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Research article

Using data-driven analysis of geochemical environmental information to infer the environmental impact of closed mines

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ABSTRACT

River sediments have the effect of aggregating geochemical environmental information, such as that related to geological and artificial pollution resulting from mine closure. This information comprises high-dimensional data and is related to the distribution and quantities of elements in river sediments. However, accessing and interpreting this geochemical information can be difficult. This study employed a data-driven analysis that can be mathematically and statistically reduced in dimension. Using high-dimensional geochemical and environmental information on river sediments, this study evaluated the environmental impact of closed mines. Sample for analysis were collected from three rivers. There are differences in the existence of mines and mine wastewater treatment methods in this river. A total of 33 elements were measured in river sediments. Frequency distribution analysis and Principal component analysis revealed that the elements had unique distribution and frequency characteristics in each river catchment. Four environmental factors could be extracted from the relationship of elements due to lower dimension. PC1 was influenced by the land use in the river area. PC2 captured the geological background. PC3 captured the mixing-diluting effect that occurs in rivers. PC4 effectively captured the effects of domestic wastewater and the effects of closed mines. The effects of the closed mines could be confirmed using the PC4 score for the Okawa River and the Akagawa River. By examining the elemental relationships obtained using these mathematical methods, it is possible to infer the effect of geological features and mines on sediment physiochemistry using existing data on river sediments.

1. Introduction

Mines have a major impact on the surrounding environment due to heavy metals contained in the mine wastewater and tailings that are left behind once the mine has closed (Li et al., 2017, Yang et al., 2018a, Yang et al., 2018b). Consequently, the long-term maintenance of closed mines that are no longer viable is an international problem. There are approximately 5000 closed mines in Japan (Mongi et al., 2007a,b). At present, mine wastewater from 80 closed metal mines is properly treated (Sawayama and Tsuchiya, 2018; Tomiyama et al., 2010). The impact of mines and closed mines on local communities and the surrounding natural environment has been extensively reported (Endou et al., 2014). Numerous assessments of the environmental effects of closed mines have been performed, and long-term monitoring of river water quality, surveys of aquatic organisms, and heavy metal pollution of river sediments, river water and soil in these areas have been undertaken (Ueda and Masuda. 2005; Neculita et al., 2006; DeNicola and Stapleton. 2014).

River sediments are defined as rocks, sand and soil that are eroded and transported downstream in river runoff (Ohta et al., 2004; Terashima et al., 2008). As a result, river sediments have characteristics that are similar to the geological characteristics of the rocks in river catchments. Sediments that have been in rivers for extended periods adsorb heavy metals and anthropogenic pollutants from the river water (Aguilar-Carrillo et al., 2018). These river sediments can therefore be considered to contain information on the various environments within the river catchment (Giovanni et al., 2018; Li et al., 2018; Ghosh et al., 2011). However, in order to correctly interpret this high-dimensional geochemical information, it is necessary to characterize the different types of information.

In recent studies, data-driven analyses centered on mathematical statistics have been used to elucidate geoscientific processes related to

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rocks and soil (Kuwatani, 2018; Nakamura et al., 2017; Pujiwati et al., 2021). Studies focusing on distributions and transport of elements in river sediments have also been reported. For example, in a data-driven analysis of river sediments in Sendai City, Miyagi Prefecture, Nakamura et al., 2016 examined geological baselines of anthropogenic heavy metals and the environmental dynamics of heavy metals. Other reports using a similar technique predicted the effects of anthropogenic heavy metals on soil, as well as considering geological contributions (Huang et al., 2018; Singh and Lee. 2015). River sediments and soil, which are important components of the environment, are likely to contain information on the environmental dynamics and distribution of heavy metals. By applying the data-driven analysis, it is possible to compress and extract essential information like relationships of elements in river sediments. Therefore, it is possible to find geochemical findings in river sediments that have not been clarified so far. Previous studies have been applied data-driven analysis to datasets such as river water (Chen et al., 2007), soil (Zhang, 2006) and sediments (Emmerson et al., 1997) to quantify elements. However, there are no discussions on the specific cause like the environmental impact of closed mines used for quantitative evaluation of the relationships between elements.

