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Atsuyuki Ohta, Noboru Imai, Yoshiko Tachibana & Ken Ikehara

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Application of spatial distribution patterns of multi-elements in geochemical maps for provenance and transfer process of marine sediments in Kyushu, western Japan

Running header: Geochemical maps of sediments in Kyushu

Atsuyuki Ohta1*, Noboru Imai1, Yoshiko Tachibana1, Ken Ikehara1

1 Geological Survey of Japan, National Institute of Advanced Industrial Science and Technology, Central 7, Tsukuba, Ibaraki 305-8568 Japan

*Correspondence: (a.ohta@aist.go.jp)

Keywords: marine sediment, stream sediment, geochemical map, particle transfer, oceanic current, pyroclastic flow deposit

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Abstract

We investigated the origin of marine sediments and their transfer by water currents using the spatial distribution patterns of multi-elements in terrestrial and marine areas of Kyushu, western Japan. Quaternary volcanic material covers Cretaceous granitic rock and Jurassic–Paleogene sedimentary rock of an accretionary complex in this region. Cluster analysis based on chemical compositions identified the origin of marine sediments from stream sediments originating from the above lithologies. Pyroclastic flow deposits associated with caldera formation, particularly that of the Kikai caldera (7.3 ka), were characterized by low Cr/Ti and La/Yb ratios. In contrast, the La/Yb ratio was very high in sediments derived from granitic rock and sedimentary rock of the accretionary complex. The spatial distributions of low Cr/Ti and La/Yb ratios suggest that marine sediments containing pyroclastic materials, which are found within an 80-km radius of the Kikai caldera, were distributed on the shelf and transported northeastward by a branch of the Kuroshio Current. The continuous distribution of the medium Cr/Ti and high La/Yb ratios from the land to the coast, slope, and deep basin on the Pacific Ocean side suggests that sediments supplied from the terrestrial area were transferred by gravitational transport from the shelf to the deep basin.
Introduction

The Geological Survey of Japan, National Institute of Advanced Industrial Science and Technology (AIST), has created geochemical maps showing the spatial distribution patterns of the concentrations of multiple elements throughout the country based on stream and marine sediments (Imai et al. 2004; Imai et al. 2010). These maps provide fundamental information on the behavior of the elements in nature and are widely used for environmental assessment and mineral exploration (e.g., Webb et al. 1978; Darnley et al. 1995). Several sub-continental and cross-border geochemical mapping projects have been conducted recently to further nationwide geochemical mapping (Xie & Chen 2001; Salminen et al. 2005; De Vos et al. 2006; De Caritat & Cooper 2011; Reimann et al. 2014b, a; Smith et al. 2014).

Japanese geochemical mapping was initially designed to provide natural background concentrations of elements to quantitatively elucidate anthropogenic contamination in urban regions and coastal sea zones (Imai et al. 2004; Imai et al. 2010). Nonetheless, the spatial distribution patterns of elements across land and marine areas may be further applicable to the study of the particle transfer process from land to the coastal sea as well as in a marine environment. Although Japan is surrounded by oceans, we still have little information about the characteristics of the surface sediment on shelves, slopes, and basins in the island arc region in terms of the erosion, transport, and deposition of the sediments. Thus, we have applied geochemical maps to the provenance and transfer analyses of marine sediments (Ohta & Imai 2011) to objectively reveal the complex factors affecting the spatial distributions of elements in marine sediments using analysis of variance (ANOVA), cluster analysis, and Mahalanobis’ generalized distances (Ohta et al. 2007; Ohta et al. 2010; Ohta et al. 2013; Ohta et al. 2017b).

We successfully identified the long-range transport process of modern silty sediments on the shelf and slope by oceanic and tidal currents using Cr and Ni geochemical maps (Ohta et al. 2007; Ohta et al. 2010; Ohta et al. 2017a). Silty sediments originated from ultramafic rocks can be easily isolated from those having different origin because the rocks extremely elevate the Cr and Ni concentrations of stream and marine sediments. In contrast, granule and sandy sediment on the shelf are mainly composed of sediments that formed during the Quaternary regression and transgression cycles (e.g., Park & Yoo 1988; Ikehara 1993); most of them formed under hydrographic conditions that are different from the modern environment. It is therefore difficult to directly identify their origin, evaluate the corresponding relationship between marine and terrestrial materials, and examine transportation process of sandy sediments (Ohta et al. 2007; Ohta et al. 2010).

Thus, the Kyushu region, southwestern Japan, was selected as a study area in which sandy sediments containing pyroclastic deposits supplied by large-scale eruptions from the Kikai caldera (7300 years ago) (Maeno & Taniguchi 2007; Fujihara & Suzuki-Kamata 2013) are widely distributed in the southern marine area of Kyushu Island (Ikehara 2014) (Fig. 1). In this region, there are various bedforms such as subaqueous sand dunes, sand ribbons, and ripples (Ikehara 1988, 1993; Ikehara & Kinoshita 1994). Shelf sand bodies with these bedforms are presumed to have formed during the transgression after the last glacial...
maximum and are also presently active owing to the branch currents of the Kuroshio Current (Fig. 1). Therefore, the Kyushu region is the suitable place to apply our geochemical datasets for finding the range over which the coarse pyroclastic deposit moved, or the boundary separating pyroclastic deposit from the sandy sediments derived from terrestrial area without volcanic materials using geochemical data of sediments. In this study, we elucidate the following three items in particular using marine and terrestrial geochemical maps: (1) the relationship between marine sediments and adjacent terrestrial materials, (2) the dispersion processes of volcanic materials erupted by very large eruptions, and (3) and the transportation of granules and sandy sediments by water currents.

**Study area**

*Marine topography and hydrographic condition*

Figure 1 shows the geographical names of the features within the study area as well as a geological map of the area. The water depth in the northern part of the study area is generally less than 200 m. The Goto Nada, Tsushima Strait, and Genkai Nada in the northwestern part are continental shelf areas, where "Nada" means an open sea where a strong ocean current, fast tidal current, or severe wave makes marine navigation challenging. The Suo Nada is in the western part of the Seto Inland Sea and is generally less than 60 m deep. It passes through the Bungo–Suido Strait to the Pacific side of Kyushu Island (Hyuga Nada). The Hyuga Nada is characterized by a narrow shelf with a depth that increases steeply toward the southeast. Submarine landslide surfaces are found along the slope of the Hyuga Nada (Ikehara 2000). The Amakusa and Yatsushiro seas, the largest inner bays of Kyushu mainland, have extreme tidal ranges and connect to the Amakusa Nada. The water depth of the Amakusa Nada gently increases toward the southwest from the southwest area of Kyushu mainland and connects to the East China Sea. Kagoshima Bay, with a maximum water depth of 80 m, is part of the Aira Caldera and is associated with thermal activity. The Osumi Strait is a marine area between the Osumi Peninsula and the islands of Yakushima and Tanegashima, where the water depth is less than 500 m. The southeast area off Tanegashima is a steep slope associated with a submarine canyon. The strong oceanic currents, which are branches of the Kuroshio Current, prevail on the coast of Kyushu mainland. The Kuroshio Current flows from the southwest to northeast at the southwest areas of the Yakushima and Tanegashima islands. A branch of the Kuroshio Current also flows through the Tsushima and Osumi straits from the southwest to northeast.

*Marine sedimentology*

Most of the marine sediments in the study area are mainly from the Holocene and Pleistocene. Coarse sand and gravel from the Goto Nada, Tsushima Strait, Genkai Nada, Bungo–Suido Channel, and Osumi Strait are composed of sediments that formed during Quaternary regression and transgression cycles (Ohshima et al. 1975; Ohshima et al. 1982; Ikehara 2000, 2013, 2014). Bedforms such as ripples and current lineations have been found in the Tsushima Strait, Genkai Nada, Osumi Strait, and northeast of Tanegashima Island; these can be explained by changes in sea level after the last glacial maximum and a strong...
bottom current related to the oceanic current that moves surface sediments (Ikehara 1988; Ikehara 1992; Ikehara 1993; Nishida & Ikehara 2013). Submarine topography and bedforms in the Bungo–Suido Channel are influenced by post-glacial sea level changes and related changes with strong tidal energies (Ikehara 1998). Silt and clay found in the depths of the Hyuga Nada and in the slope and basin off Tanegashima Island are hemipelagic deposits. Modern silty sediments derived from the adjacent terrestrial area are found in the shallows of inner bays such as the Ariake and Yatsushiro seas, the Suo Nada, and nearshore regions. The shelf and upper slope of the Hyuga Nada are covered by sandy deposits, with some turbidites are intercalated in the slope sediments (Ikehara 2000).

**Terrestrial area and riverine system**

Mainland Kyushu is mountainous and comprises many active volcanoes such as Kujusanzan, Asosan, Unzendake, Kirishimayama, Sakurajima, and Satsuma–Iojima (see small triangles in Fig. 1). It is also associated with many isolated islands such as Iki, Hirado, Fukue, Nakadori, Amakusa, Tanegashima, and Yakushima, and its major rivers include the Chikugo, Kuma, Gokase, Oyodo, and Sendai. The sediment yield, drainage basin area, and river water discharge of the major rivers in the study area are summarized in Supplementary Table 1. The sediment yield of each river system is calculated using the drainage basin area and the average rate of erosion in each river basin (after Akimoto et al. 2009). In the current environment, rivers on Kyushu Island supply 49% of the total sediment yield to the western region, including Ariake Sea, Yatsushiro Sea, and Amakusa Nada (Supplementary Table 1). The Hyuga Nada and Suo Nada receive sediment discharges of 29% and 17% from Kyushu Island, respectively (Supplementary Table 1). Little sediment is discharged from the rivers on Kyushu Island to the northern regions of the Genkai Nada, Goto Nada, and Tsushima Strait or to the southern region including the Osumi Strait and Kagoshima Bay (Supplementary Table 1).

**Terrestrial geology and metalliferous deposits**

Figure 2 shows a geological map of southwest Japan that has been simplified from the Geological Map of Japan 1:1 000 000 scale (Geological Survey of Japan AIST (ed.) 1992). The details of the geology of the Kyushu region have been provided by Miyazaki et al. (2016). A Permian accretionary complex is located in the northeast; Jurassic, Cretaceous, and Paleogene accretionary complexes are located in the central and southeast parts of the study area. The Paleogene unit is also found in the Tanegashima and Yakushima islands. These accretionary complexes are exposed across 23% of the study area. The Permian unit mainly consists of mudstone and sandstone associated with blocks of limestone, basaltic rock, and chert. The Jurassic unit is associated with exotic rocks such as serpentinite mélange, metamorphic rock, and ultramafic rock. The Cretaceous and Paleogene units consist mainly of accreted turbidites (mudstone to sandstone) associated with exotic blocks of basaltic rock. Carboniferous–Permian and Triassic–Jurassic high-pressure-type metamorphic rock is distributed in the north. These metamorphic rocks are dominated by pelitic, mafic, and psammitic schists; and their exposed area is less than 4% of the total. Cretaceous high-temperature-type metamorphic rock is found in the central region, although the area of
distribution is only 0.5% of the area. Cretaceous granitic rock is intruded in the northern region and comprises only 5% of the area. In contrast, Neogene granitic rock is intruded in the Osumi Peninsula and island of Yakushima and outcrops sporadically in accretionary complexes in 3% of the area.

Neogene–Quaternary basaltic, andesitic, and dacitic volcanic rocks are distributed in both the northern and southern regions of Kyushu; they outcrop across 22% of the area. They are associated with huge pyroclastic flow deposits, debris, and tephra that were erupted by large-scale caldera-forming processes; they comprise only 17% of the area. In particular, Aso and Ito pyroclastic rocks erupted from the Aso and Aira calderas mainly about 90 ky and 29 ky ago, respectively; they are distributed widely in the central and the southern parts, respectively (Nakada et al. 2016). The Kikai caldera to the south of the Kyushu mainland was formed by large-scale violent eruptions 7.3 ky ago. Koya pyroclastic flows from the Kikai caldera were extended to 40–80 km (Maeno & Taniguchi 2007; Fujihara & Suzuki-Kamata 2013). Asosan, Sakurajima, and Satsuma–Iojima are volcanoes associated with the Aso, Aira, and Kikai calderas, respectively (see triangular symbols in Fig. 1).

Pre-Cretaceous non-accretionary sedimentary rocks are found in the central part with an their exposure of less than 3% across the total area. Paleogene forearc sedimentary rocks with coal beds are distributed in the northern and northwestern regions of the Kyushu mainland and in the Amakusa islands (5% of the area). Neogene and Quaternary unconsolidated sediments are distributed widely in the downstream basin of the Chikugo and Oyodo rivers but are rather restricted to a small area elsewhere; they comprise 17% of the study area.

