in the bay areas may be mainly due to biogenic carbonates where the CaO content is also high in these bay areas.

Trace elements

Measureable chloride content occurs only in the Yatsushiro tidal flat samples (Table 1). The total sulfur (TS) of the Yatsushiro tidal flat is much higher than in Kuma River sediments. Yatsushiro tidal flat samples YA12-12 to YA12-17 have high elemental values—especially for Fe, Mn, P, Ti, Y, Ni, Cr, Cu, Zn, Pb and As (Table 1).

Suspended solids

Table 2 shows element concentrations for the suspended sediment (SS) of Yatsushiro Bay tidal flat and Kuma River. Yatsushiro Bay tidal flat SS shows higher values for almost all elements than those of the Kuma River, especially for Fe, Ti and total sulfur (TS). Chloride has been detected only in the Yatsushiro Bay tidal flat samples and not in the Kuma River sediments. Mn is at or below detectable levels in the suspended solids of the Yatsushiro Bay and the Kuma River.

Physical parameters

Table 3 shows chemical data of the field measurements for water and sediment of Yatsushiro Bay tidal flat and the Kuma River. The ORP, DO and EC ranges between (-40) and 300 mV, 6.7–8.3 mg/L and 0.312–6.2 mS/cm, respectively. The SEM images show that diverse diatom species are found in the Kuma River and Yatsushiro Bay.

Discussion

The Yatsushiro tidal flat area shows ripple formations formed by small-scale waves, which is a good evidence of sand deposited in the Yatsushiro tidal flat area. In addition, there are large bar formations in the bay originated by higher energy wave activity. There are also many sand bar

Table 1 Chemical analyses of Kuma River and Yatsushiro Bay bulk sediments for 2012

Sample	Major oxide (wt%)									
	SiO ₂	TiO ₂	Al ₂ O ₃	Fe ₂ O ₃	MnO	MgO	CaO	Na ₂ O	K ₂ O	P ₂ O ₅
Downstream area of Kuma River										
K12-1	72.27	1.14	12.09	7.53	0.12	2.03	1.35	1.42	1.97	0.09
K12-2	73.81	0.63	14.01	4.86	0.08	1.51	1.08	1.63	2.28	0.09
K12-3	74.31	0.64	13.65	4.81	0.09	1.54	1.07	1.57	2.23	0.09
K12-4	75.35	0.62	12.43	5.11	0.09	1.71	1.14	1.52	1.95	0.08
K12-5	74.79	0.57	13.30	4.78	0.09	1.75	1.04	1.51	2.10	0.08
Average	74.11	0.72	13.09	5.42	0.09	1.71	1.13	1.53	2.11	0.09
Yatsushiro Bay										
Yatsushiro Tidal Flat and Kuma River mouth										
Y12-1	64.08	0.93	19.22	5.30	0.14	2.51	1.41	3.15	3.01	0.25
Y12-2	73.81	0.63	14.01	4.86	0.08	1.51	1.08	1.63	2.28	0.09
Y12-3	73.06	0.67	13.68	4.97	0.10	1.64	1.47	2.18	2.14	0.09
Y12-4	73.75	0.66	13.47	4.49	0.08	1.66	1.36	2.30	2.15	0.08
Y12-5	76.95	0.43	11.96	3.82	0.09	1.41	1.70	1.77	1.81	0.07
Y12-6	75.00	0.56	13.06	4.24	0.08	1.55	1.24	2.14	2.06	0.08
Y12-6a	75.03	0.59	13.17	4.55	0.07	1.60	0.95	1.87	2.09	0.08
Y12-7	75.03	0.59	13.17	4.55	0.07	1.60	0.95	1.87	2.09	0.08
Y12-8	72.63	0.62	14.98	4.37	0.09	1.61	0.99	2.17	2.45	0.08
Y12-19a	69.57	0.82	16.52	5.47	0.08	1.74	0.99	1.97	2.68	0.15
Y12-20	70.83	0.78	15.68	5.32	0.08	1.71	1.04	1.83	2.59	0.14
Y12-21	71.11	0.73	15.61	4.74	0.09	1.65	1.11	2.27	2.57	0.12
Y12-22	77.89	0.48	11.61	4.17	0.08	1.52	0.83	1.66	1.70	0.07
Y12-22a	74.65	0.67	13.02	4.79	0.08	1.69	1.00	1.93	2.09	0.08
Western margin of the Yatsushiro Plain										
Y12-9	67.46	0.77	16.98	4.77	0.10	2.22	1.61	3.21	2.71	0.16
Y12-10	66.80	0.78	16.79	5.04	0.09	2.15	2.95	2.62	2.64	0.15
Y12-12	60.17	0.97	21.44	7.29	0.26	2.86	1.30	2.54	2.95	0.23
Y12-13	60.48	0.97	21.32	7.15	0.30	2.86	1.18	2.65	2.86	0.22
Southern area of the Uto Peninsula										
Y12-14	59.79	0.98	21.30	6.72	0.27	3.00	1.30	3.36	3.07	0.22
Y12-15	59.63	1.15	19.40	8.26	0.23	2.65	2.50	3.37	2.64	0.17
Y12-16	55.40	0.96	18.88	9.29	0.20	3.18	7.29	2.64	2.00	0.16
Y12-17	45.61	2.69	14.91	26.57	0.29	3.43	3.26	1.89	1.20	0.15
Y12-shell(<i>Atrina pectinata</i>)	na	na	na	na	na	na	na	na	na	na
Average	68.12	0.81	15.92	6.19	0.13	2.08	1.58	2.32	2.35	0.12
UCC	66	0.5	15.2	4.5	0.1	2.2	4.2	3.9	3.4	0.15