To evaluate the extent to which rivers with closed mines (with or without neutralization treatment) affect river reaches that are not affected by mines, this study evaluated the environmental impacts of rivers near closed mines by using a data-driven analysis.

2. Materials and methods

2.1. Study area and sampling

River sediments were collected from three rivers in Japan. Figure 1 shows the geological map for each river and the sampling points. The Okawa River area near Kesennuma City, Miyagi Prefecture is formed by the Shishiori River, the Okawa River and the Kamiyama River. The geology of the Okawa river area comprises sedimentary rocks. The Natori River area near Sendai City, Miyagi Prefecture, is drained by the Natori River and the Hirose River. In the Natori River area, the geology of most of the upstream areas of the river basin comprises sedimentary rocks; the remainder of the upstream areas and the middle areas comprises volcanic rocks. The Akagawa River area near Hachmantai City, Iwate Prefecture, is drained by the Akagawa River and the Matsukawa River. The Akagawa River is a tributary of the Kitakami River and the geology of this region comprises volcanic rocks.

The rivers targeted in this study are characterized by having closed mines along their length, or they drain areas where wastewater from closed mines is being treated. There are no major mines along the Natori River, but there are closed mines in the Okawa River area and Akagawa River area (Figure 1). However, the treatment facilities used to treat waste water from the mines in the Okawa River area and the Akagawa River area are different. Gold was mined at the Shikaori mine on the Okawa River; the mine was closed around 1970. As a result of on-site surveys, the existence and type of mine wastewater treatment could not be determined, and the impact of wastewater from the mine on the surrounding environment has not been investigated. The Matsuo mine in the Akagawa River area produced sulfur. The mine wastewater is strongly acidic, so it is neutralized the mine wastewater semi-permanently (Fujii, 1994).

Collection points of river sediments were set upstream and downstream of tributaries or confluences of industrial and agricultural water in river basins. The samples were collected from the river bed from 1 m to the middle of the shore. Samples were collected from 23 sampling points in the Okawa River area, 38 sampling points in the Natori River area, and 18 sampling points in the Akagawa River area.

2.2. Analytical method

After airdrying the river sediments collected from each river, gravel and wood chips etc. were removed and the particle size was adjusted to <2 mm by sieving.

An energy dispersive X-ray fluorescence analyzer (EDX-720, Shimadzu Inc., Kyoto) was used for element quantitative analysis. The quantitative analysis was performed using the absolute calibration curve method described by Yamasaki et al., 2011). Eleven major elements [sodium(Na), magnesium(Mg), aluminium(Al), silicon(Si), phosphorus(P), sulfur(S), potassium(K), calcium(Ca), titanium(Ti), manganese(Mn) and iron(Fe)] and 22 trace elements [vanadium(V), chromium (Cr), cobalt(Co), nickel (Ni), copper(Cu), zinc(Zn), arsenic(As), rubidium(Rb), strontium(Sr), yttrium(Y), zirconium(Zr), niobium(Nb), tin(Sn), caesium(Cs), barium(Ba), lanthanum(La), cerium(Ce), praseodymium(Pr), neodymium(Nd), lead(Pb) and thorium(Th)] were measured in this study.