Figure 2 also shows the distribution of major and economically mined deposits in Kyushu (Sudo et al. 2003). Many Au and Ag mineral deposits are hydrothermal or disseminated and are closely related to Neogene and Quaternary volcanic activity. Some skarn deposits bearing Cu, Zn, As, and Sn were formed in Paleozoic limestone bedrocks of the Jurassic accretionary complex where the Neogene granitic rocks intruded.

Materials and methods

Samples, sampling methods, and processing

Figure 3 shows the sampling location of the Kyushu region. For a geochemical mapping project carried out during 1999–2004 by the Geological Survey of Japan, AIST, 366 stream sediment samples were collected from locations on Kyushu Island (Imai et al. 2004) (Fig. 3). In addition, 191 stream sediment samples were collected from 23 isolated small islands around the Kyushu mainland from 2013 to 2014 for a high-density geochemical map (Ohta 2018); of these, 25 samples having wide watershed areas were used for this study (Supplementary Information). The collected stream sediments were dried in air and were sieved through a 180 μm screen. Magnetic minerals contained in samples were removed by using a magnet to minimize the effect of magnetic mineral accumulation (Imai et al. 2004; Ohta 2018).
A total of 504 marine sediment samples were collected from the marine areas around the Kyushu mainland during cruises GH83-1, GH84-1, and GH85-2 in 1983, 1984, and 1985, respectively (Ikehara 2000, 2001, 2013, 2014). In addition, 102 marine sediment samples were collected from the Tsushima Strait, Seto Inland Sea, Ariake and Yatsushiro seas, and Amakusa Nada in 2005 and from Kagoshima Bay in 2007 (Imai et al. 2010). These 606 samples were collected using a Kinoshita-type grab sampler (RIGO Inc. Co.), and the uppermost 3 cm layer of each sediment sample was separated, air-dried, ground with an agate mortar and pestle, and retained for chemical analysis.

The particle sizes of 334 marine sediments were determined on the basis of the median particle diameter of the surface sediments (Ikehara 2013, 2014). The median particle diameter was not measured for the remaining 272 samples; their classification is based on a visual inspection of texture. The elemental concentrations in the marine sediments change with variations in particle sizes. As a general rule, CaO, MnO, T-Fe$_2$O$_3$, Sr, and Co are high in coarse and medium sand because these soils contain calcareous materials and coatings of Fe-Mn oxides; the trance elements are high in silt and clay because of less effective dilution effect in the region (Ohta et al. 2010; Ohta & Imai 2011). Thus, marine sediments were roughly classified as coarse sediment comprising pebbles, granules, and very coarse–coarse–medium sands; fine sands comprising very fine and fine sand; and silt and clay consisting of coarse silt, fine silt, and clay (Fig. 3).

**Geochemical analysis**

We compiled the geochemical data of 53 elements in 391 stream sediments and 606 marine sediments around the Kyushu region that have been reported by Imai et al. (2004); Imai et al. (2010); and Ohta (2018). The details of the digestion of the samples, measurement of elemental concentrations, and quality control of measurement values are described in the above cited references. Each sample was digested using HF, HNO$_3$, and HClO$_4$. The concentrations of Na$_2$O, MgO, Al$_2$O$_3$, P$_2$O$_5$, K$_2$O, CaO, TiO$_2$, MnO, total Fe$_2$O$_3$, V, Sr, and Ba were determined using inductively coupled plasma atomic emission spectrometry (ICP–AES), while those of Li, Be, Sc, Cr, Co, Ni, Cu, Zn, Ga, Rb, Y, Nb, Zr, Mo, Cd, Sn, Sb, Cs, the lanthanides (Ln: La–Lu), Ta, Hf, Tl, Pb, Bi, Th, and U were determined by using ICP–mass spectrometry (ICP–MS) (Imai et al. 2004; Imai et al. 2010; Ohta 2018). The As and Hg analyses of stream sediment samples from the Kyushu mainland were subcontracted to ALS Chemex in Vancouver, BC (Imai et al. 2004). The As concentrations in the stream sediment samples collected from isolated islands were determined using ICP–MS after the digestion of samples using HF, HNO$_3$, HClO$_4$, and KMnO$_4$ (Ohta 2018). The Hg concentrations in stream sediments on isolated islands and marine sediments were determined by using an atomic absorption spectrometer equipped with the direct thermal decomposition of a sample (Imai et al. 2010; Ohta 2018). A summary of the geochemical characteristics of the marine and stream sediments in the Kyushu region is given in Table 1. However, the Zr and Hf concentrations were not used in this study because the heavy mineral fraction, especially that of zircon, was not digested by the HF–HNO$_3$–HClO$_4$ solution. Additionally, the Na$_2$O concentration of marine sediments was also used as a guide because such sediments were not desalinated in this study.
Geochemical map preparation

Geochemical maps of the Kyushu mainland have been re-created as mesh maps associated with the data of the surrounding isolated islands. They were created by using geographic information system software (ArcGIS 10.5; Environmental Systems Research Institute (ESRI) Japan Corporation, Tokyo, Japan) after Ohta et al. (2004). The stream sediment is considered a composite sample of the products of weathering and soil and rock erosion in the watershed area upstream of the sampling site (Howarth & Thornton 1983). A sampling site is presumed to express the average chemical concentrations in a drainage basin. The watershed area for each sample was calculated in ArcGIS based on a digital elevation model (50 m mesh data) obtained from the Geospatial Information Authority of Japan. The marine geochemical maps were created by interpolating data points using radial basis functions (Aguilar et al. 2005). The resultant marine geochemical maps were combined with the existing land geochemical maps as shown in Figs. 4a–4l. The elemental concentrations were classified separately for the stream and marine sediments because their chemical and mineralogical compositions differ significantly, as detailed by Ohta et al. (2004) and Ohta et al. (2010). The following percentile ranges were used for classification of the elemental concentration intervals in the color image maps: $0 \leq x \leq 5$, $5 < x \leq 10$, $10 < x \leq 25$, $25 < x \leq 50$, $50 < x \leq 75$, $75 < x \leq 90$, $90 < x \leq 95$, and $95 < x \leq 100\%$, where $x$ represents the elemental concentration according to Reimann (2005).

This class selection is advantageous in that the same range of percentiles (e.g., 90–95%) implies the same statistical weight even at different numerical scales (Reimann 2005).

Results

Spatial distribution patterns of concentrations of multi-elements in the terrestrial area

Geochemical maps of the 12 elements K$_2$O, CaO, MnO, TiO$_2$, Cr, Cs, La, Yb, Cu, Cd, Sb, and Pb are shown in Figs. 4a–4l. They are representative and characteristic distribution patterns of elements that were obtained by considering the similarity of the spatial distribution patterns of 49 elements across land and sea. The Li, K$_2$O, Rb, Cs, and Tl contents are high in the northern, central, and southeastern parts of Kyushu mainland and in Yakushima Island, where accretionary complex, granitic rock, and felsic volcanic rock crop out. However, the concentrations are low in the central part of Kyushu, where mafic volcanic rock, pyroclastic rock, and metamorphic rock are distributed (K$_2$O and Cs, Fig. 4). The spatial distributions of MgO, CaO, TiO$_2$, MnO, T–Fe$_2$O$_3$, Sc, V, Co, and Sr are opposite those of Li, K$_2$O, Rb, Cs, and Tl (CaO and T–Fe$_2$O$_3$, Fig. 4). Their high concentration areas correspond mainly to areas covered by the mafic volcanic rocks and pyroclastic rocks, particularly near Aso Volcano (Fig. 2). The Cr and Ni concentrations are high in the northern–central and central parts of Kyushu mainland. High-pressure-type metamorphic rock and mafic rocks of the accretionary complex crop out sporadically in these areas (Cr, Fig. 4). High enrichment of Be, Na$_2$O, rare earth elements (REEs), Nb, Ta, Th, and U was found in the northern and southern parts of the islands of Kyushu and Yakushima, which are underlain by granitic rocks (La and Yb, Fig. 4). In addition, Y and heavy REEs such as Gd, Tb, Dy, Ho, Er, Tm, Yb, and Lu were also enriched in the pyroclastic rock outcrops of central
and southern Kyushu Island (Yb, Fig. 4). The metalliferous skarn-type deposits bearing Cu, Zn, As, and Sn (circle with a dot and star symbols, Fig. 4) caused extremely high concentrations of Cu, Zn, As, Mo, Cd, Sn, Sb, Hg, Pb, and Bi (Cu, Cd, Sb, and Pb, Fig. 4). However, the Au–Ag deposits (diamond symbols, Fig. 4) did not appear to contribute to the enrichment of these elements.

**Spatial distribution patterns of multi-elements in the marine environment**

Silt and clay sediments in the Suo and Hyuga nadas, southeastern Osumi Strait, and Yatsushiro Bay are commonly enriched in Li, Be, Cr, Ni, Cu, Nb, Mo, Cd, Sn, Sb, Cs, Ta, Hg, Tl, Pb, Bi, and U. Silt and clay sediments in the basin southeast of Tanegashima Island have high concentrations of Be, K₂O, Cr, MnO, Ni, Cu, As, Rb, Mo, Cd, Sn, Sb, Cs, Ba, light REEs, Hg, Tl, Pb, Bi, and Th. The enrichment of MnO, Ni, Cu, Mo, Cd, Sn, Sb, Pb, and Bi in the deep seas was caused by early diagenetic processes under a static depositional environment; their concentrations have a definite correlation with the MnO concentration (e.g., Klinkhammer 1980; Shaw et al. 1990).

The coarse sediments and fine sands in the Tsushima Strait, Genkai Nada, and Goto Nada are rich in CaO and Sr, and poor in other elements as a result of calcareous deposits such as shell fragments and foraminifera tests. The coarse and fine sands in the Amakusa Nada are rather enriched in MgO, Cr, and Co, although their chemical compositions are similar to those in the Tsushima Strait, Genkai Nada, and Goto Nada. The sandy sediments in the southern part of the Suo Nada near the Kunisaki Peninsula are enriched in MgO, CaO, TiO₂, T–Fe₂O₃, Sc, V, Co, and Sr and are poor in K₂O, Rb, and Cs; the sediments in the northern part of the Suo Nada are in contrast to those near Kunisaki Peninsula.

The fine sands in the Hyuga Nada are rich in Li, Be, K₂O, Cr, Ni, Rb, Ba, light REEs, and Nb, and poor in P₂O₅. CaO, MnO, Sr, heavy REEs, and Cd. High concentrations of As, Sn, Hg, Pb, and Bi, in particular, were found in samples collected within a 16 km radius of the mouth of the Gokasegawa River, where upper river basin skarn-type deposits bearing Cu, Zn, As, and Sn occur. Granules and coarse sands in the Osumi Strait are highly enriched in MgO, P₂O₅, TiO₂, MnO, T–Fe₂O₃, Sc, V, Co, Zn, Sr, Mo, Cd, and heavy REEs. Marine sediments occurring between the Osumi Peninsula and Tanegashima Island are, in particular, enriched in MgO, P₂O₅, TiO₂, MnO, T–Fe₂O₃, Sc, V, and Co; however, these elements are less abundant in stream sediments derived from the adjacent terrestrial area in which unconsolidated sediment and sedimentary rock, granitic rocks, and sedimentary rocks of accretionary complexes are the dominant lithologies. In contrast, the fine sands of the Osumi Strait are more enriched in CaO and Sr, and are poorer in MgO, TiO₂, V, T–Fe₂O₃, Co, and Zn than the coarse sands owing to dilution by biogenic carbonate materials. The sediments in Kagoshima Bay have chemical compositions similar to those of the coarse sediments in the Osumi Strait irrespective of particle size; furthermore, they are highly enriched in As, Cd, Mo, Sb, Hg, and Bi owing to the hydrothermal activity (Sakamoto 1985). Almost all the elements are less abundant in the fine sands of the slope to the southeast of Tanegashima Island.
Discussion

Differences in elemental concentrations of stream sediments grouped by dominant lithology distributed in the watershed of stream sediment samples

The geochemistry of stream sediment is determined predominantly by the dominant lithology distributed in the catchment area, as we have previously suggested (e.g., Ohta et al. 2004). As a first approximation, we assume that when a specific rock type underlies more than half of the area of the river basin, it is representative of the geology of the watershed and mainly controls the chemical composition of the stream sediment (Ohta et al. 2004). Unconsolidated sediment and sedimentary rock (Sed); an accretionary complex comprising mainly sandstone, mudstone, and a mélangé matrix (Acc); granitic rock (Gr); felsic volcanic rock (Fv); mafic volcanic rock (Mv); a pyroclastic flow deposit (Py); and high-pressure-type metamorphic rock (Mp) were considered as typical lithologies in the study area. When no specific rock type extensively covered a catchment basin, the sample was classified as other (Oth). Table 2 shows the median of the elemental concentration of stream sediment classified to the parent lithology.