Table 1 continued

Sample	Trace element (ppm)														LOI	MGS	Sorting				
	As	Pb	Zn	Cu	Ni	Cr	V	Sr	Y	Nb	Zr	Th	Sc	TS				F	Br	I	Cl
Downstream area of Kuma River																					
K12-1	6	13	76	25	29	116	201	148	19	10	133	8	13	341	472	1	5	nd	2.32	2.01	2.67
K12-2	6	13	74	26	28	80	142	158	21	10	139	8	13	414	258	2	20	nd	2.99	1.57	2.24
K12-3	6	14	70	27	28	91	131	147	21	10	144	9	11	382	6,608	2	17	nd	2.92	2.56	2.28
K12-4	6	13	68	25	37	98	135	145	19	9	127	8	10	347	nd	2	14	nd	2.48	-0.05	2.15
K12-5	6	14	72	25	50	102	134	143	20	9	128	8	10	358	nd	2	16	nd	2.85	1.45	2.77
Average	6	13	72	26	34	97	149	148	20	9	134	8	11	368	2,446	2	14		2.71	1.51	2.42
Yatsushiro Bay																					
Yatsushiro Tidal Flat and Kuma River mouth																					
Y12-1	13	26	109	47	45	92	169	168	29	12	175	12	11	4,778	41	59	24	9,029	18.10	2.61	0.16
Y12-2	8	17	80	23	32	94	126	172	24	10	185	10	10	2,745	2,261	16	19	3,586	5.93	1.00	0.41
Y12-3	8	17	105	24	30	91	123	177	20	9	197	8	9	1,888	nd	9	19	2,476	3.65	0.84	0.63
Y12-4	7	13	69	15	24	108	125	177	20	9	245	8	11	2,376	nd	13	23	7,076	3.82	0.67	0.61
Y12-5	7	14	65	12	22	59	96	175	17	7	107	6	10	1,138	1,325	4	26	nd	2.69	2.87	2.48
Y12-6	8	13	71	16	28	86	132	169	19	8	147	8	10	2,059	2,429	13	24	4,989	3.63	-1.83	2.86
Y12-6a	7	19	83	33	61	113	159	164	26	12	222	10	11	887	9,113	6	12	nd	6.64	1.27	0.37
Y12-7	7	15	69	18	29	86	115	152	20	9	151	8	7	1,166	38	8	18	268	3.22	1.48	0.36
Y12-8	7	16	70	22	28	88	129	161	23	10	152	9	9	1,648	1,402	11	23	2,195	3.94	-0.48	0.35
Y12-19a	8	17	86	31	38	82	162	167	26	12	207	11	11	1,466	125	9	15	nd	5.85	0.98	0.47
Y12-20	7	19	99	32	43	115	157	166	24	11	205	10	13	1,071	1,437	9	22	nd	5.58	0.45	0.50
Y12-21	8	16	80	22	29	93	137	168	24	10	194	10	13	1,422	nd	12	23	2,345	4.26	0.55	0.35
Y12-22	7	14	70	18	31	114	138	155	20	9	173	8	11	2,009	9,684	9	26	2,038	3.58	-0.77	1.44
Y12-22a	7	12	75	17	34	108	128	146	18	7	131	6	8	694	76	5	27	442	2.53	-0.19	1.79
Western margin of the Yatsushiro Plain																					
Y12-9	8	22	196	31	38	88	145	167	24	11	174	10	11	4,854	1,845	47	15	12,503	8.89	2.55	0.19
Y12-10	8	20	192	30	40	81	144	185	24	11	167	10	14	4,098	37	31	13	8,877	8.27	2.99	0.14
Y12-12	9	24	147	41	57	107	187	152	28	13	162	13	13	2,506	111	32	12	7,014	10.11	3.64	0.08
Y12-13	8	24	138	39	57	107	189	153	28	12	164	12	14	2,433	nd	33	10	6,892	9.90	3.63	0.08
Southern area of the Uto Peninsula																					
Y12-14	8	24	138	36	52	97	186	148	28	12	158	12	14	2,296	nd	52	18	10,473	11.02	3.84	0.07
Y12-15	7	20	119	26	39	84	205	214	23	10	135	10	15	2,070	nd	44	10	10,843	9.47	3.84	0.07
Y12-16	9	19	103	18	20	69	183	403	20	8	97	7	21	2,244	nd	24	11	6,684	8.99	2.77	0.18
Y12-17	4	11	127	11		70	696	234	15	9	123	4	18	1,329	nd	6	nd	1,631	3.28	0.95	2.06

Table 1 continued

Sample	Trace element (ppm)																				
	As	Pb	Zn	Cu	Ni	Cr	V	Sr	Y	Nb	Zr	Th	Sc	TS	F	Br	I	Cl	LOI	MGS	Sorting
Y12-shell (<i>Atrina pectinata</i>)	1	5	20	1	nd	nd	nd	724	2	nd	13	nd	71	6,532	20	3	nd	1,332	na	na	na
Average	7	16	97	25	36	93	169	193	22	10	155	9	14	1,928	2,091	16	18	5,300	6.52	1.53	0.71
UCC	1.5	20	71	25	20	35	60	350	22	25	190	10.7	11	na	557	1.6	1.4	370	na	na	na