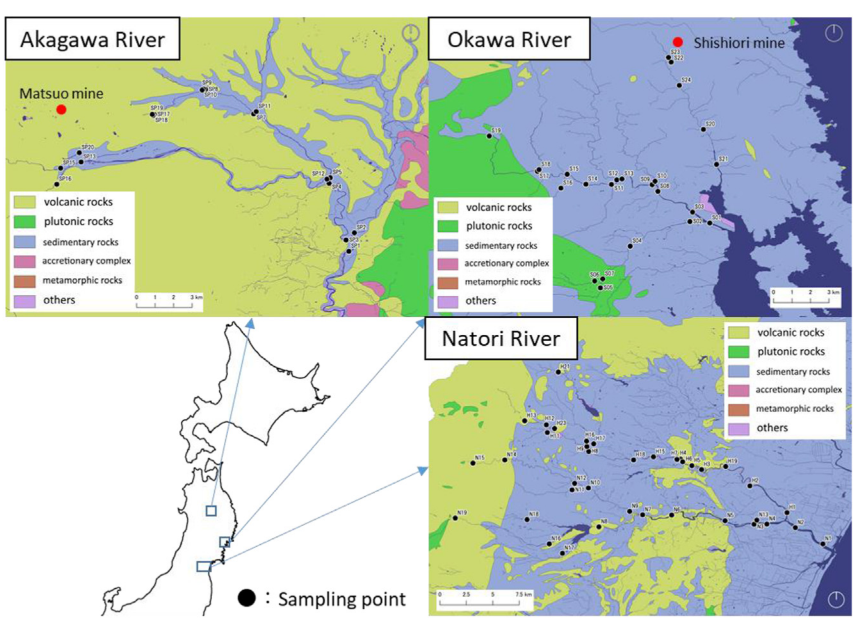

Figure 1. Geological map and location of sampling points in the catchments of this study.

2.3. Statistical analysis

In this study, data-driven analyses consisting of frequency distribution analysis and principal component analysis (PCA) were performed on major and trace elements in the obtained river sediments. These data-driven analyses were calculated using Matlab toolbox.

Frequency distribution analysis is a statistical method that divides a data group into a plurality of regions and shows how much of the data is distributed in each region.

PCA is a widely used type of multivariate analysis to simplify factors hidden in high-dimensional data (Sołek-Podwika et al., 2016; Tahri et al., 2005; Adelopo et al., 2018; Reid and Spencer. 2009). PCA is a technique that can be tolerantly understood by lowering the dimensions of the relationships between the big amount data. Therefore, it is a useful mathematical statistical method to be performed at the beginning of the analysis of complex geochemical data. Mathematically, it is a method of creating synthetic variables, called principal components (PCs) that do not correlate with each other by orthogonal transformation of high-dimensional data. By ignoring synthetic variables with a small variance (contribution rate: Eigenvalues), low-dimensional information of factors hidden from high-dimensional data becomes apparent.

3. Results and discussion

3.1. Element concentration in sediments

The measurement results for 11 major elements and 22 trace elements for each river area are shown in Figure 2. The major elements and trace elements in river sediments are characteristic to each river area. The most abundant element in all rivers was Si followed by Fe and Al. The average Si concentration of samples from the Natori River was 82 wt%, which was higher than that of samples from other rivers. The concentrations of Fe and Al were high in the Akagawa River, with averages of 10 wt% and 15 wt%, respectively. In addition, the Ca concentration from the Akagawa River was higher than that in other rivers. The concentration of Sr and Ba were highest in the Okawa River. V and Zr concentrations were highest in the Akagawa River. The concentration of trace elements in the Natori River tended to be lower than those in the Okawa River and the Akagawa River.

3.2. Frequency distribution of elements in sediments

The frequency distribution of major and trace elements in river sediments from each river area is shown in Figures 3 and 4. Figure 3 shows

