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# Assessment of dam removal from geochemical examination of Kuma River sediment, Kyushu, Japan

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**Abstract** The aim of this study was to determine if Arase dam gate removal and flushing elevated concentrations of any trace elements in Kuma River and Yatsushiro Bay sediments or caused riverine environmental change. The Arase dam gate on the Kuma River was opened in April 2010. Surface and bottom sediments were compared using 10-cm-long cores (2011) and two grain size fractions. Surface sediment data from 2002, 2012, and 2013 from the Kuma River and Yatsushiro Bay were also compared. The sediments were analyzed using XRF for 23 elements, and the grain size analysis was done. The short core surface and bottom sediments do not show major chemical changes, and therefore, may not represent post-and pre-dam sediments. Results based on 2011 samples show that the removal of the Arase dam gates in 2010 has been geoenvironmentally beneficial due to the decrease of environmentally related trace elements Pb and Zn in 2013. However, a slight increase in the levels of Cr, Cu, Zr, and Nb in 2013 indicates that periodic flushing in winter leads to elevation in these elements due to an increase in the fine fraction. Metal enrichment factors (EF) in 2002 are higher and these have decreased by 2013. Some elements exceed environmental guidelines, but this is due to natural background values, and there is no anthropogenic contamination. Thus, the environment of the river and bay has been significantly improved due to the dam opening. This result suggests that assessment and environmental monitoring studies are very important for dam management and future decision making.

**Keywords** Arase dam · Flushing · Environmental monitoring · Sediment · Kuma River · Yatsushiro Bay

## Introduction

The Arase dam was built in 1954 to generate hydroelectric power with a total storage capacity of 10,140,000 m$^3$ and an upstream flooded area of 1,230,000 m$^3$ (Planning and Construction Office of the Kumamotho Prefecture Office of the Government of Japan). The width of the dam is 210.8 m, and the height is 25 m. Following the plan of the Kumamoto Prefectural Office, the dam gate opening was begun in April 2010 (Fig. 1a). Systematic water level reduction was done by opening two control gates in the center of the dam (Fig. 1a). The decrease of the water level to the foot of the dam took 2 years, which was completed in March 2013. The removal of dam gates proceeded from the East side (right side bank) to the West side (left side bank) of the Arase dam. In 2015, two gates are to be completely removed (Fig. 1b), followed by the rest of the gates in 2016 (Fig. 1c), and by 2017, there was complete removal of the dam (Fig. 1d). A sediment removal plan was also established, which was to be completed in 2012. The bottom sediment removal was started in March 2007 at a capacity of

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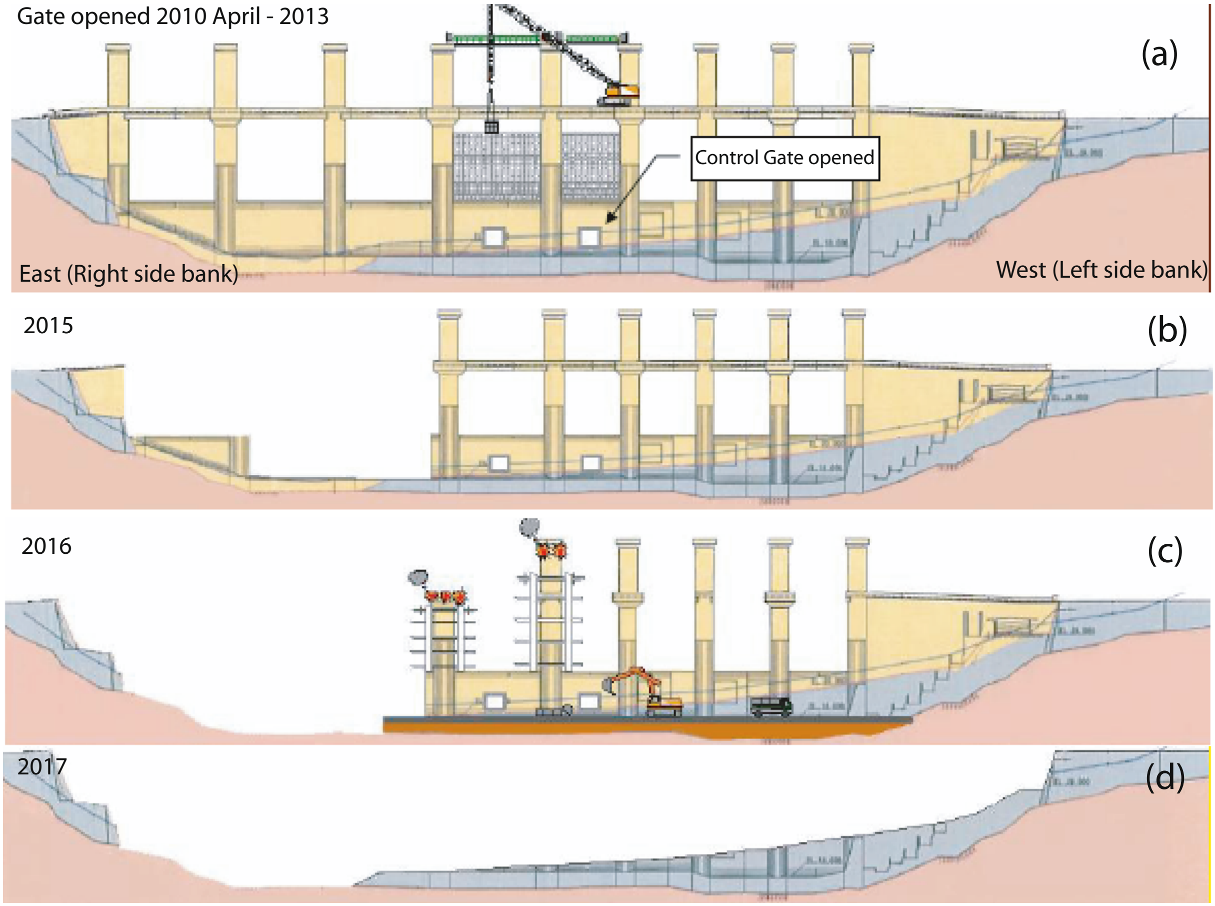

**Fig. 1** The schematic diagram showing the Arase dam removal starting from April 2010 to 2017. **a** The control gates were opened to have a systematic water level change by 2013. **b** The removal will be started in 2015 from the right corner of the dam. Initially, two gates will be completely removed and the concrete will be removed. **c** The next gates will be removed in 2016. **d** The completion of dam gate removal is expected to in 2017. These are only few figures of the whole process and the figure is sponsored by the Planning and Construction Office of the Kumamoto Prefecture Office of the government of Japan

96, 000 $m^3$. The removal is calculated to be finished by 2011. The removal of bottom sediment was estimated at 14, 776 $m^3$, 71, 469 $m^3$, 5,600 $m^3$, and 17, 000 $m^3$ for the years 2008, 2009, 2010, and 2011, respectively. The dam sediment is carried along the Kuma River to the Yatsushiro delta (Fig. 2). The Yatsushiro delta has an area of 2, 947 ha, and the Yatsushiro annual tidal change is 4 m.

Ms. Yoshiko Shiotani, Governor of Kumamoto Prefecture, decided to have the aging Arase dam removed after several public debates on environmental restoration of the Kuma River. In 2010 April 10th, all eight gates of the Arase dam were opened and the release of additional water accelerated the flow of the Kuma River. The increased flow of the river modified its bed forms. For example, the seven sand bars in the river bed known from before dam construction were restored through the transportation and accumulation of sands. New point bars were also developed along the river channel (Tsuru 2011). The changing ecology of the tidal flat in Yatsushiro Bay was described by Tsuru (2011). The pre-removal discoloration of "Aonori," green laver, vanished, and the populations of *Mya arenasia* shell, small crabs, *Upogebia pusilla*, green *Lingula jaspidea*, razor clam *Solen strictus*, Manila clam, and common orient clam, *Meretrix lusoria*, have been recovered. An emblematic feature of the recovery is the return of *Zostera marina* populations on sand bars in the Bay. All these are significant improvements in the habitat of Yatsushiro Bay. In winter, the dam gates are opened to lower water levels because water is not needed for agriculture and freezing water can damage the dam.

Thus, water levels fluctuated annually prior to the dam removal project, flushing the water above the dam every year in winter.

While the building of dams can be both environmentally and geochemically important, dam destruction will also have a large impact on the environment and on fluvial sediments (Katopodis and Aadland 2006; Hu et al. 2009; Bednarek 2001). When a river is dammed, sediments are deposited on the upstream side of the dam, and the dam's sustainability can be threatened by rapid sedimentation (Haregeweyn et al. 2012; Bednarek 2001). This can cause a number of environmental problems in the river channel, such as water stagnation and an increase of minor element concentrations in the river waters due to increasing anoxic conditions below the thermocline (Bellanger et al. 2004). These and other factors have been discussed in numerous dam sediment assessments (Jiongxin 1996; Ghrefat and Yusuf 2006; Cevik et al. 2009), and mitigation strategies based on sediment properties and sedimentation impacts (Haregeweyn et al. 2012; Trabelsi et al. 2012).

River sediments are ultimately deposited in estuaries. Estuarine geoenvironmental processes are very complex. The turbulent mixing of fresh water and seawater can generate rapid changes in many trace element concentrations (Feely et al. 1981). In addition to physical mixing of two very different water bodies, biological processes have a large impact on the aqueous environment, modifying many other variables. It is, therefore, difficult to describe the origins, pathways, and fates of dissolved and particulate materials in coastal marine systems, and especially, in estuaries. The Yatsushiro Bay environment is subject to this array of variables and is very complex.

Trace elements are a group of contaminants with high ecological significance. They tend to accumulate in suspended particulates and in sediments. Trace elements are not removed from the water column by self-purification. Trace elements tend to enter the food web via lower-level consumption and bio-accumulation, moving to the higher consumers (Ghrefat and Yusuf 2006; Khaled et al. 2006). Areas of mud deposits increase the amount of fine suspended matter in bottom waters. Anoxic conditions are also common, affecting bottom waters and sediments due to temperature changes and stratification, especially through seasonal cycles. This in turn affects river flow, water quality, and river sediments. In addition, grain size variation is a critically important factor in river sediment (Surian 2002; Owens et al. 2005). The rate of change in bed material size has important implications for river ecology (Petts et al. 2000). Thus, dam construction affects many geological features of the river, its water quality, and the channel's sedimentary environment. Consequently, dam removal will also have a major impact on river water quality, geology, and ecology.

This study is the first attempt to look into the environmental changes in the Kuma River and Arase dam sediments. Arase dam was removed based on a number of environmental issues. It is, therefore, important to look into the geo-environmental changes that have occurred after the removal of the dam. To do this, elemental concentrations of 23 major and trace elements (As, Pb, Zn, Cu, Ni, Cr, V, Sr, Y, Nb, Zr, Th, Sc, F, Br, I, Cl, $Fe_2O_3$, $TiO_2$, MnO, CaO, $P_2O_5$, and total sulfur) in the sediments of the Kuma River, Yatsushiro Bay, and the former lake above the Arase dam were examined. The objective was to monitor, document, and assess the environmental and geochemical changes that took place since 2002 in the Kuma River and Yatsushiro Bay because of the removal of Arase dam.

## Study area and its geology

The study area is located in Kumamoto prefecture, Kyushu, Japan. Arase dam is one of the few dams on the Kuma River and was the focus of the first dam removal project in Japan when the control gates were removed in April 2010. The Arase dam consists of eight gates. The dam is located ~19.9 km from the mouth of the 115-km-long Kuma River (Fig. 2). The Kuma River has a drainage area of 1,880 $km^2$, and it is considered to be one of the three most rapid rivers of Japan. The Youhaizeki bank is a bank which has a height less than 0.25 m close to the river mouth built to reduce the rate of flow and it does not have much effect on the sediments. The Kuma River falls to the Yatsushiro Bay a semi-closed estuary opening to the East China Sea. The study area is undergoes all four climatic conditions.