Major elements as oxide wt% and trace elements are in ppm

UCC values as for Taylor and McLennan (1985)

MGS mean grain size, LOI loss on ignition. MGS and sorting calculated as in Folk and Ward (1957) (nd not detected, na not analyzed)

Table 2 Analyses of Kuma River and Yatsushiro Bay suspended solids

Sample	SS weight (mg/L)	Major elements (wt%)										Trace elements (ppm)												
		TiO ₂	Fe ₂ O ₃	MnO	CaO	P ₂ O ₅	As	Pb	Zn	Cu	Ni	Cr	V	Sr	Y	Nb	Zr	Th	Sc	TS	F	Br	Cl	Mn (mg/L)
Yatsushiro Bay																								
YA 12-6	153.60	0.94	7.06	0.16	1.02	0.16	8	16	95	40	44	91	184	38	9	4	55	4	14	2,603	152	16	3,667	0.00
YA 12-9	47.91	0.66	3.65	0.13	1.15	0.19	3	11	99	29	36	64	108	10	4	2	22	1	10	3,884	40	13	23,199	0/02
YA 12-11	8.91	0.54	2.74	0.10	1.05	0.22	2	11	56	23	33	48	91	6	4	2	24	1	10	3,612	300	13	39,362	0.00
YA 12-16	388.20	0.72	6.20	0.20	1.62	0.12	8	19	130	30	35	68	137	122	18	7	110	8	15	4,025	89	45	14,388	<0.02
YA 12-18	1,765.00	0.29	1.02	0.03	0.83	0.22	2	10	26	21	24	20	38	1	2	2	19	3	2,437	34	5	18,851	<0.02	
YA 12-19	12.06	0.42	1.31	0.05	0.84	0.19	2	7	692	43	25	60	44	6	4	2	22	1	4	1,107	179	3	nd	0.01
Kuma River																								
KU 12-1	10.27	0.22	0.69	0.04	0.87	0.15	1	10	21	16	31	11	24	nd	3	2	18	1	1	637	69	2	nd	0.00
KU 12-6	6.91	0.13	0.38	0.02	0.81	0.13	1	10	28	21	30	7	23	nd	2	2	18	nd	2	595	2	2	nd	0.00

Major elements as oxide wt% and trace elements are in ppm

nd not detected

Table 3 Physical and chemical properties of sediment and water from the Kuma River and Yatsushiro Bay

Sample	Time	pH	EC (mS/cm)	ORP (mV)	DO (mg/L)	T °C	Type	Mn (mg/L)
Yatsushiro Bay								
YA 12-6	11.04	na	na	na	na	na	Water	0
YA 12-9	15.50	na	na	na	na	na	Water	0
YA 12-11	12.32	7.32	2.63	177	6.7	12.9	Water	0
YA 12-12	13.10	7.48	na	33	na	na	Sediment	NA
YA 12-16	14.33	na	na	na	na	na	Water	<0.02
YA 12-18	11.00	7.95	1.42	215	7.6	12.6	Water	<0.02
YA 12-19	13.17	7.75	0.31	231	8.3	13.1	Water	0
YA 12-19	13.53	7.66	0.32	215	8.1	14.5	Water	0
YA 12-19	14.17	7.53	0.36	219	8	13.8	Water	0
YA 12-19a	14.32	7.40	na	-40	na	na	Sediment	NA
YA 12-20	15.11	7.51	na	52	na	na	Sediment	NA
Kuma River								
KU 12-1	8.52	na	na	na	na	na	Water	0
KU 12-3	10.24	7.64	6.70	250	7.55	11.80	Sediment	NA
KU 12-6	11.04	7.75	6.60	300	7.73	12.40	Water	0
Min		7.40	0.31	-40	6.7	11.8		0
Max		7.95	6.70	300	8.3	14.5		<0.02

NA not applicable, na not analyzed

formations along the Kuma River resulted from periodic flushing of sediments after opening the dam gates.

Grain size

Marshes of the Yatsushiro Bay area are comparatively young (Yamamoto 1992) and lack fine material. The tidal flat along the western margin of the Yatsushiro Plain may have resulted from sediment accumulation rather than erosion. Large amounts of fine-grained material remain suspended in the water column, and may undergo transport towards the tidal flats of the southern area of the Uto Peninsula and then to the open sea.