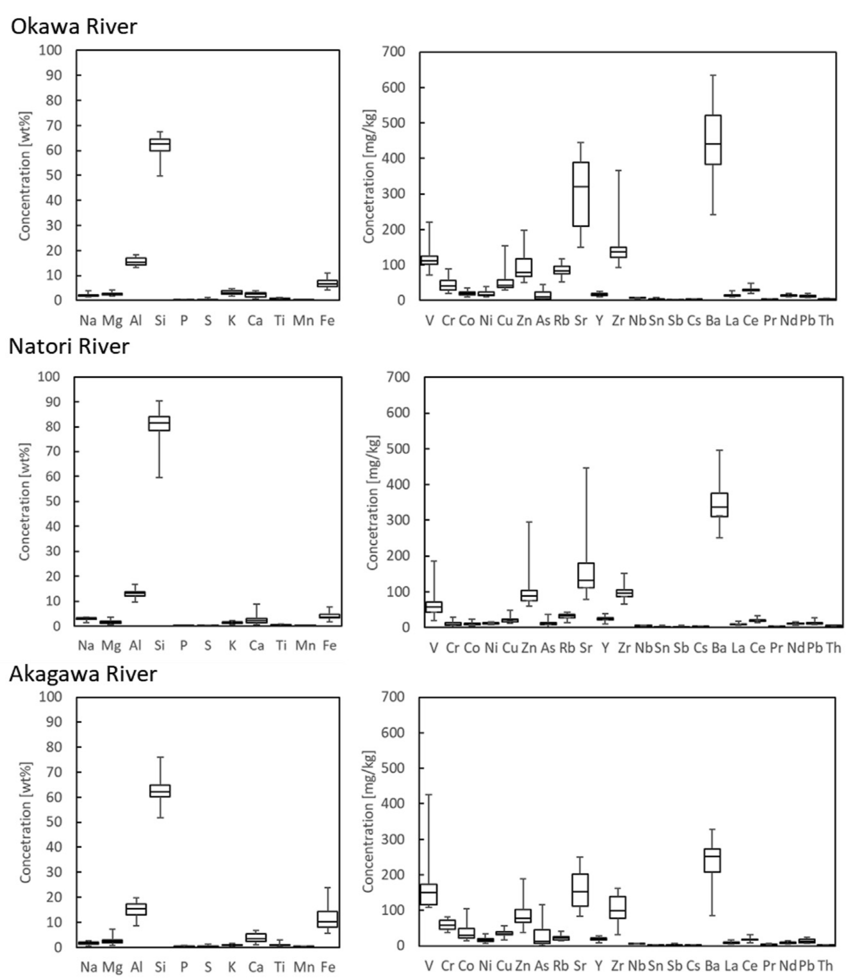

Figure 2. Boxplot of major elements and trace elements contained in river sediments. (The boxplot shows the maximum, 75%, median, 25% and minimum values, from the top down.)

the frequency distribution of 11 major elements, and Figure 4 shows the frequency distribution of 22 trace elements.

From Figure 3, Na and Si were frequently present at high concentrations in the Natori River. Mg, Al, P, S, Ti, Mn and Fe showed similar trends in the Okawa River and the Akagawa River. In addition, these elements were present at high concentrations in some river sediment samples. Due to differences among rivers, Ca was not encountered as frequently as other elements and was present at relatively low concentrations in the Akagawa River. K concentrations were highest in the Akagawa River, followed by the Natori River and Okawa River, in order.

The frequency distribution of trace elements in river sediments in each river area was also determined. From Figure 4, the frequency of V, Cr, Co, Ni, Cu, Sn and Sb are present low of frequency distribution in the Natori River. Rb, Y, Zr, Nb, Cs, La, Ce and Rb increased until minimum value. However, the frequency of these trace elements peaked in the range of 2–3. The frequency of other trace elements generally decreased from low concentrations to high concentrations.

The frequency distribution of the trace elements in river sediments from the Okawa River and the Akagawa River showed a decreasing tendency with increasing concentration. Compared to the other rivers, Cr and Ni were distributed only in low concentrations in the Natori River. In addition, the sediment concentrations of V, Co, As, Sb and Pr were high in the Okawa River, and those of Cu, Rb, Sr, Zr, Nb, Sn and La were high in the Akagawa River.

3.3. Dimensional compression using principal component analysis

PCA was performed for 11 major and 22 trace elements in river sediments (79 samples) from the Okawa River, the Natori River and the Akagawa River. Figure 5 shows the results obtained for the low-dimensional data by PCA. The PC had 33 dimensions, and the total sum of eigenvalues from PC1 to PC4 is 0.73, which indicates that 73% of the information in the dataset is aggregated in dimensions PC1 to PC4.

Figure 6 shows the relationship between the eigenvectors and the major and trace elements in PC1 to PC4. In PC1, only Na and Si were positive; all of the other elements were negative. In PC2, trace elements such as Nb were positive, and major elements such as Mn and Fe were negative. In PC3, S, As and Sb were positive, and Sr was negative. In PC4, Zn and Y were positive.