An analysis of variance (ANOVA) and Bonferroni’s multiple comparison test were applied to the geochemical data sets to objectively examine the influence of lithology on the chemical compositions of stream sediments (Hochberg & Tamhane 1987; Nagata & Yoshida 1997; Miller & Miller 2010). These statistical tests assume that the data follow a normal distribution. However, the data distribution for each elemental concentration followed neither a normal nor a log-normal distribution when the Shapiro–Wilk test was applied to our data set (Shapiro & Wilk 1965). In such cases, approaching data symmetry is the second-best alternative. Accordingly, data distribution with a skewness closer to zero, which indicated symmetric distribution, was used for the statistical tests (Ohta et al. 2005). The concentrations of the 42 elements were transformed into common logarithms; those of Al$_2$O$_3$, Ga, Rb, Er, Tm, Yb, and were unchanged for the statistical tests (Table 3).

The null hypothesis of the one-way ANOVA is that no significant difference exists in elemental concentrations among seven categories: Sed, Acc, Gr, Fv, Mv, Py, and Mp. Table 3 shows the significance probability (p) and effect size ($\eta^2$) of the one-way ANOVA analysis. The effect size ($\eta^2$) was used to form a plausible estimation of the p value irrespective of sample number because a statistical test with a large amount of input data is highly sensitive to very small differences (Richardson 2011; Fritz et al. 2012). In this study, we concluded that the null hypothesis was rejected for the elemental concentration data with $p < 0.01$ and $\eta^2 > 0.14$. Table 4 shows that the null hypothesis was rejected for all elements except Cu, Zn, As, Cd, Sn, Hg, and Pb, which indicates that most elements are significantly influenced by the parent lithology. The concentrations of these elements were influenced by metalliferous deposits rather than by a specific lithology, as described above.

Next, we examined whether the parent lithology significantly affect the concentrations of elements except Cu, Zn, As, Cd, Sn, Hg, and Pb in the stream sediment using a Bonferroni test at the 0.01 confidence level (Hochberg & Tamhane 1987). Table 4 presents the results of
direct pairwise comparisons. The elements are grouped as felsic element enrichment in rhyolitic–dacitic and granitic rocks, mafic element enrichment in andesite–basaltic and gabbroic rocks, and sulfurous elements concentrated in a sulfide deposit for descriptive purposes. The stream sediments derived from Acc are highly rich in K$_2$O, Li, Rb, Cs, Ba, and Tl compared with those from Mv, Py, and Mp. Moreover, they are poor in Na$_2$O, P$_2$O$_5$, CaO, Sc, V, TiO$_2$, MnO, T–Fe$_2$O$_3$, Sr, Nb, Eu, and heavy REEs (Gd–Lu) compared with those from Fv, Mv, Py, and Gr. The sediments derived from Mv and Py are significantly richer in MgO, CaO, Sc, TiO$_2$, V, T–Fe$_2$O$_3$, Co, and Sr than those from Sed, Acc, and Gr and are significantly poorer in alkali metal elements, Nb, Ba, Tl, Th, and U than those from Sed, Acc, and Gr. Although the stream sediments derived from Py have chemical compositions similar to those from Mv, they are significantly poorer in Cr and Ni. The stream sediments derived from Gr show more enrichment in alkali metal elements, Be, Ga, REEs, Ta, Th, and U than the other samples, although they are poor in Cr and Ni. The sediments derived from Mp are highly enriched in Cr and Ni and are poorer than the other samples in K$_2$O, Nb, and Ta. The samples derived from Fv have few systematic geochemical features compared with those of other samples. These results are consistent with the visual interpretations of the spatial distribution patterns of elemental concentrations (Figs. 4a–4l).

In terms of the sulfurous elements, the samples derived from Fv are significantly more enriched in Sb than those from Mv, Py, and Gr; the sediments from Gr are richer in Bi than those from Sed and Mv; and the sediments from Mv and Py are richer in Mo and Bi than those from Sed and Gr. The hydrothermal activity associated with Mv, Fv, and Gr may have enhanced the concentrations of the sulfurous elements. However, the spatial distributions of the major hydrothermal-type deposits do not appear to be comparable to areas in which sulfurous elements are highly concentrated (Cd and Sb, Fig. 4).

**Differences in elemental concentrations in coastal sea sediments by region and particle sizes**

We examined the supplementary process of clastic deposits from terrestrial to marine environments based on the chemical compositions of stream and marine sediments. Those of the marine sediments were influenced by various factors: (1) particle transport from the land to the sea, (2) the dilution effect of quartz and biogenic calcareous materials; (3) increases in the concentrations of alkali metal ions in silty and clayey sediments; (4) transportation of sediments by gravity flow or oceanic currents; (5) early diagenetic processes in the deep sea; (6) denudation or re-sedimentation of basement rocks; (7) and contamination from human activity (Ohta et al. 2010; Ohta et al. 2017a). Thus, we used a step-by-step approach.

The chemical compositions of marine sediments differ among regions because they originate from adjacent terrestrial materials. Some marine sediments are formed by the denudation or re-sedimentation of basement rocks, which may belong to any of the lithologies in the adjacent terrestrial area. This factor is referred to herein as a regional difference. Additionally, the elemental concentrations in the marine sediments vary with particle size, caused by the dilution of large amounts of biogenic calcareous materials in the coarse grains and by an increase in clay minerals in the fine grains. This factor is referred to herein as the particle size effect. These two effects are common in all marine sediments.
The marine sediments were grouped as those from the Tsushima Strait and Goto Nada, those from the Suo Nada, those from the Amakusa Nada including the Ariake and Yatsuhiro seas, those from the Hyuga Nada, and those from the Osumi Strait; the zonal classification is given in Fig. 3. They were further classified into coarse sediments, fine sands, and silt–clay. The median concentration of elements in the marine sediments grouped by each region and particle size were also calculated (Table 5). The silty sediments of Kagoshima Bay were separated from the silts and clays of the Osumi Strait because there are clear differences in their chemical compositions.

A two-way ANOVA test was applied to identify the factors that significantly change the chemical compositions of marine sediments. Table 6 presents the variance ratios (F), probabilities (p), and effect size ($\eta^2$) owing to the regional effect (factor A), particle size (factor B), and the interaction effect (factor A x B). The interaction effect (A x B) refers to the effect that one factor has on another (Miller & Miller 2010). When the estimated probability (p) was lower than 0.01 and the effect size ($\eta^2$) was larger than 0.14, we concluded that the factor made a significant difference in the chemical composition (Table 4). The ANOVA results suggest that the chemical compositions of 29 elements in marine sediments change significantly among different regions. The regional difference effect was not significant for $K_2O$, Ni, Rb, Sb, Cs, Ba, Hg, Bi, and U, whose concentrations were determined simply by particle size (factor B). The results suggest that these elements cannot be used to understand the influence of terrestrial materials on coastal sea sediments. The concentrations of 17 elements in the marine sediments are strongly controlled by the particle effect; however, 15 elements such as Cr and La are also significantly influenced by the regional effect. Therefore, it is more effective to subdivide marine sediments by regions and particle sizes to evaluate their geochemical similarities and differences.

Identification of probable source materials for marine sediments using cluster analysis

We identified the probable source materials for the marine sediments by using cluster analysis with stream sediments grouped by the dominant lithology. The compositional data of the marine and stream sediments need to be standardized or transformed appropriately to examine the similarity and differences in their chemical compositions when using cluster analysis (Templ et al. 2008). Although various transformation procedures for compositional data have been proposed by Aitchison (1982), Aitchison et al. (2000), and Pawlowsky-Glahn & Egozcue (2006), Ohta et al. (2017a) proposed a log transformation of the enrichment factor (EF) for geochemical data sets using following equation:

$$\log EF = \text{alr}(C)_{\text{sample}} - \text{alr}(C)_{\text{UCC}},$$

where EF is the enrichment factor for the upper continental crust (UCC) components (Taylor & McLennan 1995), and alr(C) represents the additive log-ratio transformation $\log([C]/[Al_2O_3])$. The logarithmic EF is a normalized value for all elements having respective different ranges in concentration and is also effective in removing the dilution effect (Ohta et al. 2013).
Figure 5 shows dendrograms expressing the distances between data sets determined using the Ward method (Ward Jr 1963). The data set excludes sea salt (Na$_2$O), biogenic carbonate materials (CaO and Sr), and elements of K$_2$O, Ni, Rb, Cs, Ba, and U that are not significantly impacted by the regional effect (Table 6). Furthermore, heavy metals related to mining and anthropogenic activities—including Cu, Zn, As, Mo, Cd, Sn, Sb, Hg, Pb, and Bi—were also excluded from the analysis because their concentrations do not reflect the geochemistry of the parent lithology (Table 4). Figure 5a uses a data set of the above 31 elements. Figure 5b uses a data set of immobile elements including Sc, TiO$_2$, Cr, Nb, Y, Ln, Ta, and Th, which are not strongly influenced by the weathering process. Coarse sediments and fine sands in the Osumi Strait and silt in Kagoshima Bay, which are commonly observed features in both Figs. 4a and 4b, were plotted in the same group as stream sediments derived from Py. The fine sands in the Hyuga and Amakusa nadas were clustered with stream sediments derived from Acc. The silt–clay sediments in the Amakusa and Hyuga nadas and the Osumi Strait were plotted in the same group and were related, remotely, to stream sediments. The marine sediments in the Tsushima Strait and Goto and Suo nadas were also remotely related to stream sediments. Unfortunately, all the source materials of the marine sediments could not be identified using cluster analysis.

**Discrimination study of stream and marine sediments using Cr/Ti and La/Yb ratios**

Finally, we visualized the particle transfer process in marine environments by using the chemical compositions of the sediments. The concentration ratios of immobile elements, such as Ti/Zr, La/Sc, Th/Sc, Cr/Th, were used as indices of the source rock composition of sandstone and mudstone (e.g., Roser 2000). Simultaneously, the Ti/Nb, Cr/Th, and La/Yb ratios were obtained for the provenance indicators for silt- and clay-sized marine sediments in the Yellow Sea (e.g., Yang et al. 2003). Although many combinations are possible for the provenance analysis, combinations of elemental concentration ratios effective in identifying contribution of source components are used (e.g., Roser 2000).

The cluster analysis suggested that pyroclastic rocks and accretionary complexes are the parent lithologies significantly affecting the surrounding marine sediments (Figs. 4a and 4b). As we discussed in the previous session, an ANOVA and multiple comparison tests suggested that stream sediments derived from Py have similar chemical compositions to those from Mv, which have significantly high concentrations of mafic elements such as TiO$_2$ and Fe$_2$O$_3$; nevertheless, Cr and Ni are significantly enriched in sediments from Mp but are extremely poor in those from Py (Tables 2 and 4). Therefore, the concentration ratio of immobile elements of Cr and Ti is likely useful for distinguishing sediments originating from Py, Mv, and Mp. The alkaline metal elements Ba and Tl are significantly enriched in stream sediments derived from Acc; however, their concentrations in marine sediments are predominantly determined by the particle size effect rather than the regional effect (Table 6). Heavy REEs are significantly poor in stream sediments derived from Acc and are highly enriched in those from Py and Mv. Therefore, the La/Yb ratio can be effective for highlighting the influence of sediments derived from Acc.
Figures 5a and 5b present scatter diagrams that plot the Cr/Ti and La/Yb ratios of stream sediments classified by the dominant lithology. The Cr/Ti ratios of stream sediments derived from Mp varied significantly, as did the La/Yb ratios of stream sediments derived from Sed and Gr (Fig. 6a). In general, the Cr/Ti ratio was low for stream sediments derived from Py and Fv (< 0.9) and high for sediments from Mp (> 1.4) (Fig. 6b). The La/Yb ratio was high for stream sediments derived from Acc (> 13) and low for sediments from Py (< 10) (Fig. 6b). The stream sediments derived from Mv are characterized by a middle–low La/Yb ratio (< 15) and a wide range in the Cr/Ti ratio (0.5–3.0).

Figure 6c shows a scatter diagram plotting the regionally grouped Cr/Ti and La/Yb ratios of the marine sediments. Sediments from the Hyuga Nada have a high La/Yb ratio (10–20) and an average Cr/Ti ratio (1.0–2.0), which correspond to stream sediments derived from Acc. In contrast, marine sediments with low ratios of both Cr/Ti (<1.0) and La/Yb (<9.0) were collected mostly from the Osumi Strait; these are comparable to Cr/Ti and La/Yb ratios of stream sediments derived from Py. Some samples collected near the Osumi Strait have an average Cr/Ti ratio (1.0–2.0) and an average–high La/Yb ratio (12–20), which are comparable to those of samples from the Hyuga Nada. Furthermore, some of the outliers among the samples from the Osumi Strait have a low Cr/Ti ratio (< 1.0) and a high La/Yb ratio (> 10). Marine sediments from the Tsushima Strait—including the Genkai and Goto nadas—and the Amakusa Nada are characterized as having high ratios of both Cr/Ti (2.0–4.0) and La/Yb (> 10). Sediments collected from the Suo Nada have average ratios for both Cr/Ti (1.0–2.0) and La/Yb (10–13).