Mean grain size variation in Kuma River and Yatsushiro Bay sediments of 2012 are given in Table 1. Most of the Yatsushiro tidal flat at the mouth of the Kuma River range within coarse sand to fine sand while the Western margin of the Yatsushiro Plain are fine sand to very fine sand. Sediment grain size in the southern region of the Uto Peninsula is mostly medium sand to very fine sands. Thus, the grain size gradually decrease from Kuma River > Yatsushiro tidal flat > Western margin of the Yatsushiro Plain > to the southern region of the Uto Peninsula. Grain size sorting is calculated as in Folk and Ward (1957, Table 1). The southern area of the Uto Peninsula in Yatsushiro Bay is very well sorted compared to the other samples. The river brings in coarser material, which is deposited proximal to the river mouth, while the finer particles are transported and deposited in the distal southern area, the Uto Peninsula of Yatsushiro Bay. This phenomenon occurs because of the anticlockwise water

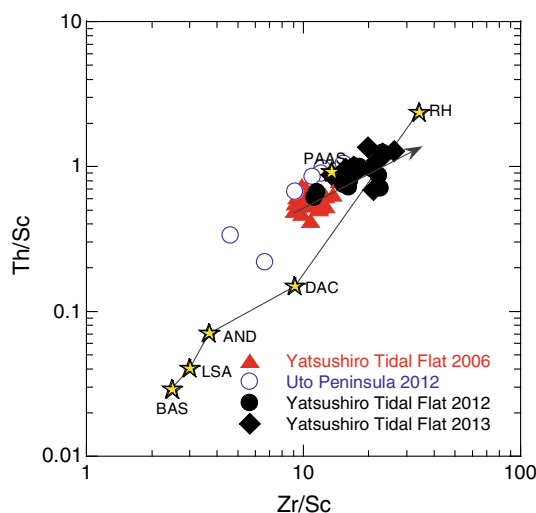


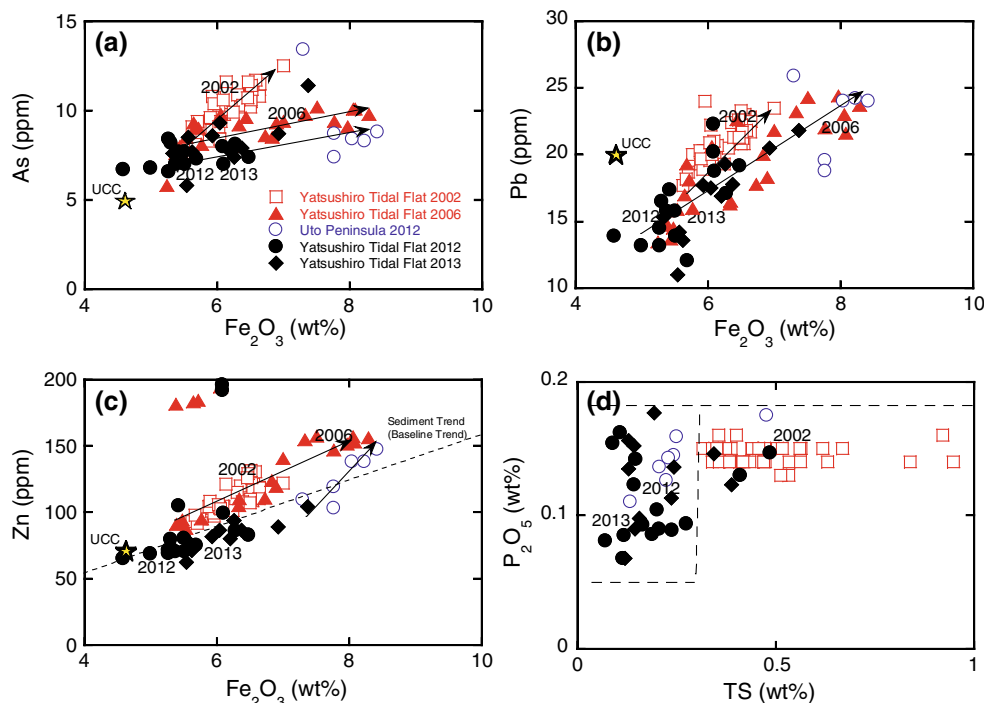
Fig. 2 Zr/Sc–Th/Sc (McLennan et al. 1993) for the Yatsushiro Bay sediments. Stars BAS, LSA, AND, DAC, RHY: average basalt, low-silica andesite, andesite, dacite and rhyolite, as plotted by Roser and Korsch (1999), representing a model source evolution trend. PAAS Post-Achaean Australian Shale (Taylor and McLennan 1985). Arrow trend line showing the shift of ratios towards the line from 2006 to 2013

circulation in the Yatsushiro Sea and because of the tidal variation. Grain size variation therefore shows a major control over the chemistry of bay and river sediments.

Provenance indicators

Possibility of provenance, sorting or accumulation of heavy minerals (zircon, monazite, or apatite) can be evaluated

Fig. 3 Plots of **a** As, **b** Pb, **c** Zn versus Fe_2O_3 and **d** P_2O_5 versus TS for Kuma River and Yatsushiro Bay sediments. Data for 2002 is from Dozen and Ishiga (2002), unpublished data from Ishiga (2006), unpublished data from Young and Ishiga (2013). *Sediment trend line* for Zn has been drawn as for Ahmed et al. (2007)



using Zr/Sc and Th/Sc ratios (McLennan et al. 1993). Bulk composition of the source can be evaluated using Th/Sc ratio which is a sensitive index (Taylor and McLennan 1985), whereas Zr/Sc ratio serves as a proxy for identifying heavy mineral concentrations, because it is highly sensitive to accumulation of zircon. Consequently, plot positions and trends on bivariate Zr/Sc – Th/Sc plots give an indication of source composition and heavy mineral concentration when compared with compositions of average volcanic and plutonic rocks. The Th/Sc – Zr/Sc plot for Yatsushiro Bay shows that the composition has changed since 2006–2013 (Fig. 2). The Yatsushiro Bay sediments have moved towards the composition of PAAS from 2006–2012, and then moving toward the line on Rhyolite to Dacite in 2012–2013 with not much change (Fig. 2). Thus, in the Yatsushiro Bay the composition changes to the background detrital composition.