The geology of the study area is highly complex. The Kuma River flows through the Chichibu terrane, the Hisatu volcanics, the Cretaceous–Paleogene sedimentary rocks, the Shimanto terrane, and associated alluvial deposits (Fig. 2). Yatsushiro Bay is located in the Chichibu terrane (Fig. 2). The Yatsushiro area is within the inner and outer zones of the Kyushu Cretaceous system (Sakai et al. 1992). The sedimentary basins of

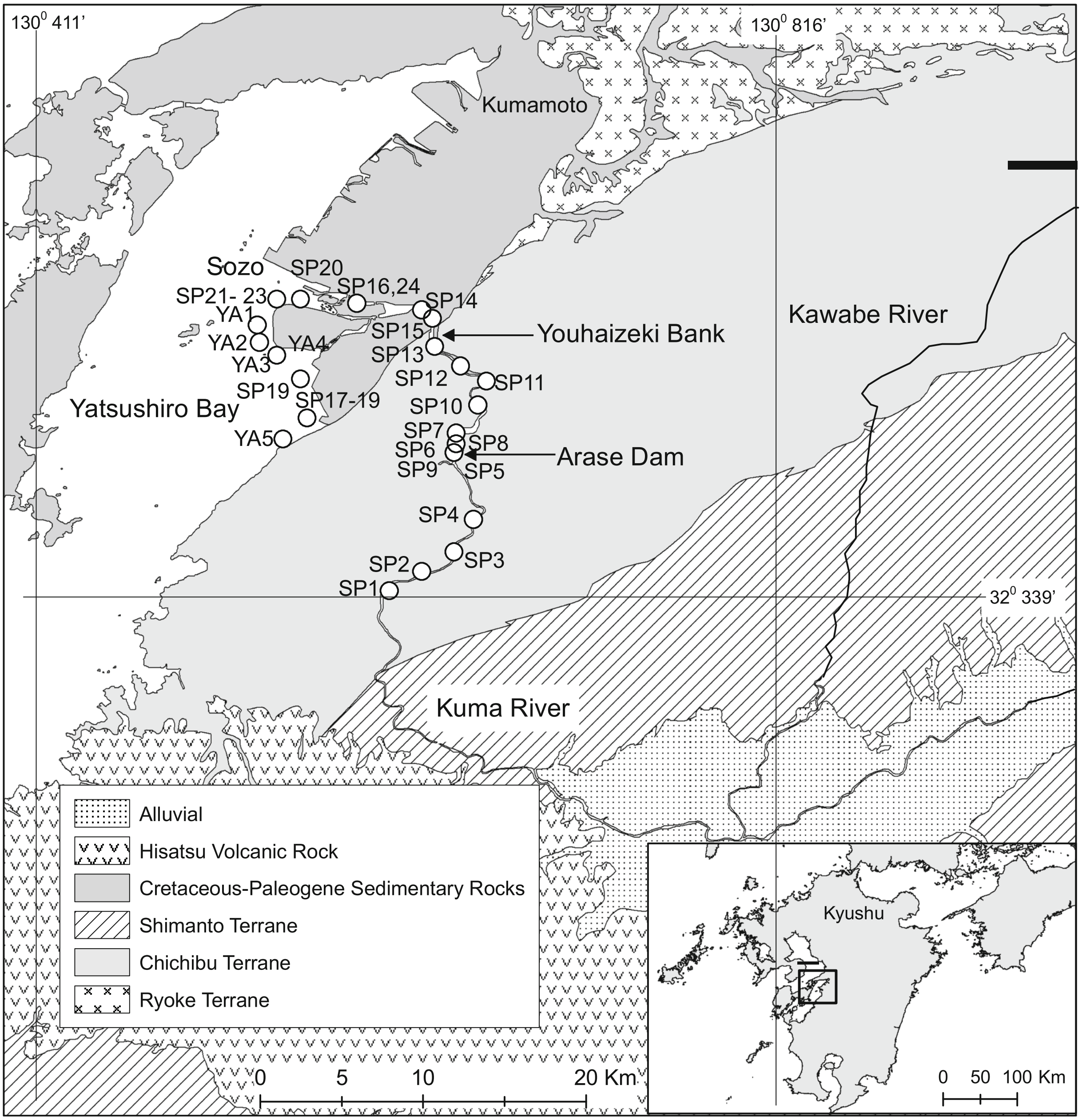

**Fig. 2** The geology of the Kuma River and the sampling locations in Kumamoto Prefecture, Kyushu Island, Japan

Kyushu belong to the Cretaceous and Tertiary periods. The Yatsushiro region and the Kuma River catchment intersect Early Cretaceous shallow-marine and turbidite basins and the Late Cretaceous non-marine, shallow-marine, and turbidite basins in the Ryoke terrane (Sakai et al. 1992). The Kawabe River joins the Kuma River after it flows through the Shimanto terrane and the Chichibu terrane (Fig. 2). The modern geologic setting may be different from that in the past, however, as Kyushu has active island arc type volcanoes, active faults, and significant crustal movement (Ogawa et al. 1992).

## Methodology

Stream sediment samples were collected along the Kuma River, Arase dam, and in the river mouth at Yatsushiro Bay using the hand pit method at 1–6 cm depths in November 2011 and May 2013. Sampling locations were selected based on sediment deposition, flow rate, river gradient, erosion, and human impact on natural sedimentation. Samples were taken at varying distances based on accessibility (Fig. 2), the locations of tributaries, geological factors, upstream and downstream of the Arase dam, and the dam site. Twenty river sediment samples and five Bay samples were collected in 2011. In 2013, 12 river sediments and eleven Bay representative samples were collected. Both sampling was done during low tide in the bay area. Samples were collected in plastic bags using a plastic spade and were stored at 4 °C in a cooling box and transferred to Shimane University and Tokyo University of Science. Sample locations (Fig. 2) matched locations of a previous study (Dozen and Ishiga 2002). Short core samples (8–10 cm in length) were collected using plastic short push cores and divided into “surface“and “bottom” samples based on color and grain size, thought to represent sediment from before and after dam removal (Fig. 3). The surface and bottom boundaries were tentatively determined by a sedimentological and geological specialist in the field. Photographs were taken of each sampling location and each sample (the photographs can be provided on request). The surface samples were composed of finer grains of darker colour while the bottom sediments were of large grains with lighter colour. The surface–bottom boundary was also checked using geochemical compositions determined by XRF and LOI values. A marked difference in chemical composition and LOI values between the surface and the bottom samples will give the boundary. The samples were collected in separate plastic bags using the grab sampling method for surface and bottom sediments. Twenty-three surface samples and 19 bottom samples were recovered. During the 2013 sample collection program, many mud drapes were seen along the river and these were also sampled. All samples were oven dried at 110 °C after transport and halved using the cone and quarter method. The fine (0.075–0.25 mm) and medium (0.25–0.85 mm) size fractions ($n=14$) were sieve separated. Grain size analysis for six size fractions was carried out at Tokyo University of Science for the surface and bottom sediments using a SALD 3000 grain size analyzer. Oven-dried sediments were treated with 30 % $H_2O_2$ for at least 24 h prior to measurement. Calculations of grain size and sorting were made following Folk and Ward (1957).

Approximately 50 g of each sample was then oven dried at 160 °C for 48 h before crushing to fine powder in an automatic agate mortar and pestle grinder. The crushed sediment samples were compressed into pellets, using a force of 200KN for 60 s. The concentrations of 23 major and trace elements (As, Pb, Zn, Cu, Ni, Cr, V, Sr, Y, Nb, Zr, Th, Sc, TS, F, Br, I, Cl, Ti, Fe, Mn, Ca, and P) were then determined by X-ray florescence spectrometry using a Rigaku RIX-2000 spectrometer equipped with an Rh-anode tube. Analytical methods, instrumental conditions, and calibration followed those described by Ogasawara (1987). The average errors for all elements are less than ±10 % relative. LOI was performed on the surface and bottom sediments. Portions (7–10 g) of the crushed materials were transferred into glass vials. The sediment samples were oven dried at 110 °C for 24 h. Gravimetric LOI (Loss on ignition) determinations were calculated from the net weight loss after ignition in a muffle furnace at 1,020 °C for at least 2 h. All standard methods have been followed and the average errors for all elements are less than ±10 % relative.

**Fig. 3** Grain size variation of the **a** fine (0.25–0.005 mm) and **b** medium (0.25–2 mm) fractions of surface and bottom sediments shown by images of location SP2 and SP11

## Results

The Kuma River sediment samples in 2011 and 2013 were all black in color. The Yatsushiro Bay sediment samples in 2011 and 2013 were more gray than black. Almost all the surface and bottom elemental concentrations in 2011 are within the ranges for UCC (Upper continental crust, Taylor and McLennan 1985) except for Zn, Fe, and Ti at a few locations (Table 1).

In the 2011 samples, chlorine was detected only in the river locations below SP19 and in Yatsushiro Bay (Fig. 2). In these surface and bottom samples, chlorine ranges are 2,449–12,887 ppm and 3,631–12,061 ppm, respectively, which are much higher than UCC 370 ppm (Taylor and McLennan 1985). The TS (total sulfur) content and LOI of the surface and bottom sediments show a considerable difference (Table 1). The bottom sediments have slightly higher TS and LOI indicating higher organic matter and reduced conditions. There is no clear chemical difference between the surface and bottom sediments (Fig. 4a, b, c, d). The average values of surface and bottom sediments for all elements except TS are very similar (Table 1). The minimum and maximum values of sediments from Arase dam, Yatsushiro Bay, and the Kuma River are given in Table 2.

The fine and medium size fractions differ from UCC for Ni, Cr, and V. They are 1.5 times to just above two times higher than UCC while the other elements are comparable to UCC. LOI is low except for a few samples (Table 1). In the 2013 samples, except Cr, V, and Fe, all others are well within UCC (Table 3). The geologic terranes in the catchment are highly evolved and thus all elements are very close to UCC.

The median grain size (Md ϕ) of the surface sediments ranges between −0.57 and 3.27 ϕ, while the bottom sediments are between −0.93 and 2.93 ϕ (Table 1). It is very clear from the grain size of surface and bottom sediments that the sediments of the Kuma River and the Yatsushiro Bay mainly consist of very coarse sand to very fine sand. Table 2 gives the chemical data of the fine and medium fractions. The summarized bulk chemical data of Dozen and Ishiga 2002 of Arase dam, Yatsushiro Bay, and Kuma River is also provided (Table 2). The elements Sc, Cl, F, and MnO were not analyzed in the 2002 data. For almost all elements, the values of Dozen and Ishiga 2002 are much higher than the 2011 and 2013 data (Tables 1 and 3). Grain size analysis of the 2011 samples shows that the clay and silt fractions have lower elemental concentrations (Online resource 1).

In the dam areas (Arase and Youhaizeki bank), bottom sediment median grain sizes (Φ) are low ( −0.93, −0.83) but are high in the dam surface sediments (2.63, 2.53, Table 1). This is due to the very low fine fraction in the dam sediments and comparatively higher coarse sand grain percentages at the dams. The median grain size of the river sediments above the Arase dam (upstream) is low in both surface and bottom sediments. The median grain size of surface sediments is also higher than that of the bottom sediments. Along the river after the dams (downstream) the median grain size is higher than upstream. The sorting of the grains (Table 1) along the river and in the bay is moderately to poorly sorted (using the Folk and Ward 1957 classification). The sorting of bottom sediments in the river is low (avg, 1.0), but it is high in the surface sediments (avg, 1.1). Thus, the dams, river, and bay sediments mainly consist of coarse grains in 2011.

Fine grain (0.05–0.25 mm) sediment content is significantly higher in the bottom sediments (Fig. 4a) in all cores. In the bottom sediment samples, fine grain sediments are low in abundance (<20 %) above Arase dam and are very high (50–90 %) below the dam (Fig. 4a). The fine fractions in surface sediments are also low above Arase dam. However, there is an increasing trend in the fines (0.05–0.25 mm) of the surface sediments below the dam towards Yatsushiro Bay (Fig. 4a). The coarser grain-size sediments (0.25–2 mm) are high in percentage in both bottom and surface sediments above the Arase dam (Fig. 4b). These coarser sediments drop in abundance below the dam and gradually decrease along the stream towards Yatsushiro Bay in both surface and bottom sediments. At the dams themselves, sediments are very low in fines (0.05–0.25 mm) in both the bottom and surface sediments. The dams have a much higher coarse sediment proportion in the bottom sediment compared to the surface sediments.

## Discussion

### Surface and bottom sediment chemical variation and grain size

Fine-grained sediment is a natural and essential component of river systems and plays a major role in the hydrological, geomorphological, and ecological

**Table 1** Elemental concentrations, loss on ignition (LOI), Md50 (median grain size), sorting, and EF of surface and bottom sediments (2011) Kuma River (KR) and Yatsushiro Bay (YB)