**Particle transfer process in marine environment and provenance of marine sediments**

Figure 7 shows the spatial distribution patterns of the Cr/Ti and La/Yb ratios of marine and stream sediments. To visualize the ratios that characterized the dominant lithology, their high and low ratios are highlighted by shaded regions. The ratios for the stream sediments varied widely (Figs. 6a and 6b) owing to the mineralogical and chemical heterogeneities of source rock, the variation in the mineralogical composition of the stream sediment in the riverbed, and the influence of coexistent lithologies except for the dominant one in the watershed. Thus, to identify the specific characteristics of stream sediment classified by the dominant lithology, the threshold values were defined by using their calculated median values (Med) and median absolute deviation (MAD). For this purpose, the MAD/0.6745 is a useful robust estimate of standard deviation (σ') (Miller & Miller 2010). The lower threshold values of the Cr/Ti and La/Yb ratios were defined as 0.814 and 9.67, respectively, which were calculated as Med + σ' of the Cr/Ti and La/Yb ratios in stream sediments derived from Py. In contrast, the higher threshold values of the Cr/Ti and La/Yb ratios were defined as 2.24 and 13.0, respectively, which were calculated as Med + σ' for the Cr/Ti ratios and Med − σ' for the La/Yb ratios in sediments derived from Acc. For reference purposes, these threshold values are also shown in Figs. 6b and 6c.

Figure 7 shows that samples having lower Cr/Ti and La/Yb ratios are distributed in the Osumi Strait, including Kagoshima Bay. The spatial distribution of these samples within a radius of 80 km of the Kikai caldera corresponds to Koya pyroclastic deposits. Samples
having lower Cr/Ti and La/Yb ratios in Fukiage Hama and Kagoshima Bay and off the Satsuma Peninsula may have been supplied by adjacent terrestrial materials, which originated from Ito pyroclastic rocks through a riverine system or by coastal erosion. The shaded region having lower Cr/Ti and La/Yb ratios continues from the area off of the Osumi Peninsula to the southern part of the Hyuga Nada, which is about 170 km from the Kikai Caldera in a straight line (area A, Figs. 7a and 7b). This distribution indicates that coarse and fine sands containing pumice have been conveyed by a strong oceanic current flow, which was reported by Ikehara (1988, 1993). However, part of the sediments from the Osumi Strait might have originated from older pyroclastic rocks supplied mainly from the Aira Caldera that were denudated and moved by sea level fall during the last glacial age (Inouchi, 1981).

In contrast, samples collected from the western and southern regions of the island of Yakushima and from the channel between the Yakushima and Tanegashima islands, which are described as outliers in the Osumi Strait samples (Fig. 6c), have a low Cr/Ti ratio but a high La/Yb ratio (area B, Figs. 7a and 7b). This feature suggests that sediments supplied from Gr in the island of Yakushima prevail in these regions because stream sediments from Gr have a low Cr/Ti ratio and a high La/Yb ratio (Fig. 6b). Furthermore, low Cr/Ti and La/Yb ratios were not found in the sediments from the southeastern region of the island of Tanegashima. Therefore, these islands appear to serve as high barriers for pyroclastic flow deposits from the Kikai caldera (Geshi 2009).

The fine sand and silt distributed in the Hyuga Nada and in the southeastern regions of the island of Tanegashima have middle–high La/Yb ratios and medium Cr/Ti ratios, which correspond to stream sediment derived from Acc (area C, Figs. 7a and 7b). Modern sandy sediments supplied through rivers were found near the coast according to the continuous spatial distribution patterns of high concentrations of heavy metals related to skarn-type deposits (Cu, Sb, and Pb, Fig. 4). Furthermore, most samples from the Hyuga Nada were collected from water depths below the wave base. Therefore, their spatial distribution across the shelf, slope, and basin can be explained by conveyance to deeper areas via gravitational transport including small-scale turbidity currents induced by storm waves and river floods. Indeed, small-scale turbidity currents occur with high frequency several times per year (e.g., Milliman & Kao 2005; Xu et al. 2010). In contrast, submarine landslides or sheet flow induced by subduction-zone earthquakes occur only once every hundred to thousand years. The spatial distributions of areas A and C in Fig. 7 clearly show that sandy sediments originating from Py erupted from the Kikai caldera 7300 years ago (Maeno & Taniguchi 2007; Fujihara & Suzuki-Kamata 2013). These sediments were conveyed by oceanic currents after the last glacial maximum (Ikehara 1992) before overlapping and mixing with surface sediments derived from Acc in the southeastern region of the study area.

The cluster analysis results suggest that silty sediments from the Amakusa Nada including the Ariake and Yatsushiho seas have no close relation to any stream sediment. The silty sediments of the Yatsushiho Sea are modern sediments supplied mainly from the Kuma River, in which the watershed is dominantly covered by sedimentary rock from accretionary complexes associated with metabasalt and ultramafic rock. Therefore, their chemical compositions would be influenced by sediments originating from Acc having high La/Yb
ratios and those from metabasalt and ultramafic rock extremely enriched in MgO, Cr, and Ni (Cr, Fig. 4).

Coarse sediments of the Tsushima Strait, Genkai Nada, and Goto Nada have high Cr/Ti and La/Yb ratios. These regions receive a modern sediment discharge of only 4% from Kyushu Island (Supplementary Table 1). Therefore, their chemical compositions are very loosely related to those of any stream sediment classified by the dominant lithology, as suggested by the cluster analysis (Fig. 5). However, some stream sediments from Amakusa Island were derived from Sed; those of the northern regions of Kyushu Island were derived from Acc; and those of the northwestern regions of Kyushu Island were derived from Mv, Sed, and Mp having comparable Cr/Ti and La/Yb ratios (Figs. 6a, 6b, and 7). These sample sites are adjacent to the Amakusa Nada, Goto Nada, Tsushima Strait, and Genkai Nada. Park & Yoo (1988) used seismic profile data to determine that most of the Tsushima Strait was exposed and eroded during the last glacial age. Nishida & Ikehara (2013) reported that the evolution of the depositional processes in the Genkai Nada was caused by sea level change after the last glacial maximum and inflow of the Tsushima Warm Current, a branch of the Kuroshio Current, into the Sea of Japan through the Tsushima Strait. Accordingly, we simply conclude that the sedimentary layers in the Tsushima Strait originated as parent rock having high Cr/Ti and La/Yb ratios, as is the case with silty sediments in the Yatsushiro Sea, and were denuded, conveyed by the strong oceanic current, and deposited at each stage during the transgression age after the last glacial maximum.

Summary

Geochemical data of 53 elements in 391 stream sediment samples and 606 marine sediment samples were used to study the provenance and transfer process of coarse and fine sands around Kyushu region, western Japan. The chemical compositions of the stream sediments were determined by the lithology dominantly distributed in the watershed area. One-way ANOVA and a multiple comparison test revealed that 1) stream sediments derived from sedimentary rocks of accretionary complexes are rich in K$_2$O, Li, Rb, Cs, Ba, and Tl; 2) those from mafic volcanic rocks and pyroclastic rocks have a high abundance of MgO, P$_2$O$_5$, CaO, Sc, V, TiO$_2$, MnO, T–Fe$_2$O$_3$, Co, Sr, and heavy REEs (Gd–Lu); 3) those from granitic rocks are enriched in alkali metal elements, Be, Ga, REEs, Ta, Th, and U; and 4) skarn-type deposits have extremely elevated concentrations of Cu, Zn, As, Mo, Cd, Sn, Sb, Hg, Pb, and Bi in a small region.

The concentrations of K$_2$O, Ni, Rb, Sb, Cs, Ba, Hg, Bi, and U in the marine sediments were determined simply by the particle sizes of the clastics rather than by their source materials. The other elements in the marine sediments were influenced by their origin. Cluster analysis using the chemical compositions of stream and marine sediments suggested that the coarse sediments and fine sands of the Osumi Strait originate from pyroclastic rocks that erupted from the Kikai caldera and that the fine sands from the Hyuga Nada and Amakusa Nada originate from sedimentary rocks of accretionary complexes distributed in the adjacent terrestrial area. Silt–clay sediments are highly enriched in alkaline metal elements, MnO, Ni, and heavy metals such as Cu and Mo, the compositions of which are weakly
related to terrestrial materials. However, the silts of Kagoshima Bay have chemical compositions similar to those of stream sediments derived from pyroclastic flow deposits and are enriched in As, Mo, Cd, Sb, Hg, and Bi owing to sea water hydrothermal activity.

The concentration ratios of immobile elements are effective at negating the dilution effect of quartz and calcareous materials in coarse marine sediment and used as an index of source rock composition in marine sediments. According to the geochemical feature of stream sediment classified to the parent lithology, Cr/Ti and La/Yb ratios are used as the most effective indicators of the dynamic transfer processes of marine sediments in the study area. The spatial distribution of the lower Cr/Ti and La/Yb ratios suggest that sandy sediments originating from pyroclastic flow deposits are distributed within an 80 km radius of the Kikai caldera, and that these have subsequently been conveyed to the Hyuga Nada via a strong oceanic current. The spatial distribution of fine sands with an average Cr/Ti ratio and a high La/Yb ratio suggest that sandy sediments on the shelf, which originate from accretionary complexes in the adjacent terrestrial area, have been conveyed to the deep sea by gravitational transport.

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Figure captions

Fig. 1. Geographical names for terrestrial and marine areas. Open triangular symbols indicate active volcanoes that have erupted over the past century. The red lines numbered 1 and 2 represent the Kuroshio Current and Tsushima Current, respectively. The blue dotted lines numbered 3 and 4 represent the Oyashio and Liman currents, respectively. The gradation map of elevation was created using the digital elevation model provided by the Geospatial Information Authority of Japan. The bathymetric depth map was delineated using a data set provided by the Japan Oceanographic Data Center.

Fig. 2. Geological map of the Kyushu region of Japan, simplified from a geological map of Japan at a scale of 1:1 000 000 (Geological Survey of Japan (ed.) 1992). Sed: sediment and sedimentary rock; Acc: sedimentary rocks of accretionary complexes associated with metagabbro, metabasalt, and ultramafic rock; Mv: mafic volcanic rocks; Fv: felsic volcanic rocks; Py: pyroclastic flow deposit and debris; Gr: granitic rocks; Mp: metamorphic rocks (mostly high-pressure type). Circles with a dot and star symbol indicate a skarn-type deposit bearing Cu, Zn, As, and Sn; diamond symbols indicate a hydrothermal-type deposit bearing As and Au. The bathymetric depth contours were delineated using a data set provided by the Japan Oceanographic Data Center.

Fig. 3. Sampling locations of stream and marine sediments. The regions of the Tsushima and Osumi Straits and the Suo Amakusa, and Hyuga nadas; are delineated by zonal classifications represented by broken lines.

Fig. 4. Spatial distributions of elemental concentrations in the Kyushu region: (a) K₂O, (b) CaO, (c) MnO, (d) T–Fe₂O₃, (e) Cr, (f) Cs, (g) La, (h) Yb, (i) Cu, (j) Cd, (k) Sb, (l) Pb. Star, circle, and diamond symbols indicate the major metalliferous deposits shown in Fig. 1.

Fig. 5. (a) Cluster dendrogram obtained using a data set of 32 elements; only those in marine sediments are affected by terrestrial materials. (b) Cluster dendrogram obtained using a data set of 21 immobile elements. Sed: sediment and sedimentary rock; Acc: sedimentary rocks of accretionary complexes associated with metagabbro, metabasalt, and ultramafic rock; Mv: mafic volcanic rocks; Fv: felsic volcanic rocks; Py: pyroclastic flow deposit and debris; Gr: granitic rocks; Mp: metamorphic rocks (mostly high-pressure type). Amakusa, Hyuga, Kagoshima, Osumi, Suo, and Tsushima indicate the marine regions of the Amakusa Nada, Hyuga Nada, Kagoshima Bay, Osumi Strait, Suo Nada, and Tsushima Nada including Goto Nada and Genkai Nada, respectively. Coarse, fine, and silt indicate coarse sediment, fine sand, and silt including clay, respectively.

Fig. 6. Scatter diagram of Cr/Ti (×100) and La/Yb ratios in the Kyushu region. (a) Stream sediments derived from sediment and sedimentary rock; sedimentary rocks of accretionary complexes associated with metagabbro, metabasalt, and ultramafic rock;
mafic and felsic volcanic rocks; pyroclastic flow deposits and debris; and granitic and metamorphic rocks (mostly high-pressure type). (b) Maximum values of the x-axis and y-axis were set to 6.0 and 30, respectively, for stream sediment data sets. (c) Marine sediments classified by region. Sed: sediment and sedimentary rock; Acc: sedimentary rocks of accretionary complexes associated with metagabbro, metabasalt, and ultramafic rock; Mv: mafic volcanic rocks; Fv: felsic volcanic rocks; Py: pyroclastic flow deposit and debris; Gr: granitic rocks; Mp: metamorphic rocks (mostly high-pressure type). UL and LL indicate the upper and lower limits of the concentration ratios, respectively. Two samples of Tsushima Strait having outliers of Cr/Ti (×100) ratio (> 8.0) are excluded.