Environmental change between 2002 and the present

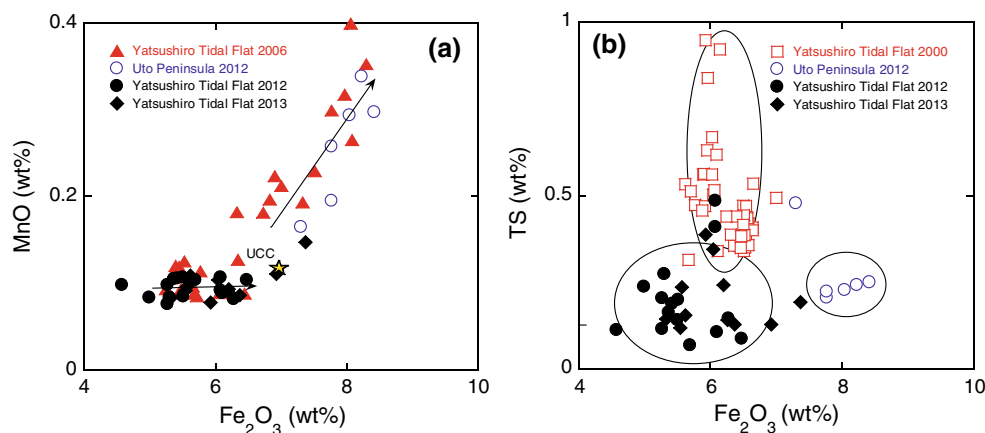
Tidal flats are good settings for the study of environmental changes in river sediments and estuaries (Kim et al. 2010; Liu et al. 2009). A large amount of sediment is brought into the Yatsushiro Bay by the Kuma River. The present environment has changed tremendously since the 2002 study of Dozen and Ishiga. Figure 3a–c shows the environmental change from 2002 to 2012.

Heavy metals are well known to be associated with the clay fraction (Robert et al. 2004). On the other hand, As, Pd and Zn are very sensitive to environmental changes and

they are easily absorbed on clay particles (Yuan et al. 2010). Iron is one of the main elements strongly associated with heavy metals and it can exist in several phases (Gurzaui et al. 2003; Violante et al. 2010). Thus, As, Pb and Zn have been cross plotted with Fe_2O_3 to show the environmental change since 2002. Arsenic, Pb and Zn all have gradually decreased since 2002–2013 (Fig. 3a–c). The 2002 and 2006 data reflect the sedimentary environment before opening of the Arase Dam. The 2012 and 2013 data represent the environmental impact on sediments after opening the Arase Dam gates and periodic seasonal flushing. The 2012 and 2013 data have not much difference and is caused due to periodic flushing of sediments. The periodic flushing cause transport of finer sediments towards the sea and thus decrease the elemental composition and thereafter retain the natural environmental conditions.

The UCC values (Taylor and McLennan 1985) are very low for As, Pb, Zn and Fe_2O_3 for almost all data except Pb for some of the 2012 and 2013 data (Fig. 3a–c). The 2002 data very clearly show a higher detrital trend for As, Pb and Zn than 2006. Therefore, before opening the dam gates these elemental concentrations have been higher. The 2012 and 2013 data spread is less than for 2002 and 2006. Therefore, a detrital trend line for the 2012 and 2013 sediments are not so significant and the values are lower. Thus, after opening the dam gates and also periodic flushing has caused the sediments to get back to the normal sediment trend or baseline trend (Fig. 3c, Ahmed et al. 2007). This indicates that opening the dam gates and flushing periodically causes a better environmental

Fig. 4 **a** The MnO and Fe₂O₃ plot, **b** total sulfur (TS) and Fe₂O₃ plot, showing redox conditions of the Yatsushiro tidal flat of 2012 data compared with unpublished data from Ishiga (2006), unpublished data from Young and Ishiga (2013)



condition for the sediments. The Uto Peninsula area sediments for 2012 show enrichments for As, Pb and Zn (Fig. 3a–c) and are related to finer sediments also as resulted in Yuan et al. (2010). Thus, the finer sediments are transported due to wave action and deposited in the Uto Peninsula area causing enrichments.

Changes in biogenic phases are represented using phosphate and TS. In Yatsushiro Bay sediments P and TS have decreased since 2002 (Fig. 3d, there is no TS data for 2006). Yatsushiro Bay consists of higher content of planktons in the suspended solids. Therefore, the P and TS increase in the Yatsushiro Bay in 2002 is likely due to the presence of a greater amount of plankton (discussed in “Suspended solids”, below) which has been lowered in 2012 and 2013.