| Sample no. | Trace elements (ppm) | | | | | | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | As | Pb | Zn | Cu | Ni | Cr | V | Sr | Y | Nb | Zr | Th | Sc | TS | F | Br |
| Surface | | | | | | | | | | | | | | | | |
| SP 2 S | 7 | 16 | 73 | 18 | 25 | 51 | 105 | 107 | 21 | 7 | 98 | 7 | 10 | 273 | 145 | 2 |
| SP 3 S | 9 | 18 | 83 | 27 | 32 | 56 | 115 | 63 | 26 | 10 | 97 | 10 | 11 | 276 | 33 | 2 |
| SP 4 S | 6 | 14 | 78 | 23 | 39 | 88 | 139 | 129 | 20 | 9 | 97 | 7 | 16 | 275 | 227 | 2 |
| SP 5 S | 6 | 14 | 65 | 19 | 29 | 52 | 96 | 113 | 19 | 6 | 92 | 7 | 10 | 265 | nd | 1 |
| SP 6 S | 7 | 15 | 78 | 21 | 29 | 77 | 137 | 125 | 20 | 8 | 99 | 6 | 15 | 273 | 75 | 1 |
| SP 7 S | 6 | 13 | 73 | 23 | 38 | 87 | 136 | 120 | 21 | 9 | 97 | 7 | 16 | 264 | 47 | 2 |
| SP 8 S | 7 | 13 | 71 | 23 | 34 | 73 | 115 | 119 | 19 | 7 | 101 | 7 | 14 | 283 | 75 | 2 |
| SP 9 S | 9 | 18 | 105 | 29 | 117 | 141 | 170 | 96 | 23 | 8 | 108 | 7 | 16 | 342 | 8 | 2 |
| SP 10 S | 7 | 15 | 74 | 23 | 35 | 73 | 129 | 117 | 21 | 9 | 107 | 7 | 12 | 287 | 116 | 2 |
| SP 11 S | 7 | 15 | 73 | 24 | 36 | 73 | 133 | 121 | 22 | 9 | 103 | 7 | 14 | 290 | 89 | 2 |
| SP 12 S | 6 | 15 | 76 | 21 | 40 | 106 | 138 | 120 | 20 | 8 | 95 | 7 | 14 | 294 | 61 | 2 |
| SP 14 S | 8 | 16 | 79 | 30 | 36 | 78 | 142 | 132 | 22 | 9 | 124 | 9 | 15 | 371 | 302 | 3 |
| SP 15 S | 6 | 14 | 66 | 20 | 42 | 71 | 93 | 116 | 18 | 6 | 89 | 6 | 9 | 262 | 74 | 1 |
| SP 16 S | 8 | 18 | 86 | 26 | 38 | 89 | 138 | 125 | 25 | 10 | 137 | 7 | 15 | 998 | 103 | 15 |
| SP 17 S | 7 | 14 | 70 | 17 | 34 | 82 | 106 | 129 | 21 | 8 | 122 | 7 | 10 | 1,486 | 144 | 13 |
| SP 18 S | 7 | 15 | 73 | 17 | 35 | 79 | 109 | 133 | 22 | 8 | 153 | 8 | 12 | 2,565 | nd | 16 |
| SP 19 S | 9 | 17 | 83 | 22 | 36 | 72 | 114 | 132 | 24 | 10 | 143 | 8 | 13 | 2,332 | 61 | 22 |
| SP 20 S | 8 | 16 | 90 | 18 | 36 | 76 | 113 | 124 | 22 | 8 | 123 | 8 | 14 | 1,391 | 89 | 10 |
| SP 21 S | 10 | 20 | 90 | 25 | 38 | 66 | 131 | 134 | 27 | 11 | 134 | 9 | 14 | 2,755 | 103 | 36 |
| SP 24 S | 8 | 13 | 78 | 13 | 34 | 85 | 111 | 121 | 20 | 7 | 110 | 7 | 13 | 659 | 74 | 5 |
| Avg. KR | 7 | 15 | 78 | 22 | 39 | 79 | 124 | 119 | 22 | 8 | 111 | 7 | 13 | 797 | 101 | 7 |
| YA 1 S | 9 | 16 | 80 | 18 | 35 | 65 | 113 | 125 | 22 | 8 | 115 | 8 | 12 | 2,277 | 61 | 19 |
| YA 2 S | 11 | 20 | 98 | 36 | 42 | 70 | 132 | 131 | 27 | 11 | 130 | 11 | 15 | 2,279 | 89 | 36 |
| YA 3 S | 7 | 13 | 66 | 21 | 36 | 69 | 99 | 127 | 19 | 7 | 98 | 7 | 10 | 1,425 | 180 | 14 |
| YA 4 S | 12 | 20 | 103 | 34 | 45 | 72 | 148 | 122 | 29 | 11 | 129 | 12 | 16 | 2,088 | 47 | 41 |
| YA 5 S | 12 | 23 | 120 | 27 | 43 | 69 | 129 | 133 | 27 | 10 | 122 | 11 | 14 | 4,199 | 192 | 41 |
| Avg. YB | 8 | 16 | 81 | 23 | 39 | 77 | 124 | 120 | 22 | 9 | 113 | 8 | 13 | 1,116 | 104 | 11 |
| Bottom | | | | | | | | | | | | | | | | |
| SP 2 B | 7 | 14 | 68 | 20 | 31 | 59 | 104 | 111 | 20 | 8 | 95 | 7 | 11 | 274 | 47 | 2 |
| SP 3 B | 10 | 18 | 83 | 26 | 35 | 59 | 117 | 62 | 26 | 11 | 102 | 11 | 11 | 290 | | 2 |
| SP 4 B | 6 | 14 | 70 | 21 | 34 | 66 | 114 | 121 | 20 | 8 | 96 | 7 | 12 | 280 | 261 | 2 |
| SP 5 B | 7 | 13 | 68 | 23 | 28 | 52 | 108 | 113 | 19 | 6 | 92 | 7 | 10 | 273 | 89 | 1 |
| SP 6 B | 7 | 14 | 78 | 23 | 32 | 76 | 143 | 120 | 21 | 8 | 101 | 7 | 14 | 300 | 89 | 2 |
| SP 7 B | 7 | 16 | 77 | 25 | 34 | 63 | 139 | 129 | 22 | 9 | 113 | 8 | 15 | 430 | | 2 |
| SP 8 B | 6 | 13 | 67 | 21 | 36 | 81 | 114 | 117 | 19 | 7 | 91 | 7 | 12 | 289 | 62 | 1 |
| SP 9 B | 8 | 16 | 99 | 30 | 121 | 164 | 164 | 91 | 21 | 6 | 96 | 6 | 17 | 281 | 176 | 2 |
| SP 10 B | 7 | 16 | 76 | 23 | 37 | 73 | 126 | 115 | 21 | 8 | 100 | 7 | 15 | 319 | 170 | 2 |

**Table 1** (continued)

| Sample no. | Trace elements (ppm) | | | | | | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | As | Pb | Zn | Cu | Ni | Cr | V | Sr | Y | Nb | Zr | Th | Sc | TS | F | Br |
| SP 11 B | 7 | 13 | 66 | 19 | 45 | 94 | 104 | 120 | 19 | 6 | 92 | 5 | 11 | 280 | 61 | 1 |
| SP 14 B | 7 | 17 | 76 | 23 | 35 | 90 | 138 | 129 | 22 | 9 | 114 | 8 | 15 | 358 | 49 | 2 |
| SP 15 B | 6 | 12 | 67 | 21 | 46 | 86 | 99 | 114 | 18 | 6 | 90 | 6 | 9 | 270 | 1 | 18 |
| SP 16 B | 8 | 19 | 89 | 28 | 39 | 82 | 138 | 131 | 26 | 11 | 159 | 9 | 13 | 1,122 | 286 | 14 |
| SP 20 B | 8 | 16 | 83 | 18 | 36 | 76 | 121 | 119 | 21 | 8 | 124 | 7 | 13 | 1,933 | 89 | 16 |
| SP21 B | 9 | 19 | 90 | 25 | 34 | 72 | 113 | 134 | 25 | 9 | 141 | 9 | 15 | 4,068 | 157 | 15 |
| SP24 B | 7 | 13 | 76 | 13 | 36 | 81 | 100 | 117 | 19 | 7 | 104 | 7 | 10 | 731 | 75 | 4 |
| Avg. KR | 7 | 15 | 77 | 22 | 41 | 80 | 121 | 115 | 21 | 8 | 107 | 7 | 13 | 719 | 115 | 5 |
| YA 1 B | 11 | 18 | 93 | 25 | 39 | 69 | 131 | 126 | 26 | 10 | 125 | 9 | 15 | 2,491 | 178 | 22 |
| YA 2 B | 10 | 19 | 90 | 28 | 45 | 80 | 135 | 126 | 27 | 10 | 126 | 9 | 16 | 2,109 | 386 | 26 |
| YA 3 B | 6 | 12 | 63 | 13 | 35 | 68 | 87 | 121 | 18 | 7 | 93 | 6 | 9 | 1,247 | 38 | 13 |
| YA 4 B | 7 | 16 | 80 | 24 | 42 | 125 | 128 | 124 | 21 | 8 | 100 | 7 | 14 | 2,070 | 23 | 21 |
| YA 5 B | 12 | 21 | 116 | 26 | 42 | 70 | 130 | 139 | 26 | 10 | 120 | 9 | 15 | 4,786 | 89 | 60 |
| Avg. YB | 9 | 17 | 88 | 23 | 41 | 83 | 122 | 127 | 24 | 9 | 113 | 8 | 14 | 2,541 | 143 | 28 |
| UCC | 4.8 | 20 | 71 | 25 | 20 | 35 | 60 | 350 | 22 | 25 | 190 | 10.7 | 11 | | 557 | 1.6 |

| Sample no. | Major elements (wt%) | | | | | | EF values | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | I | Cl | $TiO_2$ | $Fe_2O_3$ | MnO | CaO | $P_2O_5$ | LOI | d10 Φ | Sorting Φ | As EF | Pb EF | Zn EF | Cu EF | Ni EF | Cr EF |
| Surface | | | | | | | | | | | | | | | | |
| SP 2 S | 9 | nd | 0.46 | 5.74 | 0.10 | 1.08 | 0.09 | 2.83 | −0.57 | 0.61 | 1.2 | 0.6 | 0.8 | 0.6 | 1.0 | 1.1 |
| SP 3 S | 10 | nd | 0.57 | 6.07 | 0.15 | 0.84 | 0.10 | 3.30 | 1.13 | 0.86 | 1.4 | 0.7 | 0.9 | 0.8 | 1.2 | 1.2 |
| SP 4 S | 6 | nd | 0.65 | 7.11 | 0.12 | 1.58 | 0.11 | 2.62 | 1.83 | 1.13 | 0.8 | 0.5 | 0.7 | 0.6 | 1.2 | 1.6 |
| SP 5 S | 15 | nd | 0.44 | 5.41 | 0.09 | 1.21 | 0.09 | 18.18 | 1.07 | 0.72 | 1.0 | 0.6 | 0.8 | 0.6 | 1.2 | 1.2 |
| SP 6 S | 13 | nd | 0.63 | 6.38 | 0.10 | 1.29 | 0.10 | 2.52 | 1.57 | 1.04 | 1.0 | 0.5 | 0.8 | 0.6 | 1.0 | 1.6 |
| SP 7 S | 12 | nd | 0.67 | 6.61 | 0.11 | 1.42 | 0.11 | na | 2.30 | 1.05 | 0.9 | 0.4 | 0.7 | 0.6 | 1.3 | 1.7 |
| SP 8 S | 14 | nd | 0.52 | 6.21 | 0.10 | 1.33 | 0.10 | 2.57 | 2.63 | 1.27 | 1.0 | 0.5 | 0.7 | 0.7 | 1.2 | 1.5 |
| SP 9 S | 11 | nd | 0.73 | 7.79 | 0.13 | 1.18 | 0.11 | 4.57 | 0.50 | 1.02 | 1.0 | 0.5 | 0.9 | 0.7 | 3.4 | 2.3 |
| SP 10 S | 12 | nd | 0.59 | 6.19 | 0.10 | 1.30 | 0.10 | 2.86 | 1.90 | 1.12 | 1.0 | 0.5 | 0.8 | 0.7 | 1.3 | 1.5 |
| SP 11 S | 13 | nd | 0.59 | 6.13 | 0.11 | 1.41 | 0.10 | 2.99 | 2.53 | 1.03 | 1.1 | 0.5 | 0.8 | 0.7 | 1.3 | 1.5 |
| SP 12 S | 14 | nd | 0.63 | 6.50 | 0.11 | 1.24 | 0.11 | na | 2.07 | 1.12 | 0.8 | 0.5 | 0.7 | 0.6 | 1.4 | 2.1 |
| SP 14 S | 8 | nd | 0.64 | 6.25 | 0.11 | 1.48 | 0.11 | na | 2.83 | 1.05 | 1.2 | 0.6 | 0.8 | 0.9 | 1.3 | 1.6 |
| SP 15 S | 16 | nd | 0.43 | 5.45 | 0.09 | 1.23 | 0.09 | 2.35 | 1.10 | 0.66 | 1.0 | 0.6 | 0.8 | 0.7 | 1.7 | 1.7 |