Fig. 7. Spatial distributions of (a) Cr/Ti (×100) and (b) La/Yb ratios in the Kyushu region. The shaded areas with diagonal lines indicate that the Cr/Ti and La/Yb ratios are less than 0.81 and 9.7, respectively; those with horizontal lines indicate ratios greater than 2.2 and 13.0, respectively.
Supplementary Information.

Of the 191 stream sediments collected from remote islands near the Kyushu mainland, 25 samples include Am04, Am14, Am23, Am26, Am33, and Am43 from Amakusa Island; Fke02, Fke04, and Fke10 from Fukue Island; Hr04 and Hr09 from Hirado Island; Iki01 from Ikinoshima Island; Ng02 from Nagashima Island; Nk03 and Nk08 from Nakadori Island; Tn02, Tn04, Tn08, Tn21, and Tn27 from Tanegashima Island; and Yk01, Yk10, Yk14, Yk18, and Yk22 from Yakushima Island (Ohta 2018).
Table 1. Summary for the geochemistry of marine and stream sediments in Kyushu region, Japan

|                  | Marine sediment (n = 606) | Stream sediment (n = 391) |
|------------------|---------------------------|---------------------------|
|                  | Min | Median | Mean | Max | MAD | Min | Median | Mean | Max | MAD |
| Na₂O wt%         | 0.545 | 2.61 | 2.62 | 5.48 | 0.45 | 0.548 | 2.18 | 2.18 | 5.44 | 0.42 |
| MgO wt%          | 0.413 | 2.68 | 3.03 | 9.69 | 0.49 | 0.554 | 2.34 | 2.63 | 7.61 | 0.758 |
| Al₂O₃ wt%        | 0.631 | 10.27 | 9.84 | 17.96 | 1.26 | 3.32 | 12.41 | 12.37 | 27.99 | 1.77 |
| P₂O₅ wt%         | 0.0227 | 0.114 | 0.122 | 0.504 | 0.026 | 0.045 | 0.150 | 0.170 | 0.767 | 0.038 |
| K₂O wt%          | 0.193 | 1.53 | 1.46 | 2.68 | 0.34 | 0.519 | 1.71 | 1.70 | 4.71 | 0.352 |
| CaO wt%          | 0.405 | 7.48 | 8.84 | 39.4 | 2.40 | 0.173 | 2.33 | 2.53 | 23.3 | 1.18 |
| TiO₂ wt%         | 0.0242 | 0.503 | 0.627 | 2.92 | 0.118 | 0.234 | 0.79 | 0.925 | 4.81 | 0.222 |
| MnO wt%          | 0.011 | 0.079 | 0.109 | 0.973 | 0.031 | 0.013 | 0.126 | 0.139 | 0.524 | 0.0413 |
| T-Fe₂O₃ wt%      | 0.304 | 4.54 | 5.51 | 35.4 | 1.03 | 1.50 | 5.81 | 6.36 | 16.0 | 1.40 |
| Li mg/kg         | 4.70 | 32.5 | 35.5 | 122 | 12.1 | 13.2 | 36.7 | 38.4 | 112 | 8.55 |
| Be mg/kg         | 0.105 | 1.11 | 1.09 | 3.05 | 0.26 | 0.715 | 1.49 | 1.56 | 5.96 | 0.233 |
| Sc mg/kg         | 0.955 | 11.1 | 13.9 | 71.9 | 2.57 | 2.18 | 13.6 | 15.2 | 61.6 | 4.04 |
| V mg/kg          | 3.93 | 88.2 | 106 | 1030 | 26.9 | 27.6 | 114 | 135 | 737 | 37.0 |
| Cr mg/kg         | 8.11 | 28.7 | 31.9 | 77.1 | 9.98 | 7.46 | 52.9 | 68.8 | 550 | 21.9 |
| Co mg/kg         | 1.29 | 9.26 | 10.8 | 54.0 | 2.41 | 3.66 | 14.6 | 16.3 | 68.4 | 4.25 |
| Ni mg/kg         | 3.82 | 12.3 | 15.3 | 62.7 | 3.71 | 3.86 | 20.4 | 26.4 | 301 | 8.99 |
| Cu mg/kg         | 1.22 | 10.5 | 13.4 | 303 | 4.21 | 5.19 | 28.0 | 35.1 | 737 | 8.98 |
| Zn mg/kg         | 7.57 | 73.9 | 77.8 | 330 | 13.1 | 39.3 | 114 | 128 | 990 | 23.2 |
| Ga mg/kg         | 0.702 | 13.2 | 12.4 | 20.9 | 1.50 | 7.52 | 17.4 | 17.4 | 31.4 | 1.51 |
| As mg/kg         | 0.2 | 5.6 | 6.0 | 31 | 1.9 | 0.7 | 8.0 | 18 | 2010 | 4.0 |
| Rb mg/kg         | 4.66 | 52.3 | 51.7 | 115 | 16.8 | 12.3 | 72.0 | 71.5 | 170 | 17.0 |
| Sr mg/kg         | 26.7 | 351 | 450 | 3000 | 105 | 46.2 | 155 | 175 | 528 | 47.9 |
| Y mg/kg          | 1.98 | 14 | 14.5 | 35.9 | 3.23 | 5.56 | 18.0 | 18.5 | 46.8 | 4.69 |
| Nb mg/kg         | 0.456 | 4.95 | 5.00 | 24.9 | 1.19 | 3.51 | 9.21 | 10.4 | 55.0 | 1.82 |
| Mo mg/kg         | 0.133 | 0.798 | 0.958 | 8.77 | 0.304 | 0.0853 | 1.20 | 1.47 | 16.5 | 0.35 |
| Cd mg/kg         | 0.013 | 0.0695 | 0.0794 | 0.454 | 0.0205 | 0.027 | 0.127 | 0.175 | 3.74 | 0.037 |
| Sn mg/kg         | 0.098 | 1.48 | 1.54 | 7.53 | 0.474 | 0.863 | 2.55 | 4.53 | 194 | 0.546 |
| Sb mg/kg         | 0.074 | 0.423 | 0.465 | 2.34 | 0.121 | 0.137 | 0.596 | 0.963 | 33.3 | 0.196 |
| Cs mg/kg         | 0.133 | 2.64 | 2.79 | 6.74 | 1.17 | 0.41 | 4.49 | 4.61 | 14.9 | 1.24 |
| Ba mg/kg         | 31.3 | 246 | 256 | 696 | 67.2 | 111 | 385 | 379 | 980 | 54.9 |
| La mg/kg         | 2.52 | 12.9 | 13.0 | 27.0 | 2.91 | 5.90 | 19.1 | 21.6 | 194 | 2.72 |
| Ce mg/kg         | 4.31 | 27.4 | 27.3 | 58.6 | 6.05 | 12.5 | 35.5 | 41.3 | 564 | 5.89 |
| Pr mg/kg         | 0.537 | 3.23 | 3.16 | 6.71 | 0.582 | 1.55 | 4.47 | 5.04 | 38.0 | 0.597 |
| Nd mg/kg         | 2.11 | 13.4 | 12.9 | 27.2 | 1.99 | 6.64 | 17.9 | 19.9 | 137 | 2.41 |
| Sm mg/kg         | 0.375 | 2.93 | 2.78 | 5.65 | 0.425 | 1.47 | 3.72 | 4.05 | 23.5 | 0.570 |
| Eu mg/kg         | 0.142 | 0.722 | 0.704 | 1.08 | 0.087 | 0.334 | 0.928 | 0.941 | 1.94 | 0.150 |
| Gd mg/kg         | 0.366 | 2.75 | 2.66 | 4.92 | 0.447 | 1.21 | 3.46 | 3.68 | 16.3 | 0.580 |
| Tb mg/kg         | 0.062 | 0.463 | 0.458 | 1.02 | 0.088 | 0.177 | 0.589 | 0.611 | 1.78 | 0.120 |
| Dy mg/kg         | 0.325 | 2.34 | 2.37 | 5.69 | 0.485 | 1.07 | 3.02 | 3.11 | 7.99 | 0.679 |
| Ho mg/kg         | 0.061 | 0.455 | 0.469 | 1.10 | 0.099 | 0.195 | 0.584 | 0.600 | 1.39 | 0.147 |
| Er mg/kg         | 0.195 | 1.35 | 1.39 | 3.25 | 0.305 | 0.496 | 1.70 | 1.75 | 4.22 | 0.463 |
| Element | Unit | Minimum | 1st Quartile | Median | 3rd Quartile | Maximum | MAD |
|---------|------|---------|--------------|--------|--------------|---------|-----|
| Tm      | mg/kg| 0.029   | 0.215        | 0.221  | 0.543        | 0.070   | 0.269|
| Yb      | mg/kg| 0.197   | 1.34         | 1.37   | 3.39         | 0.493   | 1.65 |
| Lu      | mg/kg| 0.03    | 0.195        | 0.203  | 0.473        | 0.064   | 0.244|
| Ta      | mg/kg| 0.002   | 0.471        | 0.456  | 1.61         | 0.123   | 0.68 |
| Hg      | mg/kg| 0.007   | 0.043        | 0.061  | 0.393        | 0.029   | 0.100|
| Tl      | mg/kg| 0.028   | 0.341        | 0.343  | 0.838        | 0.115   | 0.100|
| Pb      | mg/kg| 3.55    | 16.3         | 16.2   | 68.7         | 3.17    | 8.77 |
| Bi      | mg/kg| 0.039   | 0.194        | 0.226  | 1.60         | 0.084   | 0.100|
| Th      | mg/kg| 0.435   | 4.46         | 4.51   | 26.6         | 1.49    | 1.52 |
| U       | mg/kg| 0.232   | 1.09         | 1.08   | 4.51         | 0.264   | 0.148|

Minimum (Min), maximum (Max), and median absolute deviation (MAD).
Table 2. Median elemental concentration of stream sediment for each parent lithology