MnO, TS and Fe are good indicators of oxidized and reduced conditions of sediments (Jung et al. 1996; Lacerda et al. 1999). The relationship between Mn vs. Fe and TS vs. Fe is well known, and is used for identifying bottom sediment environments and the scavenging of trace elements (Jung et al. 1996; Krupadam et al. 2006; Beck et al. 2008; Feely et al. 1983). Mn scavenging in the water column is coincident with enrichments of Cr, Ni, Cu, Zn and Pb in the suspended matter (Feely et al. 1983). In addition, Mn is scavenged by the newly formed hydrous Mn oxide coatings that exist on the surfaces of particles in estuarine and coastal waters (Feely et al. 1983). The MnO content of the southern area of the Uto Peninsula of 2006 and 2012 is higher than the other areas and plot on a higher trend line (Fig. 4a). The sediment in the southern area of the Uto Peninsula of the Yatsushiro tidal flat consists of fine sediment than other areas of the bay (Table 1). These fine particles are easily coated with hydrous Mn oxide and can scavenge trace elements as in Feely et al. (1983). Therefore, due to the finer grains, the trace element content of sediments in the southern area of the Uto Peninsula is higher than the Yatsushiro tidal flat and the western margin of the Yatsushiro Plain (Fig. 4a; Table 1). The anoxic water carrying Mn enters the tidal flat and leads to an

anoxic environment. Thus, due to high Mn and Fe in the southern Uto Peninsula tidal flats, this area is more anoxic compared to the other areas of the Yatsushiro tidal flat (Fig. 4). On the other hand, the bottom sediment redox environment of the Yatsushiro tidal flat has not undergone a large change during the period from 2006 to 2013 (Fig. 4a). The TS and Fe₂O₃ clearly show that the TS content was high in 2006 and drastically dropped in 2012/2013 and has no change in 2012–2013 (Fig. 4b). Thus, the organic matter and clay fraction show a decrease in 2012/2013. This also thus shows that the periodic flushing has brought in a favorable environmental condition for the bay sediments.

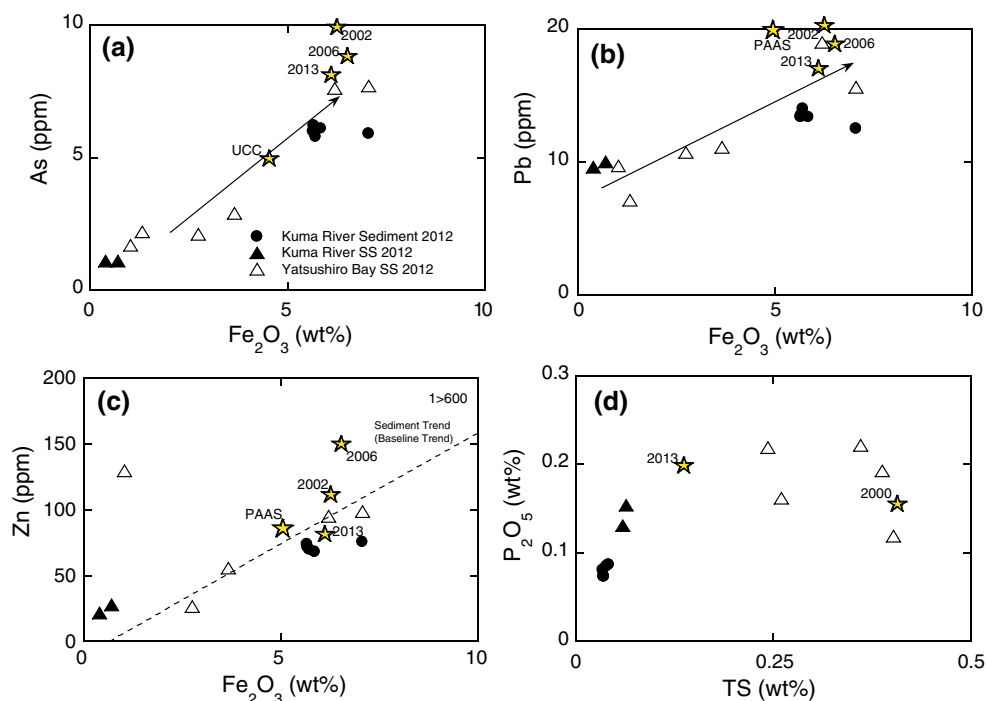
The LOI values for the Kuma River are low (Table 1, 2.32–2.99) indicating less carbonates and organic matter (OM) in 2012. The LOI of YA-1 is 18.10 and is very high. This sample contained many shell fragments and this high value is related to the shell carbonates. The southern area of the Uto Peninsula area has a comparatively high LOI (8.27–11.02). This is most likely related to high organic matter, since the CaO content is similar in almost all samples.

The pen shell, *Atrina pectinata*, belongs to the Pinnidae family and is a large (shell length up to 30 cm) suspension-feeding bivalve common along the coasts of Korea, Japan, and China. *A. pectinata* is an infaunal bivalve found in habitats ranging from muddy to sandy sediment and from tidal flats to shallow subtidal environments up to 20 m in depth (An et al. 2011). It is abundant in the tidal flats of the Yatsushiro Sea. It is possible, therefore, that *A. pectinata* shells could absorb trace elements. To test whether these shells are absorbing heavy metals, shells were collected from the bay and analyzed (Table 1). However, the low elemental values for the shells show that there is no heavy metal absorption taking place on the shell’s surface.