**Table 1** (continued)

| Sample no. | Major elements (wt%) | | | | | | EF values | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | I | Cl | $TiO_2$ | $Fe_2O_3$ | MnO | CaO | $P_2O_5$ | LOI | d10 Φ | Sorting Φ | As EF | Pb EF | Zn EF | Cu EF | Ni EF | Cr EF |
| SP 16 S | 19 | 2,939 | 0.62 | 6.22 | 0.10 | 1.13 | 0.15 | 4.86 | 2.63 | 1.58 | 1.2 | 0.6 | 0.9 | 0.7 | 1.4 | 1.8 |
| SP 17 S | 18 | 5,510 | 0.55 | 5.41 | 0.10 | 1.17 | 0.10 | na | 2.80 | 1.42 | 1.3 | 0.6 | 0.8 | 0.6 | 1.4 | 1.9 |
| SP 18 S | 15 | 7,098 | 0.59 | 5.27 | 0.08 | 1.20 | 0.10 | na | 2.97 | 1.38 | 1.3 | 0.7 | 0.9 | 0.6 | 1.5 | 1.9 |
| SP 19 S | 14 | 4,584 | 0.65 | 5.73 | 0.11 | 1.15 | 0.12 | na | 3.27 | 2.19 | 1.5 | 0.7 | 0.9 | 0.7 | 1.4 | 1.6 |
| SP 20 S | 16 | 2,449 | 0.59 | 5.92 | 0.08 | 1.09 | 0.11 | 3.76 | 2.60 | 1.42 | 1.3 | 0.6 | 1.0 | 0.5 | 1.4 | 1.7 |
| SP 21 S | 12 | 9,712 | 0.64 | 6.16 | 0.10 | 1.30 | 0.15 | 8.90 | 2.40 | 2.09 | 1.5 | 0.7 | 0.9 | 0.7 | 1.4 | 1.4 |
| SP 24 S | 18 | nd | 0.51 | 5.56 | 0.09 | 1.13 | 0.09 | 2.66 | 2.13 | 1.18 | 1.3 | 0.5 | 0.9 | 0.4 | 1.4 | 2.0 |
| Avg. KR | 13 | 5,382 | 0.59 | 6.11 | 0.10 | 1.24 | 0.11 | 4.64 | 1.98 | 1.20 | 1.1 | 0.6 | 0.8 | 0.6 | 1.4 | 1.7 |
| YA 1 S | 10 | 7,725 | 0.57 | 5.61 | 0.08 | 1.22 | 0.12 | 4.86 | 2.37 | 1.58 | 1.5 | 0.6 | 0.9 | 0.6 | 1.4 | 1.5 |
| YA 2 S | 6 | 12,887 | 0.67 | 6.48 | 0.13 | 1.17 | 0.17 | 8.51 | 2.90 | 1.67 | 1.5 | 0.7 | 1.0 | 1.0 | 1.5 | 1.4 |
| YA 3 S | 16 | 7,728 | 0.49 | 5.26 | 0.10 | 1.28 | 0.10 | 3.41 | 1.40 | 1.09 | 1.2 | 0.6 | 0.8 | 0.7 | 1.5 | 1.7 |
| YA 4 S | | 9,928 | 0.70 | 6.98 | 0.15 | 1.17 | 0.18 | 10.19 | 3.17 | 2.52 | 1.6 | 0.7 | 0.9 | 0.9 | 1.5 | 1.3 |
| YA 5 S | 8 | 9,036 | 0.66 | 6.90 | 0.10 | 1.60 | 0.15 | 8.15 | 2.90 | 0.75 | 1.6 | 0.7 | 1.1 | 0.7 | 1.4 | 1.3 |
| Avg. YB | 13 | 7,082 | 0.59 | 6.13 | 0.10 | 1.25 | 0.11 | 5.24 | 2.09 | 1.26 | 1.2 | 0.6 | 0.8 | 0.7 | 1.4 | 1.6 |
| Bottom | | | | | | | | | | | | | | | | |
| SP 2 B | 15 | nd | 0.48 | 5.57 | 0.09 | 1.22 | 0.09 | 2.76 | 1.50 | 1.83 | 1.1 | 0.6 | 0.8 | 0.7 | 1.2 | 1.4 |
| SP 3 B | 16 | nd | 0.56 | 6.17 | 0.13 | 0.83 | 0.10 | 3.31 | −0.27 | 0.87 | 1.5 | 0.7 | 0.9 | 0.7 | 1.3 | 1.2 |
| SP 4 B | 16 | nd | 0.54 | 6.03 | 0.10 | 1.41 | 0.10 | 2.66 | 0.00 | 1.00 | 0.9 | 0.5 | 0.7 | 0.6 | 1.3 | 1.4 |
| SP 5 B | 20 | nd | 0.46 | 5.57 | 0.10 | 1.18 | 0.09 | 2.30 | 0.93 | 0.86 | 1.2 | 0.5 | 0.8 | 0.7 | 1.1 | 1.2 |
| SP 6 B | 11 | nd | 0.60 | 6.39 | 0.10 | 1.27 | 0.10 | 2.77 | 2.00 | 1.15 | 1.0 | 0.5 | 0.8 | 0.6 | 1.1 | 1.5 |
| SP 7 B | 7 | nd | 0.62 | 6.21 | 0.11 | 1.46 | 0.10 | na | 2.70 | 1.05 | 1.0 | 0.6 | 0.8 | 0.7 | 1.2 | 1.3 |
| SP 8 B | 16 | nd | 0.50 | 6.17 | 0.10 | 1.31 | 0.10 | 2.48 | −0.93 | 0.61 | 1.0 | 0.5 | 0.7 | 0.6 | 1.3 | 1.7 |
| SP 9 B | 9 | nd | 0.63 | 8.02 | 0.11 | 1.19 | 0.11 | 4.04 | 0.00 | 0.56 | 0.9 | 0.5 | 0.8 | 0.7 | 3.4 | 2.6 |
| SP 10 B | 11 | nd | 0.55 | 6.13 | 0.10 | 1.26 | 0.10 | 3.18 | 2.27 | 1.12 | 1.0 | 0.6 | 0.8 | 0.7 | 1.4 | 1.5 |
| SP 11 B | 13 | nd | 0.43 | 5.68 | 0.09 | 1.22 | 0.10 | 2.52 | −0.83 | 0.77 | 1.2 | 0.5 | 0.7 | 0.6 | 1.8 | 2.1 |
| SP 14 B | 9 | nd | 0.65 | 6.18 | 0.11 | 1.38 | 0.11 | na | 2.83 | 1.06 | 1.0 | 0.6 | 0.8 | 0.7 | 1.3 | 1.9 |
| SP 15 B | nd | nd | 0.45 | 5.57 | 0.09 | 1.26 | 0.09 | 2.38 | 1.13 | 0.60 | 1.1 | 0.5 | 0.8 | 0.7 | 1.9 | 2.0 |
| SP 16 B | 16 | 3,631 | 0.67 | 6.29 | 0.10 | 1.16 | 0.15 | 5.75 | 2.93 | 1.54 | 1.2 | 0.7 | 0.9 | 0.8 | 1.4 | 1.7 |
| SP 20 B | 18 | 8,856 | 0.57 | 5.83 | 0.07 | 1.12 | 0.11 | 3.86 | 2.27 | 1.39 | 1.2 | 0.6 | 0.9 | 0.5 | 1.4 | 1.7 |
| SP21 B | 20 | 3,967 | 0.62 | 5.68 | 0.08 | 1.25 | 0.13 | 5.69 | 1.80 | 1.12 | 1.4 | 0.7 | 1.0 | 0.8 | 1.4 | 1.6 |

**Table 1** (continued)

| Sample no. | Major elements (wt%) | | | | | | EF values | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | I | Cl | $TiO_2$ | $Fe_2O_3$ | MnO | CaO | $P_2O_5$ | LOI | d10 Φ | Sorting Φ | As EF | Pb EF | Zn EF | Cu EF | Ni EF | Cr EF |
| SP24 B | 21 | nd | 0.50 | 5.55 | 0.08 | 1.10 | 0.09 | 2.76 | na | na | 1.2 | 0.5 | 0.9 | 0.4 | 1.5 | 1.9 |
| Avg. KR | 15 | 5,485 | 0.55 | 6.07 | 0.10 | 1.23 | 0.10 | 3.32 | 1.22 | 1.04 | 1.1 | 0.6 | 0.8 | 0.7 | 1.5 | 1.7 |
| YA 1 B | 14 | 6,017 | 0.65 | 6.40 | 0.09 | 1.32 | 0.14 | 6.31 | 1.90 | 1.37 | 1.5 | 0.6 | 0.9 | 0.7 | 1.4 | 1.4 |
| YA 2 B | 13 | 6,613 | 0.67 | 6.40 | 0.12 | 1.18 | 0.16 | 7.51 | 2.87 | 2.11 | 1.5 | 0.7 | 0.9 | 0.8 | 1.6 | 1.6 |
| YA 3 B | 18 | 5,981 | 0.46 | 5.06 | 0.08 | 1.22 | 0.09 | 3.03 | 1.33 | 0.95 | 1.2 | 0.5 | 0.8 | 0.5 | 1.6 | 1.7 |
| YA 4 B | 13 | 11,557 | 0.61 | 6.13 | 0.10 | 1.32 | 0.13 | 4.93 | 1.17 | 1.18 | 1.1 | 0.6 | 0.8 | 0.7 | 1.5 | 2.6 |
| YA 5 B | 10 | 12,061 | 0.65 | 6.57 | 0.09 | 1.66 | 0.13 | 10.65 | 2.90 | 1.43 | 1.8 | 0.7 | 1.1 | 0.7 | 1.4 | 1.4 |
| Avg. YB | 13 | 8,446 | 0.61 | 6.11 | 0.10 | 1.34 | 0.13 | 6.49 | 2.03 | 1.41 | 1.4 | 0.6 | 0.9 | 0.7 | 1.5 | 1.7 |
| UCC | 1.4 | 370 | 0.5 | 5 | 0.1 | 4.2 | 0.15 | | | | | | | | | |

Grain size calculations of Md50 and sorting are as for Folk and Ward (1957)

*na* not analyzed, *nd* not detected, *UCC* upper continental crust values of Taylor and McLennan (1985)

functioning of rivers (Owens et al. 2005). Surface and bottom sediments were collected assuming that opening the Arase dam gate had effect on the Kuma River and Yatsushiro Bay sediments. Thus, surface sediments would represent the recent, post-dam, situation and the bottom sediments represent the composition before opening of the dam gate. With the opening of the dam gates the flooded upstream area changed to a fluvial system dominated by flowing water. Because the fine fraction increases downstream, fine grains have been transported from above the dam and are gradually accumulating on the downstream surface sediments (Fig. 4a). In the bottom sediments, increased fine grain content along the river indicates that during flushing, trapped fine sediments in the dam lake were gradually transported downstream. When the dam gates were opened the dam area is converted to a fluvial system and the flow rate in both upstream and downstream regions increased. Therefore, most of the bed load sediments were transported downstream and also point bars were formed along the river banks. Due to a higher coarse component upstream, the sediments at the dam also have a higher coarse grain percentage, which gradually decreases downstream (Fig. 4b).

When plot Zn, $P_2O_5$, $TiO_2$, and Zr against $Fe_2O_3$ for surface and bottom core sediment samples, the difference between chemical content are not significant (Fig.. 5 a, b, c, d). The surface and the bottom sediments have the same chemical composition within the variance of the plots. LOI is low, indicating that there is little carbonate dilution taking place (Table 1). It seems, therefore, that quartz dilution is affecting the sediments, involving larger quartz grains, which would not change the chemical composition of the surface or bottom sediments. Because there are no chemical changes, these sediments are not post dam and pre dam sediments. It is likely that the grain size variation in the surface and the bottom sediments are due to the opening of the dam during winter and also removing the dam gates.

## Composition change since 2002

### *Kuma River*

This study shows that the elemental compositions of surface and bottom sediments are variable, but lie within a relatively small range of values. From this, we infer that the source of these sediments was very similar throughout the period represented by the short core

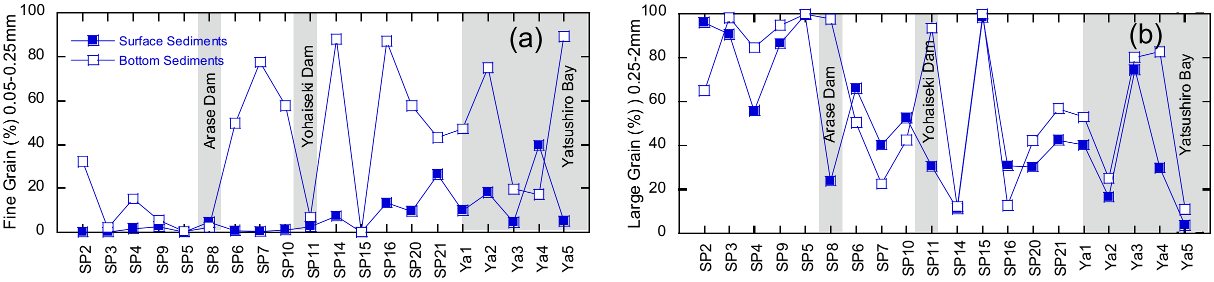

**Fig. 4** Grain size variation of the **a** fine (0.25–0.005 mm) and **b** medium (0.25–2 mm) fractions of surface and bottom sediments

collections. However, when compared with a previous study (Dozen and Ishiga 2002), it can be seen that prior to 2002 Arase dam sediments had much higher elemental concentrations for Pb, Zn, Cu, Zr, Nb, and $TiO_2$ (Fig. 6). The Kuma River sediment samples of Dozen and Ishiga (2002) show highly scattered values (larger range) indicating significant chemical variation along the stream specially seen in Zr and Nb (Fig. 6). This may be due to bed sediment heterogeneity in grain size in 2002. The elemental concentrations of most of the elements are high in 2002 and lowest in 2011 while 2013 has moderate compositions (arrows in Fig. 6). The correlations of each year have been given in the figures. Arase dam sediments show very high values in 2002 for all measured elements (Pb, Zn, Cu, Zr, Nb, and $TiO_2$) except Cr. The Fe of the river sediments is lower in 2011 than that in 2002. However, Fe content increased in 2013 (Fig. 6a, b). Fe is used as an indicator of oxygen availability in sediments and is well documented (Heggie 1992; Ahmed et al. 2007) where lower Fe content relates with higher oxygen availability. Thus, the low Fe in 2011 shows more oxygen availability in the sediments than in 2002 indicating more oxic conditions in 2011, but there is a trend toward anoxia at present in 2013. Thus, the sediment elemental composition was high in 2002 due to high fine fraction. It decreased in 2011 due to the presence of coarser sediments, but then it increased slightly in 2013 due to an increase of the finer fraction. In 2002, the Arase dam had accumulated stagnant sediments indicating high chemical compositions of fines. But in 2011, elemental concentrations in the sediments decreased due to the flushing of the finer sediments following the removal of the dam gates and also due to opening the dam gates during the winter season. In 2013, increases in elemental concentrations may be due to continued flushing, bringing finer sediments into the system leading to retention of finer sediments. The highest values of trace elements, plotting in the upper right of all plots, are data on mud drapes from the river banks in 2013 (Fig. 6a–f). These mud drapes are caused by heavy rain fall in the upstream reach of the river which transports finer particles downstream. This fine clay is deposited in the river banks due to the gradual decrease of river water level after rain events. Sediment samples from 2002, 2011, and 2013 for Kuma River and Arase dam all plot significantly below the sediment trend line of Ahmed et al. (2007). The sediment baseline trend for Zn in Ahmed et al. (2007) was drawn using South Korean baseline lagoon and lake sediment data. Thus, this suggests that it may take a few more years to reach the normal (Baseline) sediment trend line in the Kuma River.