The potential relationships between elements in river sediments was inferred from a data-driven analysis (frequency distribution and PCA) based on the concentrations of 33 elements in river sediments. The potential relationships among elements was considered the meaning of each PC and the differences in each river area. Figure 7 shows the relationship between PCs obtained from the PCA and a plot of the scores for PC 1 to 4. The relationship between PC1 and PC2 in Figure 7 was plotted at the same position in each river area, to some extent. In the Okawa River, PC1 was negative and PC2 was positive, in the Natori River, PC1 was positive and PC2 ranged from -2 to 2, and in the Akagawa River, PC1 was negative and PC2 was negative. The axes for PC1 vs PC3, PC2 vs PC3 and PC2 vs PC4 show differences in the distributions of plot scores between the Natori River and the other river areas. From the distribution of PC2 and PC3-PC4, the scores obtained for the Natori River are distributed near the center, while the scores obtained for the Okawa River and the Akagawa River are distributed on the left and right. PC2 is therefore understood to indicate the difference in characteristics between the Okawa River and the Akagawa River. For PC3 and PC4, a lot of data points are distributed in the center, and some of the data points for each

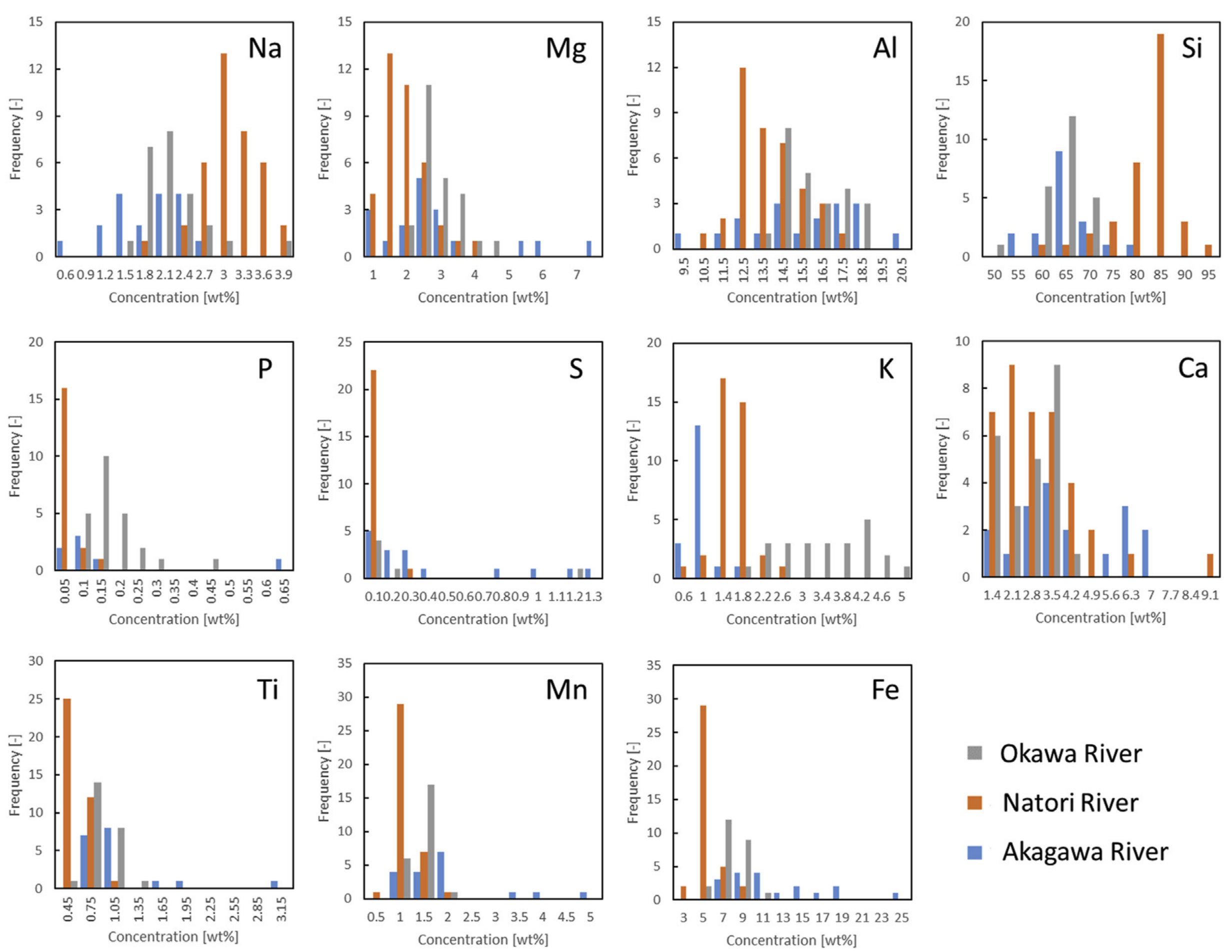

Figure 3. Frequency histogram of major elements in the three river catchments.