Water environment

ORP, DO, EC and pH were measured at the river mouth across tidal changes to see if there was any change of water

Fig. 5 Plots of **a** As, **b** Pb, **c** Zn versus Fe_2O_3 and **d** P_2O_5 versus TS for Kuma River sediments of 2012, suspended solids of Kuma River and Yatsushiro Bay in 2012. *Star* average data for 2002 is from Dozen and Ishiga (2002), unpublished data from Ishiga (2006), unpublished data from Young and Ishiga (2013)



quality. However, there was no major change in these parameters within the tidal cycle (Table 3). ORP ranged between -40 and 300 mV in the water of the Kuma River and the Yatsushiro Bay. The pH ranged between 7.40 and 7.95, remaining neutral. Redox buffering is used to interpret natural water and sediment environments. DO ranged from 6.7 to 8.3 mg/L and EC ranges between 0.312 and 6.7 mS/cm in the water and sediments of Kuma River and Yatsushiro Bay (Table 3), this indicates oxic and less polluted conditions. ORP and pH is related to the behavior of different elements and they contribute to the behavior of pollutants in the water environment (Chuan et al. 1996). ORP can control the solubility of many species of elements in a solution. In many cases, H^+ ions are also involved in these reactions; hence they are pH sensitive (Bachmann et al. 2001; Takeno 2005; Taylor et al. 2007). However, in Kuma River and Yatsushiro Bay these parameters remain relatively stable, indicating unpolluted water and sediment conditions.

Suspended solids

Suspended solids in rivers and estuaries have been studied using various methods such as, remote sensing (Wang et al. 2012; Chen et al. 2013), organic geochemistry (Hu et al. 2010; Wang et al. 2010; Carriquiry et al. 2011), magnetic approach as a tool for mining activities in rivers (Sandeep et al. 2011), adsorption of ammonia on suspended solids (Wang et al. 2010) and modeling approach (Shi et al.

2010). There are some recent studies of suspended solids carried out using trace metals showing the temporal effect (Liu et al. 2012; Shynu et al. 2012; Nair et al. 2013) and absorption desorption (Ma et al. 2011) on suspended solids. Thus, these studies are very helpful to understand the behavior of suspended solids in rivers and estuaries. However, suspended solids show very low elemental concentrations of As, Pb, Zn in the Kuma River compared to Yatsushiro Bay (Fig. 5a–c). The Kuma River sediments have comparatively higher values for As, Pb and Zn than the suspended solids (Fig. 5a–c). This indicates that the absorption and flocculation of heavy metals onto suspended solids (Shynu et al. 2012) has been reduced in the Kuma River and Yatsushiro Bay; this could be due to strong water circulation in these settings. The average sediment elemental values of 2002, 2006 and 2013 UCC and PAAS (Post Arcean Australian Shale) are higher than the suspended values (showed as stars). All the values for Zn falls on the sediment trend line (Ahmed et al. 2007). However, the P_2O_5 and TS is relatively higher in the Yatsushiro Bay suspended solids (Fig. 5d). This indicates higher nutrients in the Yatsushiro Bay suspended solids. As for a study by Zhang et al. (2008), metals adsorb more on suspended matter in higher pH water which would cause particulate phase transport of metal to become increasingly important in the seaward side of the estuary. The Kuma River and the Yatsushiro Bay pH is neutral (Table 3). Thus, though it is commonly accepted that suspended solids contain higher trace elements than sediments (Zhang et al. 2008) in the

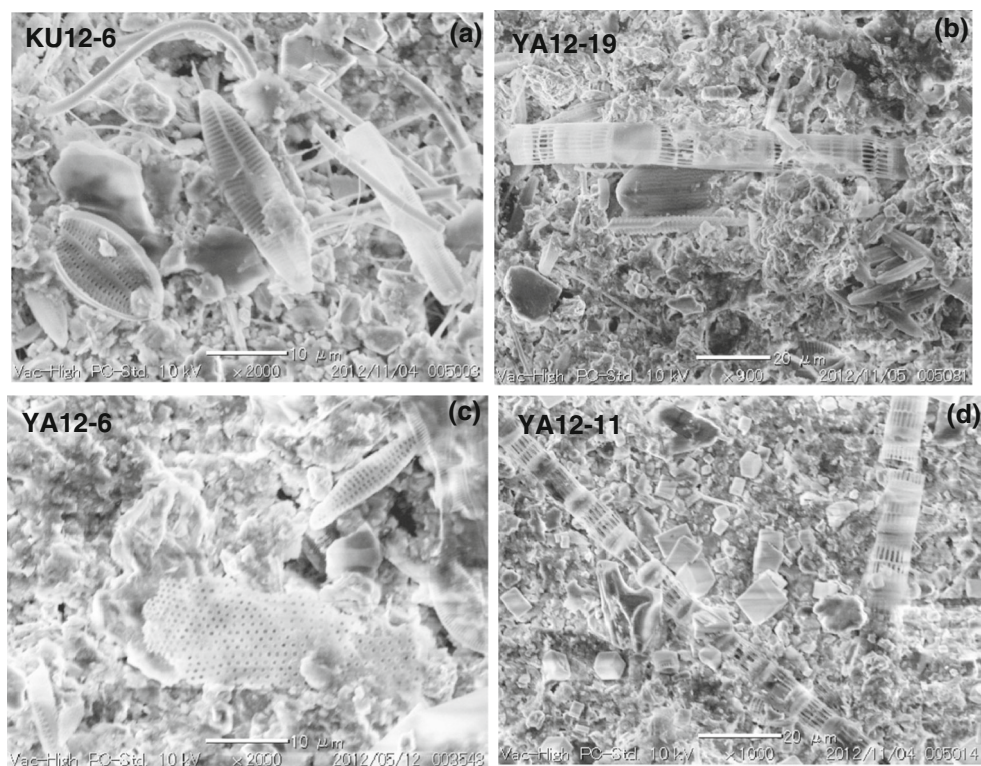


Fig. 6 Scanning electron microscope (SEM) images of diatoms **a–c** in suspended solids of the Kuma River and Yatsushiro Bay. **d** Salt crystals and diatoms

Kuma River and the Yatsushiro Bay suspended solids does not contain higher trace elements and may be due to low pH, higher wave action and large tidal changes causing rapid sediment transport.