### *Yatsushiro Bay*

Yatsushiro Bay sediments also had much higher elemental concentrations in 2002 than they do today, as seen for Pb, Zn, Cu, Zr, and Nb (Fig. 7). In 2011 and 2013, Yatsushiro Bay surface samples had low Pb, Zn, and Cu and a good correlation with Fe (Fig. 7a, b). From this, we infer that the environmental conditions have recovered because fine sediments are actively being deposited in the bay area at present (Figs. 7 and 4a). The correlations of 2002, 2011, and 2013 clearly show the chemical difference in each year. Like the Kuma River, Yatsushiro Bay sediments also show high elemental concentrations in 2002, decreasing in 2011, and slightly increasing in 2013. Thus, the sediment compositions of the Kuma River and the Yatsushiro Bay are consistent with each other and are subjected to the same sedimentary processes. The Yatushiro Bay sediments in 2002 plot very close to the sediment trend line of Ahmed et al. 2007. However, the 2011 and 2013 sediment data plot a bit below the trend line. This suggests that the sediments have been altered since 2002 and that

**Table 2** Elemental concentrations of fine (0.075–0.25 mm) and medium (0.25–0.85 mm) sediments of the Kuma River and Yatsushiro bay of 2011

| Sample no. | Trace elements (ppm) | | | | | | | | | | | | | | | | Major elements (wt%) | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | As | Pb | Zn | Cu | Ni | Cr | V | Sr | Y | Nb | Zr | Th | Sc | TS | F | Br | I | Cl | $TiO_2$ | $Fe_2O_3$ | MnO | CaO | $P_2O_5$ |
| SP2S-fine | 6 | 17 | 74 | 25 | 34 | 69 | 115 | 123 | 21 | 8 | 96 | 8 | 12 | 293 | 144 | 2 | 17 | nd | 0.54 | 5.74 | 0.10 | 1.31 | 0.10 |
| SP4S-fine | 7 | 13 | 74 | 22 | 35 | 96 | 147 | 139 | 20 | 9 | 99 | 7 | 16 | 279 | 115 | 2 | 7 | nd | 0.65 | 6.82 | 0.12 | 1.66 | 0.11 |
| SP6S-fine | 7 | 14 | 87 | 24 | 34 | 130 | 255 | 121 | 20 | 11 | 116 | 7 | 18 | 274 | 36 | 2 | nd | nd | 1.07 | 8.62 | 0.13 | 1.33 | 0.11 |
| SP8S-fine | 7 | 16 | 82 | 24 | 35 | 109 | 171 | 134 | 22 | 10 | 106 | 7 | 15 | 298 | 178 | 2 | 1 | nd | 0.74 | 7.26 | 0.13 | 1.42 | 0.11 |
| SP8B-fine | 10 | 20 | 111 | 32 | 44 | 79 | 153 | 118 | 28 | 11 | 123 | 11 | 16 | 796 | | 6 | 5 | nd | 0.73 | 7.10 | 0.14 | 1.31 | 0.22 |
| SP11S-fine | 6 | 15 | 74 | 25 | 36 | 83 | 127 | 129 | 20 | 9 | 99 | 7 | 16 | 289 | 75 | 2 | 12 | nd | 0.60 | 6.14 | 0.11 | 1.48 | 0.10 |
| SP14S-fine | 7 | 16 | 80 | 34 | 36 | 95 | 135 | 134 | 21 | 9 | 106 | 6 | 14 | 346 | 103 | 2 | 14 | nd | 0.61 | 6.14 | 0.11 | 1.46 | 0.11 |
| SP19 fine mix | 8 | 18 | 83 | 22 | 36 | 77 | 114 | 133 | 23 | 9 | 127 | 8 | 13 | 2,488 | 146 | 20 | 15 | 7,852 | 0.63 | 5.68 | 0.11 | 1.15 | 0.12 |
| Sozou S fine | 7 | 14 | 77 | 14 | 32 | 89 | 120 | 129 | 20 | 7 | 118 | 7 | 15 | 615 | 324 | 5 | 16 | nd | 0.62 | 5.64 | 0.10 | 1.11 | 0.09 |
| Sozou B fine | 7 | 14 | 80 | 16 | 34 | 95 | 123 | 131 | 20 | 7 | 111 | 7 | 13 | 697 | 46 | 4 | 18 | nd | 0.61 | 5.69 | 0.09 | 1.11 | 0.09 |
| YA-1 fine mix | 9 | 17 | 88 | 23 | 35 | 77 | 119 | 128 | 23 | 9 | 114 | 7 | 14 | 2,313 | 47 | 19 | 13 | 6,803 | 0.61 | 5.85 | 0.08 | 1.25 | 0.13 |
| YA-4 fine mix | 9 | 19 | 104 | 34 | 53 | 134 | 197 | 127 | 26 | 12 | 137 | 9 | 17 | 2,686 | 215 | 26 | | 15,972 | 0.84 | 7.88 | 0.13 | 1.37 | 0.17 |
| SP2S-med | 6 | 14 | 69 | 23 | 31 | 64 | 101 | 112 | 20 | 7 | 93 | 7 | 11 | 265 | 103 | 2 | 19 | nd | 0.47 | 5.37 | 0.09 | 1.23 | 0.09 |
| SP4S-med | 6 | 13 | 79 | 25 | 37 | 82 | 133 | 117 | 20 | 8 | 94 | 6 | 14 | 264 | 61 | 2 | 11 | nd | 0.62 | 6.95 | 0.12 | 1.48 | 0.10 |
| SP6S-med | 6 | 15 | 75 | 21 | 30 | 64 | 108 | 127 | 20 | 7 | 95 | 8 | 14 | 265 | | 2 | 9 | nd | 0.47 | 5.58 | 0.09 | 1.31 | 0.09 |
| SP8S-med | 6 | 13 | 74 | 21 | 35 | 74 | 115 | 123 | 19 | 7 | 91 | 7 | 14 | 266 | 192 | 2 | 6 | nd | 0.50 | 6.06 | 0.11 | 1.39 | 0.10 |
| SP8B-med | 9 | 20 | 110 | 34 | 43 | 83 | 144 | 112 | 27 | 10 | 114 | 10 | 15 | 787 | 160 | 6 | 11 | nd | 0.67 | 6.92 | 0.13 | 1.33 | 0.20 |
| SP11S-med | 7 | 15 | 76 | 25 | 36 | 66 | 111 | 109 | 22 | 8 | 98 | 8 | 12 | 286 | 89 | 2 | 11 | nd | 0.53 | 5.72 | 0.10 | 1.24 | 0.09 |
| SP14S-med | 8 | 17 | 82 | 26 | 39 | 70 | 112 | 113 | 23 | 9 | 100 | 8 | 16 | 409 | | 3 | 14 | nd | 0.53 | 5.79 | 0.11 | 1.35 | 0.10 |
| SP19 med mix | 9 | 14 | 79 | 17 | 39 | 76 | 101 | 119 | 20 | 7 | 103 | 7 | 11 | 2,322 | | 16 | 18 | 14,599 | 0.52 | 5.47 | 0.10 | 1.12 | 0.11 |
| Sozou S med | 8 | 14 | 80 | 13 | 37 | 61 | 88 | 112 | 18 | 6 | 89 | 6 | 9 | 603 | 103 | 5 | 14 | nd | 0.46 | 5.51 | 0.09 | 1.07 | 0.09 |
| Sozou B med | 8 | 12 | 79 | 15 | 35 | 69 | 94 | 111 | 17 | 6 | 86 | 6 | 10 | 679 | 133 | 4 | 18 | nd | 0.43 | 5.55 | 0.08 | 1.07 | 0.09 |
| YA-1 med mix | 10 | 15 | 75 | 18 | 35 | 68 | 96 | 121 | 20 | 7 | 98 | 7 | 13 | 2,666 | 128 | 16 | 18 | 10,856 | 0.53 | 5.35 | 0.07 | 1.29 | 0.11 |
| YA-4 med mix | 6 | 14 | 71 | 20 | 39 | 94 | 103 | 123 | 18 | 6 | 89 | 5 | 13 | 1,559 | 227 | 13 | 16 | 9,228 | 0.50 | 5.60 | 0.09 | 1.30 | 0.11 |
| Dozen and Isiga 2,000 | | | | | | | | | | | | | | | | | | | | | | | |
| Arase | | | | | | | | | | | | | | | | | | | | | | | |
| Min | 7 | 17 | 89 | 30 | 28 | 60 | 132 | 131 | 22 | 9 | 160 | 10 | na | 633 | na | 2 | 15 | na | 0.62 | 6.12 | na | 1.08 | 0.13 |
| Max | 13 | 28 | 160 | 45 | 41 | 76 | 153 | 155 | 30 | 12 | 207 | 13 | na | 1,574 | na | 22 | 42 | na | 0.73 | 7.08 | na | 1.52 | 0.29 |
| Yatsushiro | | | | | | | | | | | | | | | | | | | | | | | |
| Min | 8 | 18 | 92 | 26 | 29 | 62 | 116 | 147 | 24 | 9 | 156 | 9 | na | 3,157 | na | 45 | 31 | na | 0.61 | 5.62 | na | 1.08 | 0.13 |

**Table 2** (continued)

| Sample no. | Trace elements (ppm) | | | | | | | | | | | | | | | | Major elements (wt%) | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | As | Pb | Zn | Cu | Ni | Cr | V | Sr | Y | Nb | Zr | Th | Sc | TS | F | Br | I | Cl | $TiO_2$ | $Fe_2O_3$ | MnO | CaO | $P_2O_5$ |
| Max | 13 | 24 | 131 | 38 | 41 | 87 | 148 | 172 | 29 | 11 | 194 | 13 | na | 9,477 | na | 87 | 61 | na | 0.71 | 7.00 | na | 1.99 | 0.16 |
| Kuma River | | | | | | | | | | | | | | | | | | | | | | | |
| Min | 7 | 13 | 75 | 21 | 20 | 50 | 91 | 84 | 17 | 6 | 109 | 7 | na | 368 | na | 1 | 25 | na | 0.41 | 5.16 | na | 0.78 | 0.01 |
| Max | 12 | 29 | 195 | 32 | 28 | 74 | 141 | 184 | 27 | 10 | 166 | 12 | na | 2,044 | na | 36 | 58 | na | 0.64 | 7.58 | na | 1.81 | 0.39 |

*na* not analyzed, *nd* not detected

it may take a few more years to reach the normal (baseline) sediment trend line.

Arsenic is usually associated with organic rich sediment where pyrite forms under low temperature conditions (Smedley and Kinniburgh 2001), conditions which are common in river or bay environments. The UCC value for As is 4.8 given by Rudnick and Gao (2005). The 2011 Yatsushiro Bay sediments have high As (10–12 ppm, Table 1) in both surface and bottom sediments, two times higher than UCC. All river sediments are less than 10 ppm in 2011 (Table 1). Thus, the sediments of the bay environment, containing more organic matter (related with LOI content, Table 1), may be polluted with As in some locations and also may derive As from the background mafic rocks of the area. Organics are associated with anoxia and can lead to pyrite formation. The presence of pyrite was observed in the sediments using a binocular microscope. Because arsenic is highly toxic, the accumulation of As is an important issue and must be monitored regularly. However, in 2013, the As content decreased in the bay and thus also indicates environmental recovery.

Br and Zn in Arase dam sediments were used by Dozen and Ishiga (2002) to trace the accumulation of algae, which was preserved in the sediments under reducing conditions. The present Kuma River core bottom sediments are slightly higher in Br than the surface sediments, but no difference in Zn content. A comparison of Dozen and Ishiga (2002) data with the 2011 data (Figs. 6 and 7, and Tables 1, 2, and 3) indicates that conditions have changed since the dam opening. Although high bromides and Zn were found in 2002, at present both have decreased notably. Therefore, it seems that biogenic processes and redox conditions have changed in the most recent years (2011 to 2013).

### *Provenance indicators*

The possible role of provenance, sorting or accumulation of heavy minerals such as zircon, monazite, or apatite, can be evaluated using Zr/Sc and Th/Sc ratios (McLennan et al. 1993). The Th/Sc ratio is a sensitive index of the bulk composition of the source (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 bivariant Zr/Sc–Th/Sc plots give an indication of source composition and heavy mineral concentration when compared with