|     | Sed | Acc | Fv | Mv | Py | Gr | Mp | Oth |
|-----|-----|-----|----|----|----|----|----|-----|
|     | unit |     |    |    |    |    |    |     |
|     | n = 41 | n = 83 | n = 9 | n = 48 | n = 56 | n = 29 | n = 13 | n = 111 |
| Na$_2$O wt% | 1.76 | 1.76 | 1.86 | 2.23 | 2.45 | 3.03 | 2.68 | 2.30 |
| MgO wt% | 1.72 | 1.65 | 1.53 | 3.43 | 3.17 | 2.06 | 2.63 | 2.64 |
| Al$_2$O$_3$ wt% | 9.88 | 12.30 | 12.00 | 12.70 | 13.90 | 14.80 | 9.31 | 12.60 |
| P$_2$O$_5$ wt% | 0.131 | 0.114 | 0.157 | 0.159 | 0.164 | 0.180 | 0.167 | 0.160 |
| K$_2$O wt% | 1.78 | 2.10 | 1.73 | 1.18 | 1.27 | 2.17 | 1.44 | 1.68 |
| CaO wt% | 0.87 | 0.72 | 2.05 | 3.47 | 3.35 | 2.56 | 2.11 | 2.55 |
| TiO$_2$ wt% | 0.67 | 0.57 | 1.08 | 1.10 | 1.12 | 0.64 | 0.64 | 0.84 |
| MnO wt% | 0.076 | 0.096 | 0.123 | 0.152 | 0.159 | 0.116 | 0.101 | 0.140 |
| T-Fe$_2$O$_3$ wt% | 4.56 | 4.67 | 6.10 | 7.78 | 7.70 | 5.25 | 4.98 | 6.31 |
| Li mg/kg | 41 | 43 | 40 | 26 | 28 | 49 | 30 | 36 |
| Be mg/kg | 1.5 | 1.7 | 1.4 | 1.2 | 1.3 | 2.0 | 1.2 | 1.5 |
| Sc mg/kg | 9.0 | 10 | 14 | 17 | 18 | 10 | 11 | 15 |
| V mg/kg | 77 | 88 | 140 | 170 | 170 | 85 | 88 | 130 |
| Cr mg/kg | 61 | 53 | 47 | 78 | 30 | 31 | 110 | 50 |
| Co mg/kg | 13 | 12 | 12 | 20 | 19 | 12 | 15 | 17 |
| Ni mg/kg | 24 | 22 | 13 | 26 | 10 | 12 | 50 | 20 |
| Cu mg/kg | 24 | 31 | 28 | 27 | 26 | 41 | 30 |     |
| Zn mg/kg | 110 | 97 | 130 | 120 | 130 | 110 | 130 | 120 |
| Ga mg/kg | 14 | 17 | 17 | 19 | 18 | 22 | 14 | 17 |
| As mg/kg | 5.4 | 8.7 | 8.0 | 6.0 | 5.9 | 10 | 7.0 | 9.0 |
| Rb mg/kg | 75 | 90 | 75 | 41 | 52 | 100 | 65 | 70 |
| Sr mg/kg | 100 | 110 | 190 | 230 | 220 | 160 | 130 | 170 |
| Y mg/kg | 13 | 12 | 17 | 17 | 22 | 26 | 15 | 21 |
| Nb mg/kg | 9.1 | 8.2 | 8.4 | 11 | 10 | 12 | 7.2 | 9.2 |
| Mo mg/kg | 0.81 | 1.1 | 1.5 | 1.3 | 1.4 | 0.75 | 0.88 | 1.3 |
| Cd mg/kg | 0.13 | 0.11 | 0.15 | 0.13 | 0.14 | 0.12 | 0.12 | 0.13 |
| Sn mg/kg | 2.3 | 2.6 | 2.9 | 2.4 | 2.3 | 4.0 | 2.5 | 2.6 |
| Sb mg/kg | 0.55 | 0.81 | 1.2 | 0.46 | 0.47 | 0.38 | 0.61 | 0.63 |
| Cs mg/kg | 4.0 | 5.5 | 5.5 | 2.8 | 3.5 | 5.0 | 4.0 | 4.9 |
| Ba mg/kg | 380 | 430 | 370 | 340 | 360 | 380 | 280 | 380 |
| La mg/kg | 20 | 19 | 18 | 18 | 18 | 24 | 16 | 19 |
| Ce mg/kg | 35 | 38 | 36 | 30 | 36 | 44 | 29 | 37 |
| Pr mg/kg | 4.4 | 4.5 | 4.1 | 4.0 | 4.3 | 5.5 | 3.7 | 4.6 |
| Nd mg/kg | 17 | 18 | 16 | 17 | 18 | 22 | 15 | 19 |
| Sm mg/kg | 3.3 | 3.5 | 3.4 | 3.5 | 3.8 | 5.0 | 3.2 | 3.9 |
| Eu mg/kg | 0.73 | 0.78 | 0.90 | 0.98 | 0.99 | 1.1 | 0.84 | 0.99 |
| Gd mg/kg | 2.9 | 3.0 | 3.2 | 3.2 | 3.7 | 4.7 | 3.0 | 3.8 |
| Tb mg/kg | 0.43 | 0.48 | 0.58 | 0.55 | 0.68 | 0.82 | 0.52 | 0.66 |
| Element | Unit | Data 1 | Data 2 | Data 3 | Data 4 | Data 5 | Data 6 | Data 7 | Data 8 |
|---------|------|--------|--------|--------|--------|--------|--------|--------|--------|
| Dy      | mg/kg| 2.2    | 2.2    | 3.0    | 2.8    | 3.7    | 4.2    | 2.6    | 3.4    |
| Ho      | mg/kg| 0.42   | 0.41   | 0.57   | 0.55   | 0.74   | 0.83   | 0.51   | 0.67   |
| Er      | mg/kg| 1.2    | 1.2    | 1.7    | 1.6    | 2.2    | 2.3    | 1.5    | 2.0    |
| Tm      | mg/kg| 0.18   | 0.18   | 0.25   | 0.26   | 0.35   | 0.30   | 0.23   | 0.32   |
| Yb      | mg/kg| 1.2    | 1.1    | 1.5    | 1.6    | 2.3    | 1.7    | 1.4    | 2.0    |
| Lu      | mg/kg| 0.18   | 0.16   | 0.22   | 0.25   | 0.33   | 0.24   | 0.20   | 0.29   |
| Ta      | mg/kg| 0.64   | 0.73   | 1.0    | 0.69   | 0.82   | 1.1    | 0.35   | 0.76   |
| Hg      | mg/kg| 0.061  | 0.070  | 0.070  | 0.065  | 0.040  | 0.030  | 0.070  | 0.060  |
| Tl      | mg/kg| 0.45   | 0.57   | 0.59   | 0.34   | 0.42   | 0.54   | 0.36   | 0.50   |
| Pb      | mg/kg| 21     | 23     | 24     | 20     | 21     | 28     | 21     | 23     |
| Bi      | mg/kg| 0.20   | 0.31   | 0.34   | 0.16   | 0.27   | 0.37   | 0.20   | 0.29   |
| Th      | mg/kg| 5.9    | 6.5    | 6.1    | 4.7    | 6.1    | 8.3    | 5.2    | 6.7    |
| U       | mg/kg| 1.4    | 1.4    | 1.7    | 1.0    | 1.6    | 2.5    | 1.3    | 1.7    |
Table 3. Skewness of the unchanged and the log-transformed data, data transformation, and significance probability ($p$) and size effect ($\eta^2$) of one-way ANOVA

|                | Skewness | Data Transformation | One-way ANOVA |
|----------------|----------|---------------------|---------------|
|                | Unchanged| Log-transformed     | $p$          | $\eta^2$     |
| Na$_2$O        | 0.5      | -1.0                | <0.01        | 0.29         |
| MgO            | 1.2      | 0.0                 | <0.01        | 0.34         |
| Al$_2$O$_3$    | 0.5      | -1.1                | <0.01        | 0.23         |
| P$_2$O$_5$     | 2.9      | 0.6                 | <0.01        | 0.15         |
| K$_2$O         | 0.7      | -0.6                | <0.01        | 0.45         |
| CaO            | 3.7      | -0.6                | <0.01        | 0.52         |
| TiO$_2$        | 2.9      | 0.8                 | <0.01        | 0.37         |
| MnO            | 1.6      | -0.4                | <0.01        | 0.22         |
| Fe$_2$O$_3$    | 1.1      | 0.0                 | <0.01        | 0.35         |
| Li             | 1.3      | 0.0                 | <0.01        | 0.31         |
| Be             | 3.3      | 0.9                 | <0.01        | 0.41         |
| Sc             | 1.8      | -0.1                | <0.01        | 0.36         |
| V              | 1.5      | 0.2                 | <0.01        | 0.39         |
| Cr             | 3.3      | 0.2                 | <0.01        | 0.27         |
| Co             | 1.9      | 0.1                 | <0.01        | 0.24         |
| Ni             | 5.6      | 0.3                 | <0.01        | 0.25         |
| Cu             | 12       | 0.9                 | <0.01        | 0.06         |
| Zn             | 5.8      | 1.0                 | 0.013        | 0.06         |
| Ga             | 0.5      | -0.7                | <0.01        | 0.37         |
| As             | 17       | 1.1                 | <0.01        | 0.09         |
| Rb             | 0.3      | -0.9                | <0.01        | 0.48         |
| Sr             | 1.2      | 0.1                 | <0.01        | 0.39         |
| Y              | 0.6      | -0.4                | <0.01        | 0.31         |
| Nb             | 3.7      | 1.1                 | <0.01        | 0.18         |
| Mo             | 5.5      | 0.4                 | <0.01        | 0.15         |
| Cd             | 9.8      | 1.1                 | 0.39         | 0.02         |
| Sn             | 12       | 2.7                 | 0.02         | 0.05         |
| Sb             | 12       | 1.4                 | <0.01        | 0.26         |
| Cs             | 1.0      | -0.9                | <0.01        | 0.24         |
| Ba             | 0.7      | -0.7                | <0.01        | 0.16         |
| La             | 7.6      | 2.6                 | <0.01        | 0.24         |
| Ce             | 9.2      | 2.7                 | <0.01        | 0.22         |
| Pr             | 7.2      | 2.5                 | <0.01        | 0.24         |
| Nd             | 7.0      | 2.2                 | <0.01        | 0.23         |
| Sm             | 6.1      | 1.6                 | <0.01        | 0.24         |
| Eu             | 0.6      | -0.5                | <0.01        | 0.23         |
| Gd             | 4.3      | 0.7                 | <0.01        | 0.23         |
| Tb             | 1.7      | -0.1                | <0.01        | 0.25         |
| Element | Log-transformed | Unchanged | p-value 1 | p-value 2 |
|---------|----------------|-----------|-----------|-----------|
| Dy      | 1.0            | -0.2      | Log-transformed | <0.01 | 0.28 |
| Ho      | 0.5            | -0.4      | Log-transformed | <0.01 | 0.29 |
| Er      | 0.4            | -0.5      | Unchanged | <0.01 | 0.28 |
| Tm      | 0.4            | -0.5      | Unchanged | <0.01 | 0.29 |
| Yb      | 0.4            | -0.4      | Unchanged | <0.01 | 0.30 |
| Lu      | 0.4            | -0.4      | Unchanged | <0.01 | 0.31 |
| Ta      | 2.6            | 0.0       | Log-transformed | <0.01 | 0.20 |
| Hg      | 17             | 0.8       | Log-transformed | <0.01 | 0.08 |
| Tl      | 1.6            | -1.0      | Log-transformed | <0.01 | 0.28 |
| Pb      | 15             | 3.0       | Log-transformed | <0.01 | 0.10 |
| Bi      | 13             | 0.9       | Log-transformed | <0.01 | 0.21 |
| Th      | 6.3            | 1.6       | Log-transformed | <0.01 | 0.25 |
| U       | 2.9            | 0.5       | Log-transformed | <0.01 | 0.26 |
| Cr/Ti   | 4.1            | -0.2      | Log-transformed | <0.01 | 0.40 |
| La/Yb   | 6.5            | 1.4       | Log-transformed | <0.01 | 0.36 |
| A*  | B*  | Felsic elements | Mafic elements | Sulphophile elements |
|-----|-----|-----------------|----------------|---------------------|
| Sed | Acc | Cs              | Al             | As, Bi              |
| Sed | Fv  | Ca, Sc          |                |                     |
| Sed | Mv  | K, Li, Rb, Tl, Th, U | Eu             | Mg, Al, Ca, Ti, Mn, Fe, Sc, V, Co, Ga, Sr | Mo |
| Sed | Py  | K, Li, Rb       | Na, Eu, Ta     | Cr, Ni              | Mg, Al, Ca, Ti, Mn, Fe, Sc, V, Ga, Sr, Y, Tb–Lu | Mo |
| Sed | Gr  | Na, K, Rb, Nb, La–Eu, Ta, Th, U | Cr, Ni         | Al, Ca, Ga, Sr, Y, Gd–Lu | Sb | Bi |
| Sed | Mp  | Na              | Na             | Mg, Cr              |                     |
| Acc | Sed | Cs              | Al             | Mg, Cr              |
| Acc | Fv  |                | Ca, Ti         |                     |
| Acc | Mv  | K, Li, Rb, Cs, Ba, Ti, Th | Na, Nb, Eu     | Mg, P, Ca, Ti, Mn, Fe, Sc, V, Co, Ga, Sr, Y, Ho–Lu, Sb, Pb, Bi |
| Acc | Py  | K, Li, Rb, Cs, Ba, Ti | Na, Nb, Eu     | Cr, Ni              | Mg, P, Ca, Ti, Mn, Fe, Sc, V, Co, Sr, Y, Gd–Lu | As, Sb, Pb |
| Acc | Gr  | Na, Nb, La–Eu, Ta, Th, U | Cr, Ni         | Al, P, Ca, Ga, Sr, Y, Gd–Lu | Mo, Sb |
| Acc | Mp  | K, Rb, Ba, Ta, Ti | Na             | Ca, Cr, Ni          |                     |
| Mv  | Sed | Eu              | K, Li, Rb, Tl, Th, U | Mg, Al, Ca, Ti, Mn, Fe, Sc, V, Co, Ga, Sr | Mo |
| Mv  | Acc | Na, Nb, Eu      | K, Li, Rb, Cs, Ba, Tl, Th | Mg, P, Ca, Ti, Mn, Fe, Sc, V, Co, Ga, Sr, Y, Ho–Lu | Sb, Pb, Bi |
| Mv  | Fv  | K, Rb, Cs, Tl   | K, Rb, Cs, Tl  | Cr, Ni              | Ho–Lu               | Bi |
| Mv  | Py  | U               | U              | Cr, Ni              | Ho–Lu               | Bi |