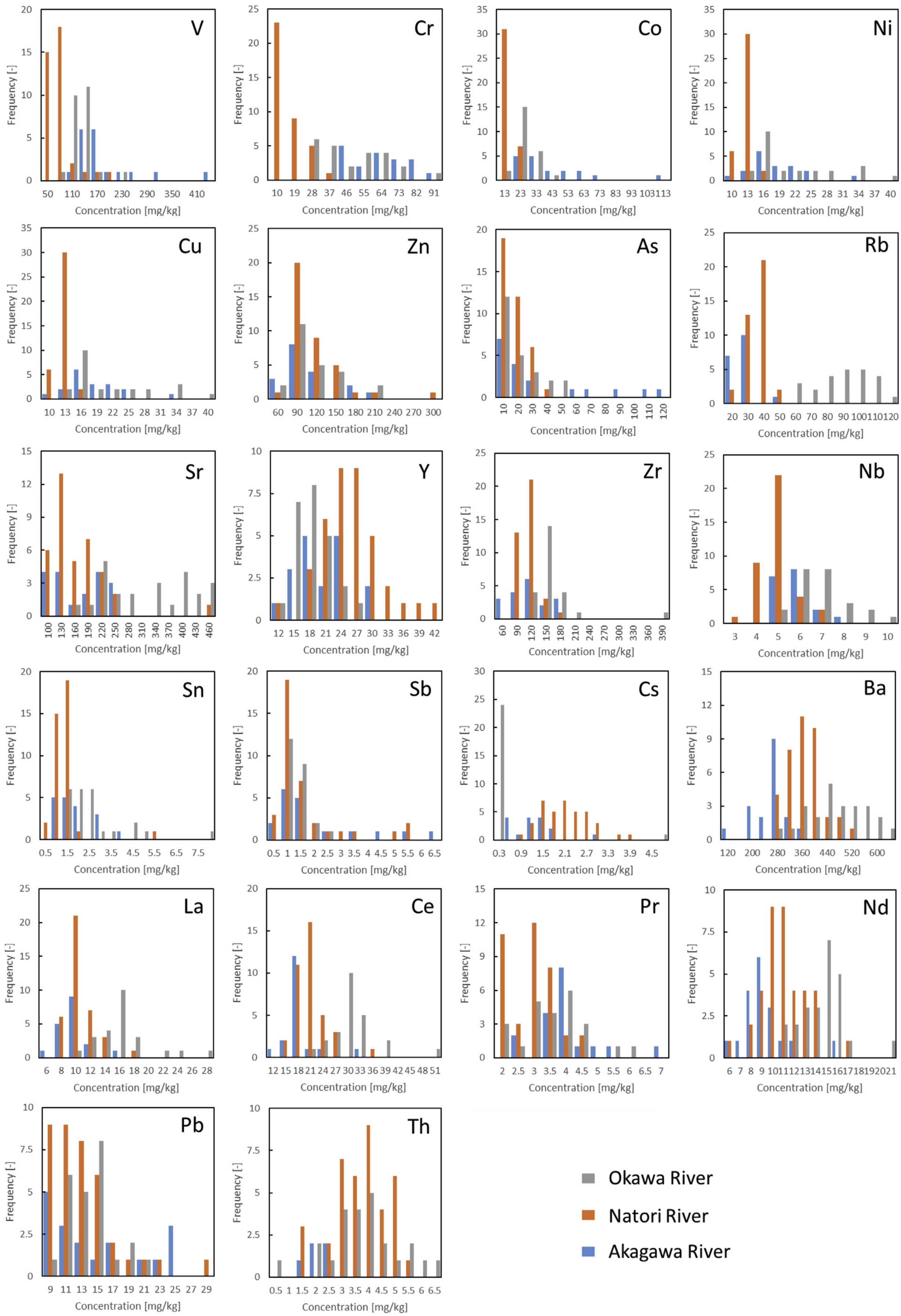

Figure 4. Frequency histogram of trace elements in the three river catchments.