**Table 3** Elemental concentrations of Kuma River and Yatsushiro Bay for 2013

| Sample no. | Trace elements (ppm) | | | | | | | | | | | | | | | | Major elements (wt%) | | | | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | As | Pb | Zn | Cu | Ni | Cr | V | Sr | Y | Nb | Zr | Th | Sc | TS | F | Br | I | Cl | $TiO_2$ | $Fe_2O_3$ | MnO | CaO | $P_2O_5$ | As EF | Pb EF | Zn EF | Cu EF | Ni EF | Cr EF |
| Kuma River | | | | | | | | | | | | | | | | | | | | | | | | | | | | | |
| 1A | 6 | 13 | 70 | 26 | 35 | 93 | 156 | 141 | 20 | 10 | 127 | 9 | 10 | 339 | nd | 2 | 13 | nd | 0.70 | 6.52 | 0.12 | 1.31 | 0.09 | 0.92 | 0.44 | 0.68 | 0.70 | 1.21 | 1.84 |
| 1B | 6 | 13 | 71 | 25 | 33 | 94 | 154 | 138 | 19 | 9 | 136 | 8 | 10 | 340 | 21 | 2 | 15 | nd | 0.75 | 6.30 | 0.11 | 1.24 | 0.08 | 1.02 | 0.48 | 0.71 | 0.71 | 1.19 | 1.91 |
| 1C | 10 | 23 | 119 | 44 | 50 | 100 | 194 | 154 | 32 | 15 | 205 | 13 | 14 | 876 | 332 | 13 | nd | nd | 1.00 | 8.03 | 0.17 | 1.41 | 0.22 | 1.25 | 0.65 | 0.94 | 0.98 | 1.40 | 1.61 |
| 2A | 7 | 14 | 67 | 28 | 31 | 65 | 129 | 129 | 21 | 9 | 131 | 9 | 7 | 335 | 21 | 1 | 13 | nd | 0.63 | 5.71 | 0.10 | 1.07 | 0.08 | 1.23 | 0.54 | 0.74 | 0.87 | 1.22 | 1.45 |
| 2B | 6 | 16 | 72 | 28 | 31 | 88 | 151 | 136 | 21 | 10 | 145 | 8 | 8 | 371 | nd | 2 | 17 | nd | 0.77 | 6.33 | 0.11 | 1.13 | 0.09 | 0.88 | 0.56 | 0.73 | 0.78 | 1.11 | 1.78 |
| 2C | 8 | 21 | 100 | 37 | 40 | 96 | 175 | 157 | 27 | 13 | 191 | 12 | 12 | 770 | 32 | 8 | 3 | nd | 0.87 | 7.03 | 0.13 | 1.37 | 0.19 | 1.14 | 0.66 | 0.90 | 0.95 | 1.26 | 1.76 |
| 3A | 10 | 25 | 125 | 47 | 51 | 100 | 200 | 151 | 32 | 14 | 206 | 14 | 14 | 953 | 32 | 13 | nd | nd | 0.94 | 8.04 | 0.17 | 1.40 | 0.23 | 1.19 | 0.70 | 0.99 | 1.05 | 1.42 | 1.60 |
| 3B | 10 | 25 | 125 | 46 | 51 | 97 | 191 | 152 | 32 | 14 | 205 | 14 | 12 | 984 | 103 | 13 | nd | nd | 0.92 | 8.08 | 0.18 | 1.41 | 0.22 | 1.27 | 0.69 | 0.98 | 1.02 | 1.41 | 1.55 |
| 3C | 7 | 16 | 74 | 27 | 36 | 85 | 151 | 135 | 21 | 10 | 151 | 9 | 10 | 360 | nd | 2 | 12 | nd | 0.74 | 6.19 | 0.11 | 1.09 | 0.09 | 1.08 | 0.57 | 0.76 | 0.80 | 1.30 | 1.77 |
| 4A | 10 | 24 | 121 | 43 | 50 | 106 | 195 | 155 | 31 | 14 | 201 | 13 | 16 | 916 | 103 | 14 | nd | nd | 0.96 | 7.99 | 0.21 | 1.45 | 0.22 | 1.24 | 0.67 | 0.96 | 0.96 | 1.40 | 1.71 |
| 4B | 7 | 15 | 73 | 29 | 50 | 105 | 146 | 134 | 21 | 9 | 136 | 9 | 7 | 355 | 2 | 13 | nd | nd | 0.68 | 6.31 | 0.11 | 1.13 | 0.08 | 1.08 | 0.54 | 0.73 | 0.81 | 1.77 | 2.15 |
| 5 | 7 | 15 | 70 | 27 | 46 | 77 | 146 | 131 | 21 | 9 | 133 | 9 | 9 | 344 | 173 | 2 | 13 | nd | 0.72 | 6.12 | 0.11 | 1.05 | 0.08 | 1.09 | 0.53 | 0.73 | 0.81 | 1.69 | 1.62 |
| Min | 6 | 13 | 67 | 25 | 31 | 65 | 129 | 129 | 19 | 9 | 127 | 8 | 7 | 335 | 2 | 1 | 3 | | 0.63 | 5.71 | 0.10 | 1.05 | 0.08 | | | | | | |
| Max | c10 | 25 | 125 | 47 | 51 | 106 | 200 | 157 | 32 | 15 | 206 | 14 | 16 | 984 | 332 | 14 | 17 | | 1.00 | 8.08 | 0.21 | 1.45 | 0.23 | | | | | | |
| Average | 8 | 18 | 91 | 34 | 42 | 92 | 166 | 143 | 25 | 11 | 164 | 10 | 11 | 579 | 91 | 7 | 12 | | 0.81 | 6.89 | 0.14 | 1.26 | 0.14 | 1.12 | 0.59 | 0.82 | 0.87 | 1.36 | 1.73 |
| Yatsushiro Bay | | | | | | | | | | | | | | | | | | | | | | | | | | | | | |
| 1 | 8 | 15 | 73 | 21 | 36 | 69 | 129 | 176 | 23 | 11 | 197 | 10 | 8 | 1,458 | nd | 12 | 6 | 2,494 | 0.69 | 5.33 | 0.08 | 1.15 | 0.09 | 1.43 | 0.65 | 0.87 | 0.72 | 1.51 | 1.65 |
| 3 | 9 | 18 | 81 | 29 | 37 | 79 | 141 | 179 | 24 | 10 | 176 | 9 | 10 | 3,885 | nd | 21 | 14 | 5,684 | 0.76 | 5.93 | 0.08 | 1.51 | 0.12 | 1.45 | 0.67 | 0.87 | 0.89 | 1.41 | 1.71 |
| 4 | 9 | 18 | 86 | 28 | 35 | 83 | 146 | 175 | 24 | 10 | 174 | 10 | 10 | 3,449 | nd | 30 | 19 | 10,572 | 0.76 | 6.04 | 0.10 | 1.57 | 0.15 | 1.54 | 0.65 | 0.91 | 0.83 | 1.30 | 1.76 |
| 5 | 8 | 14 | 71 | 25 | 34 | 85 | 129 | 156 | 20 | 8 | 129 | 8 | 10 | 1,565 | 61 | 12 | 19 | 7,405 | 0.65 | 5.62 | 0.11 | 1.38 | 0.10 | 1.37 | 0.54 | 0.80 | 0.79 | 1.35 | 1.94 |
| 6 | 11 | 22 | 104 | 40 | 53 | 94 | 170 | 151 | 30 | 13 | 194 | 13 | 10 | 1,944 | 60 | 39 | 11 | 9,307 | 0.88 | 7.37 | 0.15 | 1.02 | 0.18 | 1.55 | 0.67 | 0.90 | 0.99 | 1.61 | 1.63 |
| 7 | 8 | 17 | 80 | 24 | 38 | 94 | 158 | 156 | 22 | 9 | 153 | 10 | 10 | 2,434 | nd | 16 | 19 | 7,062 | 0.78 | 6.20 | 0.09 | 1.28 | 0.14 | 1.24 | 0.61 | 0.82 | 0.70 | 1.37 | 1.94 |
| 8 | 6 | 11 | 63 | 14 | 30 | 114 | 157 | 164 | 19 | 9 | 195 | 6 | 9 | 1,196 | nd | 9 | 16 | 4,525 | 0.83 | 5.54 | 0.09 | 1.14 | 0.07 | 1.05 | 0.45 | 0.72 | 0.46 | 1.20 | 2.65 |
| 9 | 9 | 14 | 78 | 17 | 30 | 90 | 134 | 171 | 21 | 9 | 143 | 8 | 10 | 2,368 | 49 | 16 | 20 | 9,254 | 0.68 | 5.57 | 0.10 | 1.69 | 0.11 | 1.53 | 0.57 | 0.89 | 0.56 | 1.20 | 2.08 |
| 19A | 9 | 21 | 89 | 34 | 44 | 91 | 167 | 155 | 28 | 14 | 215 | 11 | 10 | 1,293 | 61 | 16 | 4 | 3,123 | 0.88 | 6.93 | 0.11 | 1.07 | 0.16 | 1.26 | 0.67 | 0.81 | 0.88 | 1.44 | 1.70 |
| 19B | 8 | 18 | 86 | 33 | 48 | 100 | 159 | 154 | 26 | 12 | 214 | 11 | 9 | 1,293 | 5 | 13 | 12 | 2,929 | 0.85 | 6.37 | 0.09 | 1.08 | 0.14 | 1.24 | 0.63 | 0.86 | 0.92 | 1.69 | 2.01 |
| 20 | 7 | 19 | 94 | 33 | 46 | 103 | 157 | 157 | 25 | 11 | 194 | 10 | 8 | 1,426 | 156 | 20 | 17 | 6,579 | 0.81 | 6.26 | 0.09 | 1.04 | 0.15 | 1.18 | 0.69 | 0.95 | 0.93 | 1.65 | 2.11 |
| Min | 6 | 11 | 63 | 14 | 30 | 69 | 129 | 151 | 19 | 8 | 129 | 6 | 8 | 1,196 | 5 | 9 | 4 | 2,494 | 0.65 | 5.33 | 0.08 | 1.02 | 0.07 | | | | | | |

**Table 3** (continued)

| Sample no. | Trace elements (ppm) | | | | | | | | | | | | | | | | Major elements (wt%) | | | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | As | Pb | Zn | Cu | Ni | Cr | V | Sr | Y | Nb | Zr | Th | Sc | TS | F | Br | I | Cl | $TiO_2$ | $Fe_2O_3$ | MnO | CaO | $P_2O_5$ | As EF | Pb EF | Zn EF | Cu EF | Ni EF | Cr EF |
| Max | 11 | 22 | 104 | 40 | 53 | 114 | 170 | 179 | 30 | 14 | 215 | 13 | 10 | 3,885 | 156 | 39 | 20 | 10,572 | 0.88 | 7.37 | 0.15 | 1.69 | 0.18 | | | | | | |
| Average | 8 | 17 | 82 | 27 | 39 | 91 | 150 | 163 | 24 | 11 | 180 | 10 | 9 | 2,028 | 65 | 18 | 14 | 6,267 | 0.78 | 6.11 | 0.10 | 1.27 | 0.13 | 1.35 | 0.62 | 0.85 | 0.79 | 1.43 | 1.93 |

*nd* not detected

compositions of average volcanic and plutonic rocks. The Th/Sc–Zr/Sc plot for Kuma River and Yatsushiro Bay sediments for the years 2011 and 2013 shows that the composition has changed between the two sampling intervals (Fig. 8a). The surface and the bottom sediments of 2011 have the same composition. The Kuma River sediments have moved towards the composition of PAAS, while both Kuma River and the Yatsushiro Bay compositions are moving toward the Rhyolite to Dacite line (Fig. 8a). This is further shown by Zr/Ti–Th/Ti (Fig. 8b; Roser et al. 2000) which also gives the same result as for Th/Sc–Zr/Sc.

Patterns of similarity in elemental content can be portrayed using cluster analysis of the sediment sample data. The cluster analysis included grain size data to eliminate a grain size effect and to present the controlling elements on the basis of provenance-related elements for 2011 (Sr, Nb, Y, Zr, Th, Sc, $TiO_2$, CaO, MnO, F, Br, Cl, I) and elements not tied to provenance (As, Pb, Zn, Cu, Ni, V, Cr, $P_2O_5$, $Fe_2O_3$,TS—total sulfur). The cluster analysis was done using the complete linkage method, a distance measure of absolute correlation and was partitioned into five clusters for best results.

The five clusters of the non-provenance elements (Fig. 9a) are cluster one: SP2, SP4, and SP14. Cluster two: SP3, SP5, and SP15. Cluster three has only SP9 which is from a tributary stream. Cluster four: Sp8, SP6, SP7, SP10, SP11, SP12, and SP24. Cluster five has SP16, SP17, Sp18, SP19, SP20, SP21, Ya1, Ya2, Ya3, Ya4, and Ya5. These clusters clearly separate the upstream, downstream, and the bay area except for SP14, SP15, and SP24.

The five clusters for the provenance elements (Fig. 9b) are cluster one: SP2 and SP9. Cluster two: SP3, Sp5, SP15, and Ya3. Cluster three: SP4, SP6, SP7, SP10, SP12, SP24, and Ya1. Cluster four: SP8, SP11, SP14, SP16, Sp17, SP18, SP20, Ya2, and Ya5. Cluster five has SP19, SP21, and Ya4. The provenance cluster does not show a clear separation of upstream, downstream and bay areas. When the provenance and non-provenance element clusters are compared, the provenance elements clusters are clearly weaker. The cluster analysis (Fig. 9a, b) shows that variation in the elements not related to provenance dominate the sediment composition. Thus, the elements that are related to environmental change: As, Pb, Zn, Cu, Ni, Cr, V, and Fe, and total sulfur are related to the chemical behavior of the river and bay sediments. However, a more detailed study of Yatsushiro Bay is needed during low tide for

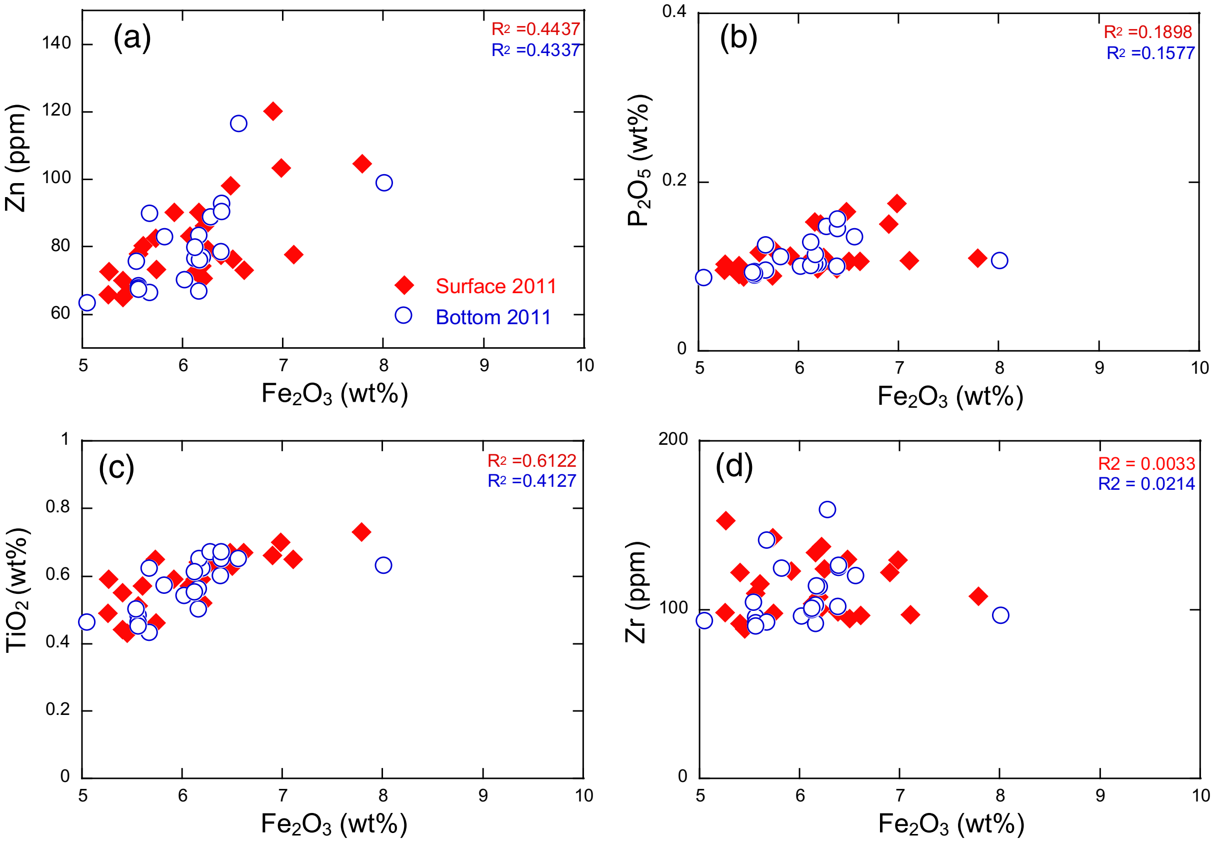

**Fig. 5** **a** Zn–Fe$_2$O$_3$, **b** P$_2$O$_5$–Fe$_2$O$_3$, **c** TiO$_2$–Fe$_2$O$_3$, and**d** Zr–Fe$_2$O$_3$ plots showing difference of composition showing provenance and environmental status for bottom and surface sediments in 2011

a better environmental picture. In order to better understand the variation in the chemistry of the sediments, a chemical analysis of the fine and medium fractions is discussed in the next section.