Table 4. Results of the Bonferroni multiple comparison procedure at the 0.01 confidence interval.
| A | B | Felsic element | Mafic element | Sulphophile elements |
|---|---|---|---|---|
| Py | Sed | Na, Eu, Ta | K, Li, Rb | Mg, Al, Ca, Ti, Mn, Fe, Sc, V, Ga, Sr, Y, Tb–Lu | Cr, Ni | Mo |
| Py | Acc | Na, Nb, Eu | K, Li, Rb, Cs, Ba, Ti | Mg, P, Ca, Ti, Mn, Fe, Sc, V, Co, Sr, Y, Gd–Lu | Cr, Ni | As, Sb, Pb |
| Py | Fv | | | | |
| Py | Mv | U | Ho–Lu | Cr, Ni | | Bi |
| Py | Gr | Na, K, Li, Rb, Cs, La–Sm, Th, U | Mg, Ti, Fe, Sc, V, Co | Ga, Gd | | Mo |
| Py | Mp | Nb, Eu, Ta | Al, Ti, Fe, Sc, V, Ga, Sr, Tm–Lu | Cr, Ni | | |
| Gr | Sed | Na, K, Rb, Nb, La–Eu, Ta, Th, U | Al, Ca, Ga, Sr, Y, Gd–Lu | Cr, Ni | Bi | Sb |
| Gr | Acc | Na, Nb, La–Eu, Ta, Th, U | Al, P, Ca, Ga, Sr, Y, Gd–Lu | Cr, Ni | Mo, Sb | | |
| Gr | Fv  | Na, La–Sm, Th, U | Ga, Gd | Ga, Y, Gd–Ho | Mg, Ti, Fe, Sc, V, Cr, Co, Ni | Bi | Mo |
|----|-----|-----------------|--------|--------------|--------------------------------|----|----|
| Gr | Mv  | Na, K, Li, Rb, Cs, La–Sm, Ta, Th, U | Ga, Gd | Mg, Ti, Fe, Sc, V, Co | Mo |
| Gr | Py  | Na, K, Li, Rb, Cs, La–Sm, Ta, Th, U | Ga, Gd | Mg, Ti, Fe, Sc, V, Co | Mo |
| Gr | Mv  | K, Rb, Nb, Ba, La–Eu, Ta, Th, U | Al, Ga, Gd, Tb, Dy | Cr, Ni |
| Mp | Sed | Na | Mg, Cr |
| Mp | Acc | Na | K, Rb, Ba, Ta, Th, U | Ca, Cr, Ni |
| Mp | Fv  | Ta | Cr, Ni |
| Mp | Mv  | Nb, Ta | Ni | Ti, Fe, V, Ga, Sr |
| Mp | Py  | Nb, Eu, Ta | Cr, Ni | Al, Ti, Fe, Sc, V, Ga, Sr, Tm–Lu |
| Mp | Gr  | K, Rb, Nb, Ba, La–Eu, Ta, Th, U | Cr, Ni | Al, Ga, Gd, Tb, Dy |

*Abbreviations of Sed, Acc, Fv, Mv, Py, Gr, and Mp are the same as Table 2. A > (or <) B indicates that elemental concentrations of sediments originated from A is significantly higher (or lower) than those of sediments originated from B.*
Table 5. Median elemental concentrations of marine sediments classified by regions and particel sizes

|                  | Tsushima St. | Suo Nada | Amakusa Nada | Hyuga Nada | Osumi St. | Silt in Bay* |
|------------------|--------------|----------|--------------|------------|-----------|-------------|
|                  | C. sed d*    | F. san d* | C. sed d*    | F. san d* | C. sed d* | F. san d* |
| **n**            | 21           | 25       | 9            | 7          | 8         | 11         |
| **wt. %**        |              |          |              |            |           |            |
| Na               | 1.4          | 1.7      | 2.9          | 2.8        | 3.9       | 2.1        |
| O                | 3.0          | 2.1      | 4.0          | 4.2        | 6.1       | 3.2        |
| Mg               | 1.4          | 2.2      | 3.0          | 2.2        | 3.1       | 3.2        |
| O                | 6.0          | 3.0      | 3.0          | 2.9        | 6.0       | 3.0        |
| Al               | 4.3          | 5.3      | 9.0          | 7.4        | 8.1       | 7.3        |
| O₃               | 9.0          | 4.6      | 9.3          | 4.3        |           | 9.0        |
| P                | 54           | 78       | 99           | 85         | 39        | 98         |
| O₅               | 1.5          | 1.3      | 1.9          | 2.1        | 1.6       | 1.7        |
| O                | 1.0          | 5.2      | 9.6          | 9.9        |           | 8.2        |
| Ca               | 12.12        | 13.4     | 14.8         | 15.1       | 17.0      | 14.8       |
| TiO              | 93.7         | 37.8     | 37.8         | 37.8       |           | 37.8       |
| Mn               | 0.0          | 0.0      | 0.0          | 0.0        | 0.0       | 0.0        |
| O                | 19.22        | 22.1     | 19.6         | 66.8       |           | 19.6       |
| Fe₂               | 1.2          | 1.8      | 5.0          | 3.5        | 4.2       | 4.1        |
| O₃               | 9.0          | 4.9      | 9.4          | 4.7        | 7.0       | 1.3        |
| mg/kg            |              |          |              |            |           |            |
| Li               | 16.0         | 20.6     | 26.1         | 61.10      | 110       | 28.1       |
| Be               | 0.4          | 0.6      | 1.2          | 1.5        | 1.6       | 0.8        |
| Sc               | 2.2          | 3.7      | 9.1          | 6.4        | 8.9       | 9.1        |
| V                | 19.24        | 24.1     | 55.4         | 46.76      | 74.1      | 55.56      |
| Cr               | 17.27        | 27.1     | 38.4         | 40.62      | 62.1      | 29.32      |
| Co               | 2.43         | 3.74     | 14.9          | 9.71       | 11.1      | 9.34       |
| Ni               | 8.41         | 11.1     | 15.1         | 17.4       | 24.1      | 12.13      |
| Cu               | 3.20         | 5.2      | 9.5          | 13.8       | 26.1      | 7.14       |
| Zn               | 17.27        | 27.1     | 77.83        | 130.1      | 130.0     | 56.58      |
| Ga               | 4.66         | 6.19     | 14.14        | 16.1       | 16.0      | 9.83       |
| As               | 6.50         | 3.70     | 6.8          | 5.8        | 9.2       | 7.1        |
| Rb               | 53           | 51       | 41.87        | 43.0        | 43.0      | 28.51      |
| Sr               | 650          | 680      | 380          | 139.1      | 130.0     | 740.420    |
| Y                | 4.85         | 7.18     | 13.1         | 13.0       | 13.0      | 11.12      |
| Nb               | 1.82         | 2.72     | 6.2          | 6.1        | 8.8       | 5.1        |
| Mo               | 0.20         | 0.30     | 0.6          | 0.8        | 0.6       | 0.6        |
| Cd               | 0.00         | 0.0      | 0.0          | 0.0        | 0.2       | 0.0        |
| Sn               | 0.30         | 0.50     | 1.5          | 2.4        | 3.5       | 1.0        |

Note: * Concentrations in mg/kg.
|     | 0.3 | 0.3 | 0.3 | 0.4 | 0.4 | 0.3 | 0.5 | 0.4 | 0.8 | 0.2 | 0.4 | 0.7 | 0.4 |
|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|
| Sb  | 1.0 | 1.3 | 2.1 | 4.0 | 4.4 | 1.5 | 2.9 | 3.6 | 2.0 | 3.6 | 4.8 | 1.3 | 2.3 | 5.3 | 2.4 |
| Cs  | 290 | 240 | 300 | 260 | 210 | 170 | 260 | 190 | 300 | 310 | 340 | 160 | 230 | 420 | 210 |
| Ba  | 5.1 | 8.9 | 13  | 13  | 16  | 11  | 14  | 17  | 12  | 16  | 16  | 10  | 12  | 19  | 11  |
| Ce  | 12  | 20  | 28  | 28  | 27  | 22  | 29  | 35  | 25  | 33  | 34  | 22  | 26  | 38  | 22  |
| Pr  | 1.2 | 2.0 | 3.1 | 3.1 | 3.8 | 2.4 | 3.2 | 3.8 | 3.0 | 3.7 | 3.9 | 2.7 | 3.1 | 4.3 | 2.8 |
| Nd  | 4.6 | 8.0 | 13  | 13  | 15  | 10  | 12  | 15  | 13  | 15  | 15  | 12  | 13  | 17  | 12  |
| Sm  | 0.8 | 4   | 1.5 | 2.7 | 2.7 | 3.1 | 2.1 | 2.6 | 3.1 | 2.8 | 3.0 | 3.2 | 2.9 | 3.0 | 3.5 | 2.8 |
| Eu  | 0.3 | 0.4 | 0.7 | 0.6 | 0.6 | 0.7 | 0.7 | 0.7 | 0.7 | 0.6 | 0.6 | 0.7 | 0.7 | 0.7 | 0.7 | 0.7 |
| Ho  | 0   | 3   | 6   | 2   | 4   | 3   | 1   | 3   | 7   | 9   | 9   | 8   | 5   | 6   | 6   |
| Gd  | 0.8 | 5   | 1.3 | 2.6 | 2.4 | 2.8 | 2.1 | 2.4 | 2.9 | 2.5 | 2.6 | 2.9 | 2.9 | 2.9 | 3.1 | 3.0 |
| Tb  | 0.1 | 0.2 | 0.4 | 0.4 | 0.4 | 0.3 | 0.4 | 0.4 | 0.3 | 0.4 | 0.4 | 0.5 | 0.5 | 0.5 | 0.5 | 0.5 |
| Dy  | 0.7 | 0   | 1   | 2.3 | 2.1 | 2.5 | 1.8 | 2.1 | 2.4 | 2.0 | 2.1 | 2.3 | 2.7 | 2.6 | 2.6 | 3.0 |
| Ho  | 0.1 | 0.2 | 0.4 | 0.4 | 0.4 | 0.3 | 0.3 | 0.4 | 0.3 | 0.3 | 0.4 | 0.5 | 0.5 | 0.5 | 0.5 | 0.6 |
| Eu  | 0.4 | 1   | 0.6 | 1.3 | 1.3 | 1.4 | 1.1 | 1.2 | 1.3 | 1.1 | 1.1 | 1.3 | 1.6 | 1.6 | 1.5 | 1.9 |
| Tm  | 0.0 | 0.0 | 0.2 | 0.2 | 0.2 | 0.1 | 0.1 | 0.2 | 0.1 | 0.1 | 0.2 | 0.2 | 0.2 | 0.2 | 0.2 | 0.3 |
| Yb  | 0.3 | 6   | 1.3 | 1.3 | 1.3  | 1.1 | 1.2 | 1.2 | 1.1 | 1.1 | 1.3 | 1.5 | 1.5 | 1.5 | 1.5 | 2.0 |
| Lu  | 0.0 | 0.0 | 0.1 | 0.1 | 0.2  | 0.1 | 0.1  | 0.1 | 0.1 | 0.1 | 0.2 | 0.2 | 0.2 | 0.2 | 0.2 | 0.2 |
| Hg  | 0.0 | 0.0 | 0.0 | 0.0 | 0.0  | 0.0 | 0.0  | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 |
| Tl  | 8  | 9   | 8   | 0   | 9    | 7   | 5    | 7   | 8   | 4   | 1   | 8   | 9   | 9   | 2   | 69  |
| Pb  | 0.0 | 0.0 | 0.2 | 0.4 | 0.6  | 0.5  | 0.1  | 0.3  | 0.4  | 0.3  | 0.4  | 0.5  | 0.1  | 0.2  | 0.4  | 0.4  |
| Bi  | 8  | 1   | 2   | 0   | 3    | 0.6 | 0.1  | 0.1  | 0.3  | 0.1  | 0.2  | 0.4  | 0.1  | 0.1  | 0.3  | 0.4  |
| Th  | 1.4 | 2.5 | 3.8 | 5.9 | 6.2  | 2.4 | 4.7  | 5.7  | 3.4  | 5.5  | 6.4  | 2.6  | 4.2  | 6.9  | 4.1  |
| U   | 0.5 | 0.9 | 1.0 | 1.5 | 1.5  | 0.6  | 1.2  | 1.6  | 0.8  | 1.1  | 1.7  | 0.7  | 1.1  | 1.3  | 1.2  |