Diatoms are distributed worldwide, respond quickly to environmental change, and reflect both physical and chemical characteristics of the overlying water column. A study by Xiangbin et al. (2013) shows the relationship between silicon, suspended matter and diatoms. The study shows the transportation and retention of silica which is essential for growth of diatoms affect the ecological environment (Xiangbin et al. 2013). Thus, diatoms are helpful in determining the environmental conditions particularly when studying suspended solids. The diatoms of the Kuma River and Yatsushiro Bay were identified and classified using SEM images. Many species of diatoms were found in the Kuma River and Yatsushiro Bay (Fig. 6a–d).

The most common species were: *Skeletonema* spp, *Neodelphineis pelagic*, *Thalassiosira* spp, *Nitzschia* spp, *Gyrosigma*, *Tryblionella* spp, *Delphineis* spp and *Achnanthes* spp (Fig. 6). These species have also been found by Park et al. (2012) in the Ariake Sea, just north of the Yatsushiro Sea (Fig. 1).

Skeletonema is the most abundant species in Yatsushiro Bay and the Kuma River sediments. *Skeletonema* grows

primarily in marine and brackish waters in estuaries (Fukuyo et al. 1990). Species within the genus are known to form large blooms in eutrophic and saline inland lakes and large rivers. Specimens of *Skeletonema* may be overlooked in samples due to their very light silicification (Spaulding and Edlund 2009). The habitat of *Neodelphineis pelagic* is marine (Krayesky et al. 2009) and this taxon was very common in Yatsushiro Bay samples.

Nitzschia is a very common genus with a large number of species that are often difficult to identify. Some *Nitzschia* species reach greatest abundance in waters high in organic pollution (Spaulding and Edlund 2009). *Gyrosigma* is characteristic of epipelagic and endopelagic habitats. This genus is widely distributed in fresh waters, with some species found in brackish waters (Spaulding and Edlund 2009). Although *Tryblionella* was described in 1853, many recent floras include species within the *Nitzschia* genus. Species within the genus *Tryblionella* occur in both freshwater and marine waters (Spaulding 2011) and are common on mud flats (Park et al. 2012). The abundance and taxa of diatoms in the Kuma River and Yatsushiro Bay indicate high nutrient conditions in the bay and river environments.

The SEM images also show the presence of salt crystals and high concentrations of clay particles. Salt crystals were

found in Yatsushiro Bay suspended solids, but not in those of the Kuma River (Fig. 6c). The most abundant salt crystals were found in the sample YA12-11. YA12-19 is from the river mouth and did not contain any salt crystals. This clearly explains the high chloride concentration in the marine bay samples (Table 2). The Kuma River (KU12-1) was low in clay content, but at the river mouth (KU12-6) clay content was high, indicating transport of fine particles into the bay. In the bay, the southern area of the Uto Peninsula samples (YA12-16, YA12-18 and YA12-19) showed high clay content. In summary, diatoms are associated with abundant organic matter and show that the bay marine environment is particularly rich in nutrients. Images of diatom species in Yatsushiro Bay and the Kuma River can be provided on request.

Conclusions

Sediments at the river mouth show less sorting than those of the Yatsushiro tidal flat, which is moderately sorted. Well-sorted fine-grain sediments are found in the northern portion of Yatsushiro Bay. Opening the Arase Dam has caused impounded Kuma River sediments to be flushed into Yatsushiro Bay. The river deposits coarser material proximal to the river mouth, while the finer particles are transported and deposited in the distal, northern parts of the Yatsushiro Bay because of the anticlockwise circulation of bay waters. Grain size has a major control over the chemistry of bay and river sediments. However, there are also sand bar formations found along the Kuma River resulting from the opening of the Arase Dam. The southern area of the Uto Peninsula tidal flat is made up of silty sand, while the southwest and northern parts of the Yatsushiro Plain and the river mouth is sandy tidal flat. The chemistry of Yatsushiro Bay sediments has changed tremendously between 2002 and 2013 with not much difference from 2012 to 2013. The trace element concentrations of As, Zn, Pb is higher in 2002 than 2012/13. Therefore, the 2002 sediments represent more of an urban trend while the 2012 and 2013 sediments represent a trend of more of normal sedimentation. SEM images of the suspended matter are rich in diatoms indicating high nutrient conditions in both the bay and river environments. Water chemistry parameters ORP, DO, EC and pH show that there is no pollution and the marine environment is oxic. Thus, the marine environment has been recovered after opening and periodic seasonal flushing of the Arase Dam water and sediments.

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