## Fine (0.075–0.25 mm) and medium (0.25–0.85 mm) fraction of 2011

The grain size of river bed sediments is an important property of streams because it is one of the major factors controlling channel morphology and hydraulics. The downstream variation in sediment size, which is characterized by a complex pattern rather than by a simple decreasing trend, can be portrayed by sediment fraction data (Surian 2002). Since there was no major change in the chemical composition of the surface and bottom sediments of the Kuma River, the chemical composition of two grain size fractions in 2011 surface sediments was investigated (Table 2, Online Resource 2). The Kuma River sediments are mainly comprised of sand sized fractions. It is widely agreed that the process of downstream fining of sands depends on some combination of abrasion and sorting, although arguments continue on their relative importance (Ferguson et al. 1998). The high strontium, Ti, Zr, and V in the fine fraction below the Arase dam (Table 2, Online Resource 2) indicate downstream fining. These elements are in heavy minerals and to accumulate in the fine fraction undergoing fractionation. The rate of change in bed material grain size has important implications for downstream changes in flow resistance and for sediment transport (Reid et al. 1997). The very high elemental concentrations at Yatsushiro Bay show that river sediments are transported along the stream bed and are deposited in Yatsushiro Bay. However, no obvious dilution or removal due to currents or wave actions can be seen in Yatsushiro Bay because Zn and Cu are elevated at this location. Co-precipitation of iron hydroxide along with the scavenging of other metals has been suggested as the principal mechanism explaining the accumulation

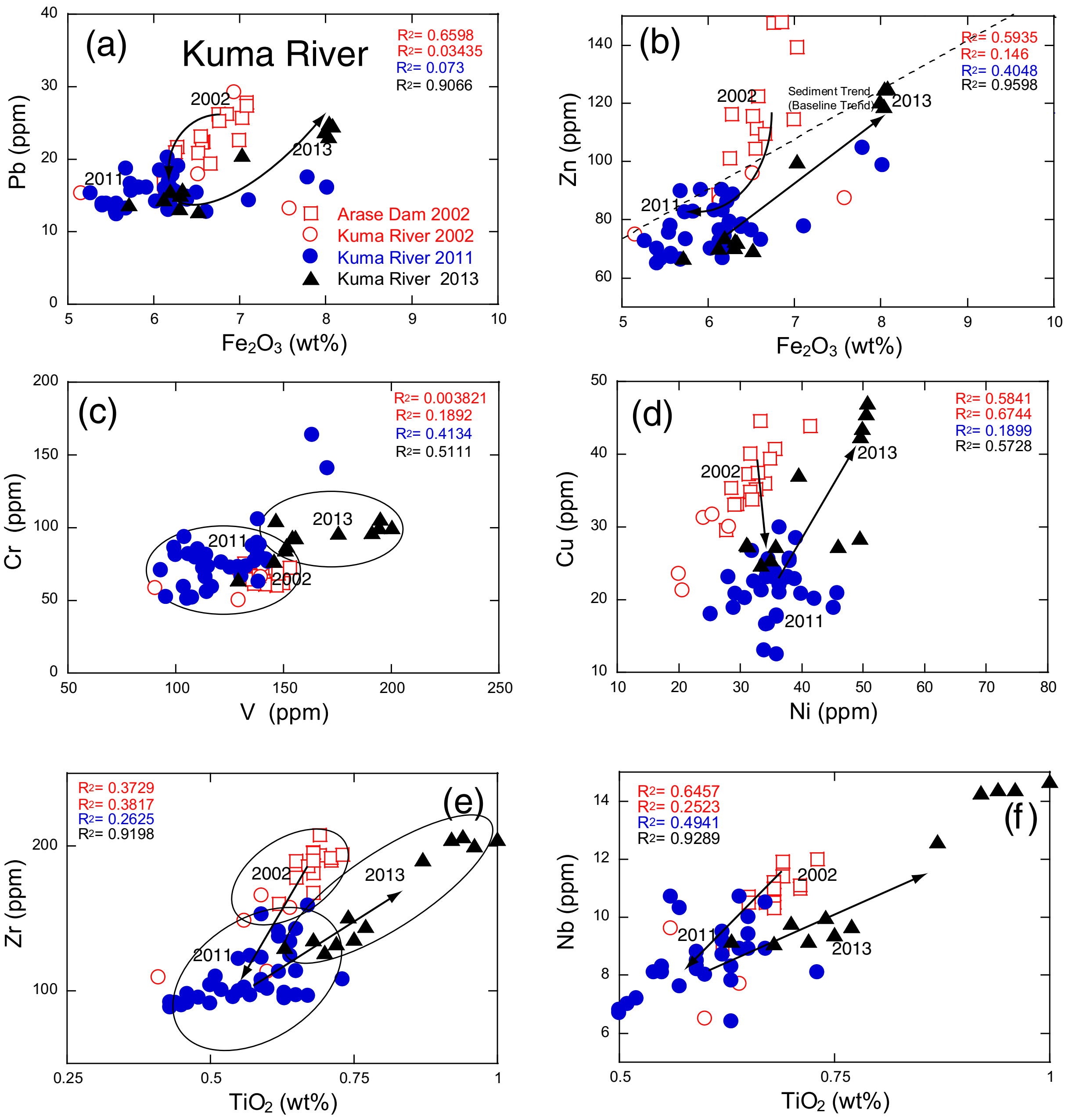

**Fig. 6** Variation diagrams of the Kuma River for **a** Pb–$Fe_2O_3$, **b** Zn–$Fe_2O_3$, **c** Cr–V, **d** Cu–Ni, **e** Zr–$TiO_2$, and **f** Nb–$TiO_2$. The data is compared with results from Dozen and Ishiga (2002) from the same locations. The plots **a**, **b**, **c**, and **d** are mainly related with environmentally related elements and the elements Cr, Zr, Nb, and $TiO_2$ is related with grain size. The correlation years 2002, 2011, and 2013 are given in each plot. The *arrows* show that 2002 has the highest values and the 2011 values have been decreased and 2013 has been increased or is the same as 2011

of Cu and Zn in estuarine sediments (Balachandran et al. 2006). The 2011 data for Zn and Cu shows much higher values (Online Resource 2) than UCC (Zn—71 ppm, Cu—25 ppm) in the Yatsushiro Bay. The river values are much lower, except for one location for Cu in the fine fraction. Therefore, the background values are also lower. Thus, the 2011 sediments in Yatsushiro Bay may be contaminated by Zn and Cu. At SP8 bottom sediments have elevated values for Ti, Fe, Zn, P, Ni, Pb, Cu, and As (Online Resource 2). At

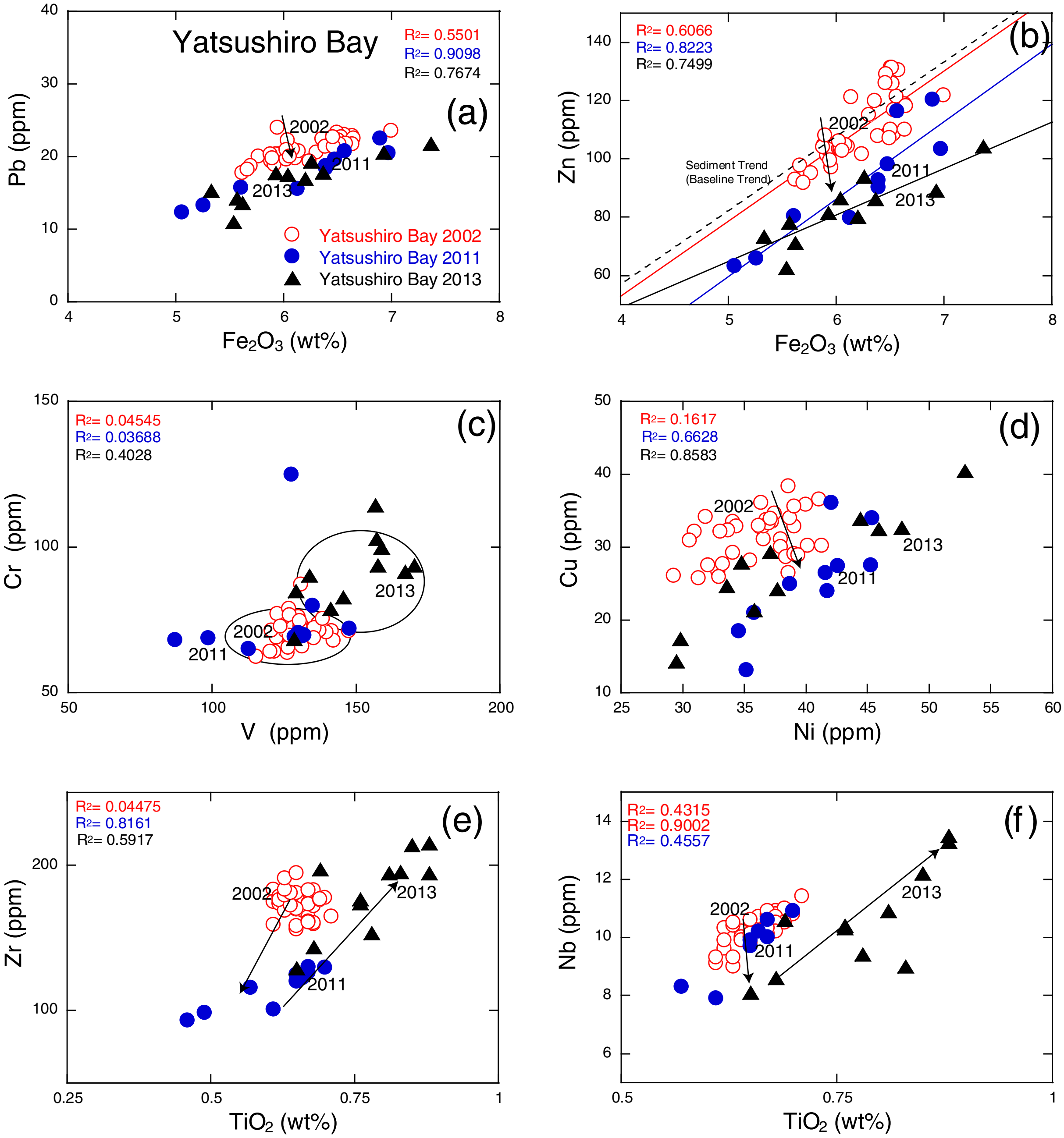

**Fig. 7** Variation diagrams of the Yatsushiro Bay for **a** Pb–Fe$_2$O$_3$, **b** Zn–Fe$_2$O$_3$, **c** Cr–V, **d** Cu–Ni, **e** Zr–TiO$_2$, and**f** Nb–TiO$_2$. The data is compared with results from Dozen and Ishiga (2002) from the same locations. The plots **a**, **b**, **c**, and **d** are mainly related with environmentally related elements and the elements Cr, Zr, Nb, and TiO$_2$ is related with grain size. The correlation years 2002, 2011, and 2013 are given in each plot. The *arrows* show that 2002 has highest values, and the 2011 values have been decreased and 2013 has been increased or is the same as 2011

this location, fine sediment has high values of Ti, Fe, Zr, and As. All other measured elements in the SP8 sediments have the same concentrations in the fine (0.075–0.25 mm) and medium (0.25–0.85 mm) fractions. SP8 is the sampling point for Arase dam, and is a location on a curve in the river. After the dam removal some of the sediments flushed from the river bed are deposited in the river bank forming sand point bars and the rest are

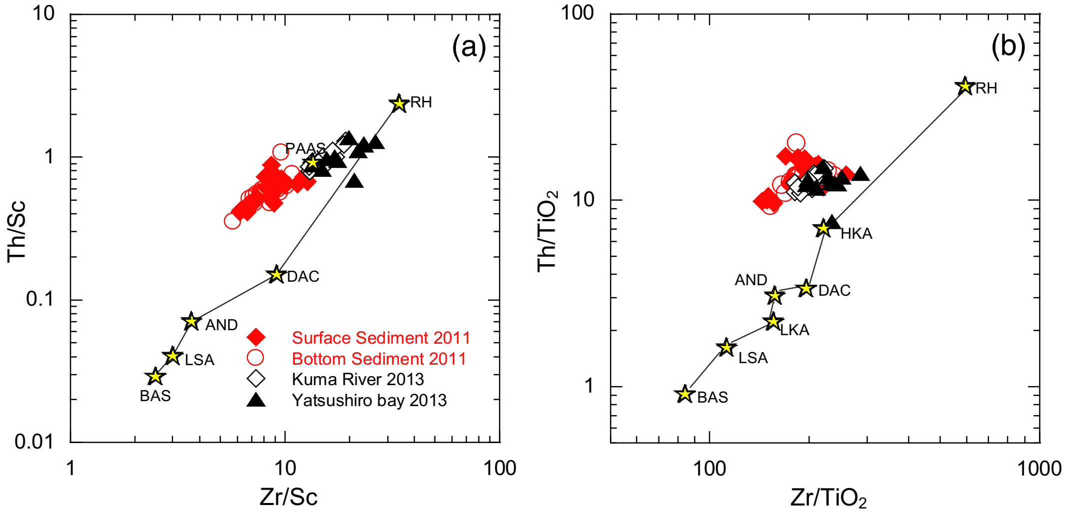

**Fig. 8** **a** Zr/Sc–Th/Sc (McLennan et al. 1993) and **b** Zr/Ti–Th/Ti (Roser et al. 2000) for the surface and bottom sediments for the Kuma River in 2011 and Kuma River and Yatsushiro Bay sediments in 2013. Stars BAS, LSA, AND, DAC, RHY: average basalt, low-silica andesite, andesite, dacite, and rhyolite, as plotted by Roser and Korsch (1999), representing a model source evolution trend. *PAAS* Post-Archaean Australian Shale (Taylor and McLennan 1985)

transported downstream. The SP8 bottom sediment represents the sediments accumulating while the Arase dam was in place. High Ti, P, Ni, As, Pb, Zn, and Cu in both size fractions also indicates that pre-removal sediments are still present in the dam lake site. Therefore, these sediments will need more time to be removed from the dam lake site and the natural conditions of the river sediment are not yet restored.