*Cr: Sed, F. sand, silt in Bay indicate coarse sediments, fine sand, and silt collected from Kagoshima Bay, respectively.*
Table 6. Skewness of the unchanged and the log-transformed data, data transformation, and significance probability (p) and size effect ($\eta^2$) of two-way ANOVA

| Skewness          | Data transformation | p       | $\eta^2$       | Major factor |
|-------------------|---------------------|---------|----------------|--------------|
| Unch. * Log-trans. * |  | Region | Particel size | Int.† | Region | Particel size | Int.† | Major factor |
| **MgO** | 1.9 | 0.2 | Log-trans. | <0.01 | <0.01 | <0.01 | 1 | 0.15 | 0.12 | 0.12 | Region |
| **Al2O3** | -1.0 | -2.9 | Unch. | <0.01 | <0.01 | <0.01 | 1 | 0.37 | 0.02 | 0.02 | Region |
| **P2O5** | 2.4 | 0.1 | Log-trans. | <0.01 | <0.01 | <0.01 | 1 | 0.22 | 0.03 | 0.07 | Region |
| **K2O** | -0.5 | -1.6 | Unch. | <0.01 | <0.01 | <0.01 | 1 | 0.12 | 0.26 | 0.08 | Particle size |
| **CaO** | 2.2 | -0.3 | Log-trans. | <0.01 | 0.02 | <0.01 | 1 | 0.28 | 0.01 | 0.05 | Region |
| **TiO2** | 2.1 | -0.5 | Log-trans. | <0.01 | <0.01 | <0.01 | 1 | 0.40 | 0.04 | 0.06 | Region |
| **MnO** | 3.5 | 0.2 | Log-trans. | <0.01 | <0.01 | <0.01 | 1 | 0.36 | 0.05 | 0.03 | Region |
| **Fe2O3** | 2.9 | -0.5 | Log-trans. | <0.01 | <0.01 | <0.01 | 1 | 0.36 | 0.06 | 0.07 | Region |
| **Li** | 1.4 | -0.3 | Log-trans. | <0.01 | <0.01 | <0.01 | 1 | 0.28 | 0.26 | 0.03 | Both |
| **Be** | -0.1 | -1.6 | Unch. | <0.01 | <0.01 | <0.01 | 1 | 0.27 | 0.20 | 0.03 | Region |
| **Sc** | 2.4 | -0.4 | Log-trans. | <0.01 | <0.01 | <0.01 | 1 | 0.46 | 0.05 | 0.06 | Region |
| **V** | 4.4 | -0.3 | Log-trans. | <0.01 | <0.01 | <0.01 | 1 | 0.43 | 0.05 | 0.03 | Region |
| **Cr** | 0.6 | -0.1 | Log-trans. | <0.01 | <0.01 | <0.01 | 1 | 0.21 | 0.22 | 0.09 | Both |
| **Co** | 2.0 | -0.4 | Log-trans. | <0.01 | <0.01 | <0.01 | 1 | 0.36 | 0.05 | 0.07 | Region |
| **Ni** | 1.9 | 0.5 | Log-trans. | <0.01 | <0.01 | <0.01 | 1 | 0.11 | 0.41 | 0.07 | Particle size |
| **Cu** | 12.5 | 0.3 | Log-trans. | <0.01 | <0.01 | <0.01 | 1 | 0.14 | 0.35 | 0.04 | Region |
| **Zn** | 1.6 | -1.1 | Log-trans. | <0.01 | <0.01 | <0.01 | 1 | 0.43 | 0.01 | 0.08 | Region |
| **Ga** | -1.3 | -2.9 | Unch. | <0.01 | <0.01 | <0.01 | 1 | 0.42 | 0.03 | 0.02 | Region |
| **As** | 1.7 | -1.0 | Log-trans. | <0.01 | <0.01 | <0.01 | 1 | 0.08 | 0.03 | 0.04 | Region |
| **Rb** | 0.0 | -1.3 | Unch. | <0.01 | <0.01 | <0.01 | 1 | 0.04 | 0.30 | 0.13 | Particle size |
| **Sr** | 3.4 | 0.6 | Log-trans. | <0.01 | <0.01 | <0.01 | 1 | 0.18 | 0.03 | 0.04 | Region |
| **Y** | 0.5 | -0.8 | Unch. | <0.01 | 0.93 | 0.05 | 1 | 0.33 | 0.00 | 0.02 | Region |
| **Nb** | 1.8 | -1.6 | Log-trans. | <0.01 | <0.01 | <0.01 | 1 | 0.21 | 0.10 | 0.03 | Region |
| **Mo** | 4.9 | 0.5 | Log-trans. | <0.01 | <0.01 | <0.01 | 1 | 0.17 | 0.15 | 0.01 | Both |
| **Cd** | 3.1 | 0.2 | Log-trans. | <0.01 | <0.01 | <0.01 | 1 | 0.14 | 0.18 | 0.05 | Particle |
| Element | Log-Trans. | Log-Trans. | 1 | 0.35 | 0.18 | 0.02 | Region |
|---------|-----------|-----------|---|------|------|------|--------|
| Sn      | 1.5       | 1.0       | <0.01 | <0.01 | 0.35 | 0.18 | 0.02 |
| Sb      | 1.7       | 0.3       | <0.01 | <0.01 | 0.05 | 0.31 | 0.05 |
| Cs      | 0.3       | -0.9      | Unch. | <0.01 | <0.01 | 0.09 | 0.49 | 0.06 |
| Ba      | 0.6       | -1.1      | Unch. | <0.01 | <0.01 | 0.06 | 0.29 | 0.18 |
| La      | 0.0       | -1.2      | Unch. | <0.01 | <0.01 | 0.14 | 0.32 | 0.07 |
| Ce      | -0.1      | -1.2      | Unch. | <0.01 | <0.01 | 0.14 | 0.27 | 0.07 |
| Pr      | -0.3      | -1.6      | Unch. | <0.01 | <0.01 | 0.20 | 0.25 | 0.05 |
| Nd      | -0.5      | -1.8      | Unch. | <0.01 | <0.01 | 0.24 | 0.18 | 0.04 |
| Sm      | -0.7      | -1.9      | Unch. | <0.01 | 0.01  | 0.30 | 0.08 | 0.02 |
| Eu      | -0.9      | -2.1      | Unch. | <0.01 | 0.94  | 0.35 | 0.00 | 0.03 |
| Gd      | -0.4      | -1.7      | Unch. | <0.01 | 0.05  | 0.32 | 0.02 | 0.02 |
| Tb      | -0.1      | -1.4      | Unch. | <0.01 | 0.09  | 0.32 | 0.01 | 0.02 |
| Dy      | 0.2       | -1.2      | Unch. | <0.01 | 0.56  | 0.32 | 0.00 | 0.02 |
| Ho      | 0.4       | -1.1      | Unch. | <0.01 | 0.71  | 0.32 | 0.00 | 0.02 |
| Er      | 0.4       | -1.0      | Unch. | <0.01 | 0.95  | 0.33 | 0.00 | 0.02 |
| Tm      | 0.4       | -1.0      | Unch. | <0.01 | 0.96  | 0.32 | 0.00 | 0.02 |
| Yb      | 0.5       | -0.9      | Unch. | <0.01 | 0.97  | 0.32 | 0.00 | 0.02 |
| Lu      | 0.5       | -0.9      | Unch. | <0.01 | 0.98  | 0.32 | 0.00 | 0.02 |
| Ta      | 0.3       | -3.1      | Unch. | <0.01 | <0.01 | 0.18 | 0.17 | 0.03 |
| Hg      | 1.5       | -0.5      | Log-Trans. | <0.01 | <0.01 | 0.06 | 0.38 | 0.02 |
| Tl      | 0.1       | -1.2      | Unch. | <0.01 | <0.01 | 0.16 | 0.34 | 0.05 |
| Pb      | 1.7       | -0.8      | Log-Trans. | <0.01 | 0.03  | 0.14 | 0.24 | 0.02 |
| Bi      | 3.2       | -0.1      | Log-Trans. | <0.01 | 0.92  | 0.07 | 0.32 | 0.00 |
| Th      | 1.9       | -0.8      | Log-Trans. | <0.01 | <0.01 | 0.14 | 0.31 | 0.03 |
| U       | 1.3       | -0.5      | Log-Trans. | <0.01 | <0.01 | 0.41 | 0.18 | 0.05 |
| Cr/Ti   | 2.8       | -0.2      | Log-Trans. | <0.01 | <0.01 | 0.26 | 0.09 | 0.07 |
| La/Yb   | 0.3       | -0.7      | Unch. | <0.01 | <0.01 | 0.26 | 0.09 | 0.07 |

*Unch and Log-Trans. Indicates that composition data were unchanged and log-transformed for ANOVA, respectively.

†Int. indicates interaction effect.
### Appendix Table 1. Sediment yield and river water discharge data for each river system of the study area

| River system | Average rate of sediment yield* | Drainage basin area† | Sediment yield | Water discharge in 2000† | Discharged area | Relative rate to total sediment yield |
|--------------|---------------------------------|-----------------------|----------------|--------------------------|----------------|--------------------------------------|
|              | m³/km²/year                      | km²                   | m³/yr          | m³/yr (×10³)             |                |                                     |
| Onga         | 122                             | 695                   | 85             | 581                      | Genkai Nada   | 3.2%                                 |
| Matsu-ura    | 115                             | 275                   | 32             | 248                      | Genkai Nada   | 1.2%                                 |
| Chikugo      | 144                             | 2315                  | 333            | 2470                     | Ariake Sea    | 12%                                  |
| Yabe         | 139                             | 460                   | 64             | 440                      | Ariake Sea    | 2.4%                                 |
| Rokkaku      | 122                             | 95                    | 12             | 93                       | Ariake Sea    | 0.4%                                 |
| Kase         | 112                             | 256                   | 29             | 308                      | Ariake Sea    | 1.1%                                 |
| Hon-myo‡     | 140‡                            | 36                    | 5              | 52                       | Ariake Sea    | 0.2%                                 |
| Kikuchi      | 124                             | 906                   | 112            | 1248                     | Ariake Sea    | 4.2%                                 |
| Shirakawa    | 333                             | 477                   | 159            | 685                      | Ariake Sea    | 5.9%                                 |
| Midorikawa   | 140                             | 681                   | 95             | 940                      | Ariake Sea    | 3.6%                                 |
| Kama         | 137                             | 1856                  | 254            | 3288                     | Yatsushiro Sea| 9.5%                                 |
| Yamakuni     | 183                             | 483                   | 88             | 418                      | Suo Nada      | 3.3%                                 |
| Oita         | 230                             | 494                   | 114            | 498                      | Suo Nada      | 4.3%                                 |
| Ono          | 164                             | 1239                  | 203            | 1616                     | Suo Nada      | 7.6%                                 |
| Banjo        | 192                             | 278                   | 53             | 392                      | Suo Nada      | 2.0%                                 |
| Gokase       | 320                             | 1044                  | 334            | 1922                     | Hyuga Nada    | 13%                                  |
| Omaru        | 436                             | 396                   | 173            | 960                      | Hyuga Nada    | 6.5%                                 |
| Oyodo        | 171                             | 1564                  | 267            | 3348                     | Hyuga Nada    | 10%                                  |
| Sendai       | 192                             | 1348                  | 259            | 2529                     | Amakusa Nada  | 9.7%                                 |
| Kimotsuki    | 185                             | 450                   | 83             | 1050                     | Kagoshima Bay | 3.1%                                 |

*Akimoto et al. (2009)
†Ministry of Land, Infrastructure, Transport and Tourism
(http://www.mlit.go.jp/river/toukei_chousa/, accessed in Nov. 24, 2017)
‡Average rate of sediment yield of Takase River is assumed to be the same as that of
Midorikawa River.
Figure 2 (Ohta et al.)

- **Geology**
  - Sed
  - Py
  - Acc
  - Gr
  - Mv
  - Mp
  - Fv

- Geographical markers:
  - **Au, Ag deposit**
  - **Cu, Zn, Sn deposit**
  - **As, Zn deposit**

- Study area:
  - Sea of Japan
  - Sea of Japan
  - Sea of Japan
  - Pacific Ocean
  - Japan

- **Figure 2 (Ohta et al.)**
Figure 3 (Ohta et al.)
Figure 4 (Ohta et al.)
Figure 3 (Ohta et al.)

- **Cr (mg/kg)**
  - Land: 7.46, 18.3, 21.8, 34.3, 52.9, 135, 163, 550
  - Sea: 3.11, 16.4, 20.6, 28.7, 41.6, 51.7, 55.9, 77.1

- **Cs (mg/kg)**
  - Land: 0.41, 1.81, 2.18, 3.23, 4.49, 5.70, 6.84, 7.86, 14.9
  - Sea: 0.13, 0.70, 0.98, 1.53, 2.64, 3.90, 4.92, 5.37, 6.74

- **La (mg/kg)**
  - Land: 5.90, 13.2, 14.3, 16.4, 19.1, 22.1, 26.6, 30.3, 194
  - Sea: 2.52, 7.62, 10.2, 12.9, 16.0, 18.1, 19.6, 27.0, 8.11

- **Yb (mg/kg)**
  - Land: 0.49, 0.82, 0.93, 1.24, 1.65, 2.19, 2.68, 4.11
  - Sea: 0.20, 0.47, 0.72, 1.02, 1.34, 1.66, 2.35, 3.39

Legend:
- **Purple**: mafic and ultramafic rocks
Figure 4 (Ohta et al.)
Figure 5 (Ohta et al.)
Figure 6 (Ohta et al.)
Figure 7 (Ohta et al.)

La/Yb

2.49 - 4.98
4.90 - 5.88
5.89 - 8.14
8.15 - 11.21
11.3 - 14.0
14.1 - 18.2
18.3 - 20.8
20.9 - 144

Cr/Ti (×100)

0.10 - 0.28
0.28 - 0.38
0.39 - 0.58
0.59 - 1.14
1.15 - 1.66
1.68 - 2.26
2.27 - 2.95
2.96 - 11.6