Although there is no change seen in the bulk surface and bottom sediments, there is an elemental difference in the surface sediments between different grain size fractions. The difference in fine fraction composition shows sediments have high concentrations of the heavy minerals. In the fine fractions, titanium, Fe, Zr, and V are high at the Arase dam location (SP8). These may relate to the heavy minerals zircon, magnetite, and ilmenite which may have been deposited before opening of the dam. It is a matter of concern that As content in the dam sediments are high. For As the fine fraction yields more than 10 ppm and the medium size fraction are 9 ppm. Arsenic levels reported in Rudnick and Gao (2005) indicate values of 4.8, and the Arase dam lake values are as much as two times higher. Therefore, this is a matter of some concern for the pre-dam removal sediments. However, after dam removal, the As concentration has dropped to 6–7 ppm in both fractions. Nevertheless, these values may also relate to natural factors such as background values.

Calcium, Sr, Zn, V, Ti, Zr, and Fe are high in the surface sediments of SP4, which is in the farthest upstream area. Ti, Fe, and V are much higher in the fine fraction than in the medium size fraction at SP4.

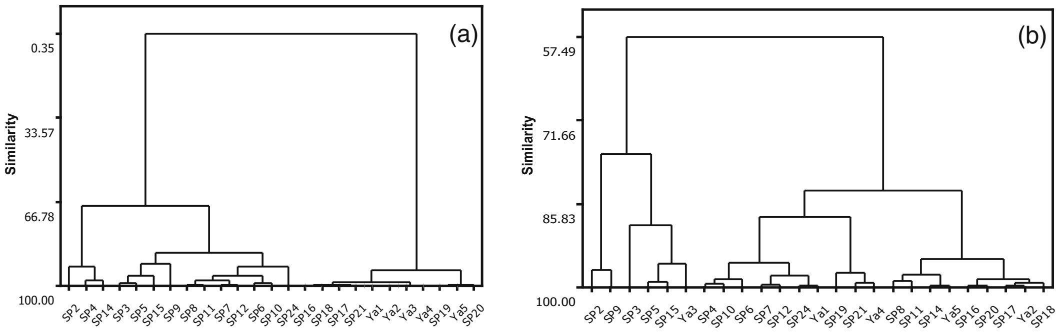

**Fig. 9** **a** Non-provenance elements cluster analysis plot. **b** Provenance elements cluster analysis plot of the Kuma River sediments

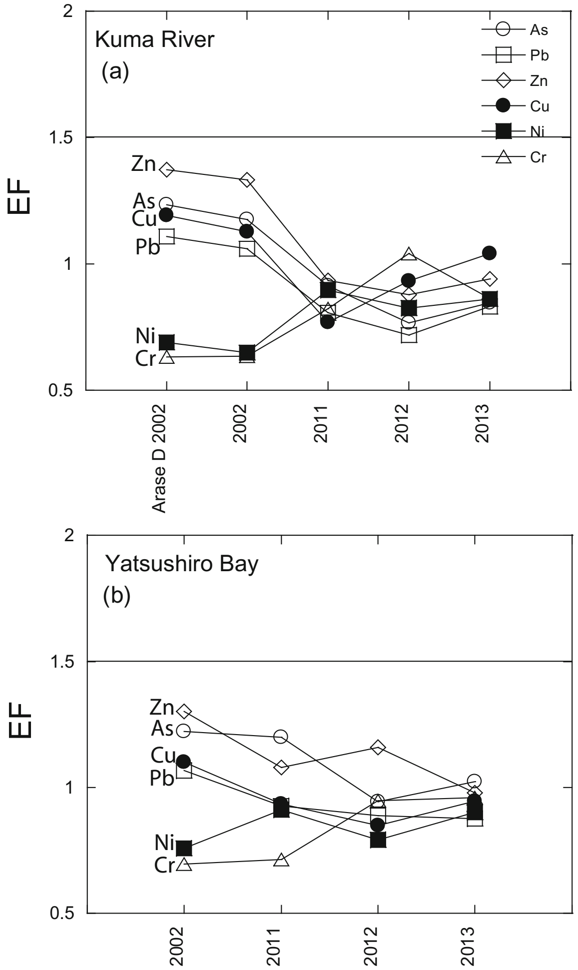

**Fig. 10** **a** The EF values of As, Pb, Zn, Cu, Ni, and Cr for the Kuma River for the years 2002, 2011, 2012, and 2013. **b** The EF values of As, Pb, Zn, Cu, Ni, and Cr for the Yatsushiro Bay for the years 2002, 2011, 2012, and 2013

Carbonates may be the main source of Ca and Sr, while all other elements relate to heavy minerals.

At the Sozo location, sediment elemental concentrations for both fractions are decreasing. This may be due to the vicinity of Yatsushiro Bay. Sediment undergoing transport moves through the Sozo region (river mouth) and into the bay, thus showing lower values at Sozo.

Therefore, considering all discussion above, it is evident that studies such as this one are very useful not only as a record of historical data for future studies. These studies are also useful for forming dam management strategies. When the processes that control the river and its sedimentation are better understood, they can be used for dam construction/destruction or maintenance issues in the future.

## Present environmental status

Trace metal concentrations (As, Pb, Zn, Cu, Ni, and Cr) in the Kuma River and Yatsushiro Bay sediments were compared with four established international standards to evaluate present pollution status for the elements analyzed (Table 4). Because comparison with a single guideline could be misleading four guidelines were used. The guidelines used were NYSDEC (New York State Department of Environmental Conservation) values of lower effect level (LEL) and severe effect level (SEL), ISQG (Interim Sediment Quality Guidelines) and probable effect level (PEL). The NYSDEC metals criteria are derived from Ministry of Ontario guidelines and NOAA data that make use of the screening level approach. The LEL for each metal is thus the lowest of either the Persaud et al. (1992) LEL or the Long and Morgan (1990) effect range—low. Similarly, the SEL for each metal is the lowest of either the Persaud et al. (1992) SEL or the Long and Morgan (1990) effect range—moderate. If either criterion is exceeded, sediments are considered contaminated. If both criteria are exceeded, the sediment is classified as severely impacted. If both the LEL and SEL criteria are exceeded, the metal may have a severe impact on the health of biota. If only the LEL criterion is exceeded, the metal may have a moderate impact on biotic health (NYSDEC New York State Department of Environmental Conservation 1999; Graney and Eriksen 2004). The Canadian Council of Ministers of the Environment also developed national Interim Sediment Quality Guidelines (ISQG) based on co-occurrence of chemical and biological data from the assessment of Great Lakes contaminated sediments (SAIC 2002). Using guideline values derived from large well-assessed data sets such as those above should result in reasonable sediment classifications, even though there may be differences in threshold levels.

Metal enrichment factor (EF) values of 0.5–1.5 suggest that the trace metals concerned may be derived entirely from crustal materials or natural weathering processes (Zhang and Liu 2002). Values greater than 1.5 suggest that a significant portion of the trace metal has been delivered from non-natural

**Table 4** Average values of elemental concentrations of Kuma River and Yatsushiro Bay for 2002 to 2013 and environmental guidelines (Japanese Island arc avg; Togashi et al. 2000)

| Element | As | Pb | Zn | Cu | Ni | Cr |
|---|---|---|---|---|---|---|
| Averages of raw data | | | | | | |
| Kuma River | | | | | | |
| Arase Dam (2002) | 10.9 | 23.3 | 126.3 | 37.0 | 32.4 | 65.5 |
| 2002 | 10.3 | 22.1 | 121.9 | 34.7 | 30.3 | 65.1 |
| 2011 | 7.3 | 15.4 | 78.2 | 21.9 | 39.2 | 78.8 |
| 2012 | 6.0 | 13.4 | 71.8 | 25.7 | 34.4 | 97.2 |
| 2013 | 7.8 | 18.2 | 90.5 | 33.7 | 41.8 | 92.3 |
| Yatsushiro Bay | | | | | | |
| 2002 | 10.3 | 21.4 | 114.7 | 32.6 | 34.0 | 68.8 |
| 2011 | 10.0 | 18.3 | 93.5 | 27.4 | 40.1 | 68.9 |
| 2012 | 7.8 | 17.8 | 104.2 | 25.4 | 36.9 | 92.2 |
| 2013 | 8.2 | 16.9 | 82.4 | 27.1 | 39.0 | 91.0 |
| Environmental guidelines | | | | | | |
| Japan Island Arc Avg. | 7.1 | 16.9 | 74.1 | 25 | 38 | 84 |
| LEL | 6 | 31 | 120 | 16 | 16 | 26 |
| SEL | 33 | 110 | 270 | 110 | 50 | 110 |
| ISQG | 7 | 30 | 124 | 19 | na | 52 |
| PEL | 42 | 112 | 271 | 108 | na | 160 |

(anthropogenic) sources (Zhang et al. 2007). EF values were calculated using the formula given in Zhang et al. (2007):

$$EF = \frac{(Me/Fe)Sample}{(Me/Fe)Background}$$

Heavy metal contents are strongly correlated with $Fe_2O_3$, suggesting that Fe oxides play a major role in controlling abundances (Ahmed et al. 2010). Clay particles often appeared to be coated with Fe (oxy) hydroxides, which can act as carriers of metallic pollutants by absorption, as observed byAhmed et al. (2010) and Galan et al. (2003). Therefore, in this study, $Fe_2O_3$ was used to normalize elemental abundances.

The calculated EF values of As, Pb, Zn, Cu, Ni, and Cr for the Arase dam, Kuma River, and Yatsushiro Bay for the years 2011 and 2013 is given in Tables 1 and 3. The average EF for each year was plotted to show the change from 2002 to 2013 (Fig. 10a, b).

In the Kuma River, EF of Cr and Ni gradually increase from 2002 to 2013 while As, Pb, Zn, and Cu decreased in 2011 and increased in 2013 (Fig. 10a) due to a higher abundance of fine sediments . The EF for Kuma River sediments are less than 1.5 for all elements. We, therefore, conclude that there is no anthropogenic contamination following the classification of Zhang et al. (2007).

The EF values of Yatushiro Bay sediments are not much different from those of the Kuma River sediments (Fig. 10b). Cr increased gradually from 2002 to 2013. Ni increased by 2011, decreased in 2012, and increased in 2013. The levels of As, Zn, Cu, and Pb decreased gradually from 2002 to 2013. Yatsushiro Bay sediments, like those of the Kuma River, have EF values lower than 1.5, implying no anthropogenic contamination, based on Zhang et al. (2007). Although both Kuma River and Yatsushiro Bay sediments have average elemental contents higher than UCC (Table 4), this seems to be due to the background values of Kuma River and Yatsushiro Bay basement geology and not any anthropogenic input.

## Conclusions

In the short cores from 2011, surface and bottom sediments do not seem to represent post-dam removal and dam conditions, respectively, and do not show significant chemical change. This may imply that the beginning of dam removal did not cause a large change in river sediment composition in 2011. However, when compared with 2002 data, elemental composition of the sediment has decreased significantly by 2011 and 2013. The increase in elemental concentration in 2013 compared to 2011 may be influenced by dam removal as well as periodic dam flushing during the winter season. On the other hand, grain size differences in the short core bottom and surface sediments show that opening the dam has flushed most of the fine sediments towards Yatsushiro Bay. The two sediment grain size fractions show clear chemical changes along the stream. We conclude that grain size is a main controlling factor for the chemical composition of Kuma River and Yatsushiro Bay sediments. Grain size data shows progressive fining of sediment moving downstream, correlated with increases in Sr, Zr, and Ti. Sediments are poorly sorted in the river channel. Yatsushiro Bay has high levels of the measured elements in the fine fraction for almost all samples. This indicates that the fine sediments transported by the river are accumulating in the bay. The chemical composition from 2002, 2011, and 2013 shows that elemental contents decreased from 2002 to 2011 and then increased slightly in 2013. The removal of the Arase dam in 2010 and periodic flushing of the upstream dam lake in winter reduced the levels of elements of concern in 2011 and did not cause an increase. The increase of elemental concentration in 2013 seen in both environmental related elements (Pb, Zn, and Cu) and heavy mineral related elements (Zr, Nb, and $TiO_2$) is due to an increase in the fine sediment fraction in the bay area. An EF analysis does not show any anthropogenic contamination in either the river or the bay sediments. Therefore, flushing the dam has led to a recovery of the bay environment. We also conclude that continued environmental monitoring studies are very important for dam management and control.

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