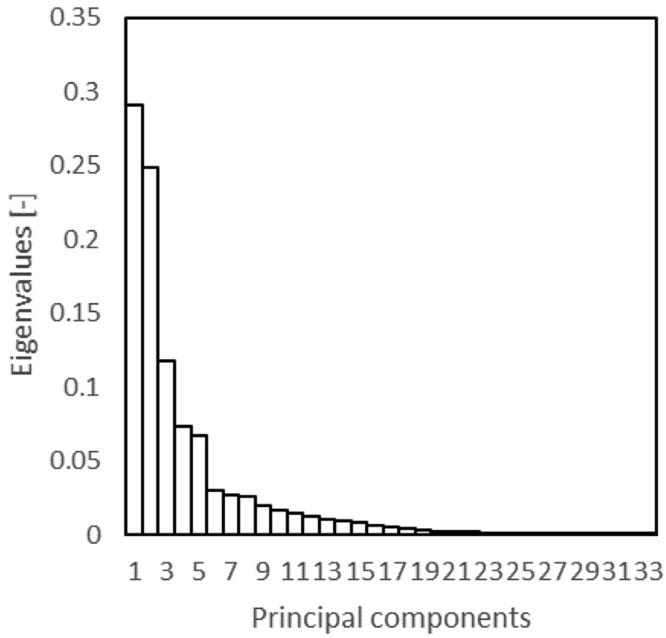

Figure 5. Eigenvalues of the PCA of element concentration in all sediments. The horizontal axis indicates the dimension, while the vertical axis indicates the eigenvalues, in which the sum is normalized to 1.

river have high positive or negative scores. It is considered that PC3 and PC4 do not capture the characteristics of each river area, but rather the relationship among element in the river area.

Figure 8 shows the scores for each PC on a map obtained from the PCA. As for the distribution of scores on the map of PC1, the scores for the Natori River are higher than those for the other river areas. Compared with the scores for the Okawa River and the Akagawa River, PC2 has a high score for the Okawa River and a low score for the Akagawa River. The score for the Natori River is almost 0 in the whole area. These findings indicate that the difference between the Natori River and other areas by PC1, and the difference in the characteristics of the Okawa River and the Akagawa River by PC2, can be confirmed from the map shown in Figure 7.

The scores for PC3 decreased from the upstream reaches to the downstream reaches, some places where the score was high in the Okawa River and the Akagawa River. No such trends were observed for the Natori River. The score distribution for PC4 showed that some points in the Okawa River and the Akagawa River areas were high. The scores obtained for the Natori River were higher overall than those for the other river areas.

3.3.1. Characteristics of PC1

PC1 (Eigenvalues: 29%), which is the most informative of the PCs, is characterized by the elemental relationship of Na and Si. The score clearly shows the difference between the Natori River and other river areas. From Figure 2, the Si concentration of the Natori River is 10–20 wt % higher than that in other river areas. The median values for Na in each river area is 2.0 wt% in the Okawa River, 2.9 wt% in the Natori River and 1.8 wt% in the Akagawa River. The sodium (Na) concentration was relatively high in the Natori River, implying that the concentration of the other major elements is higher in the Okawa River and the Akagawa River.

Figure 9 shows the land use of each river area (ALOS/AVNIR-2) (ALOS Research and Application Project, 2020). The Natori River flows through Sendai City and most of the area around Sendai City is urban. The land in the Okawa River and Akagawa River catchments contains large areas of deciduous broadleaved forest, coniferous forest and rice paddy fields, i.e., land use types more commonly associated with mountainous areas than urban areas. In the Natori River catchment, which has a vast urban area, the rivers are well maintained, so is difficult for rocks and soil to flow into the river from surrounding environments, is little supply of earth and sand from the river. Therefore, it is considered that Si is the main element, and Na is associated with human activity in the Natori River catchment. The Si and Na are likely to remain at high concentration. Other elements are also likely to be retained in this downstream environment in this area. Various soils and minerals in the Okawa River and the Akagawa River tend to be readily transported down

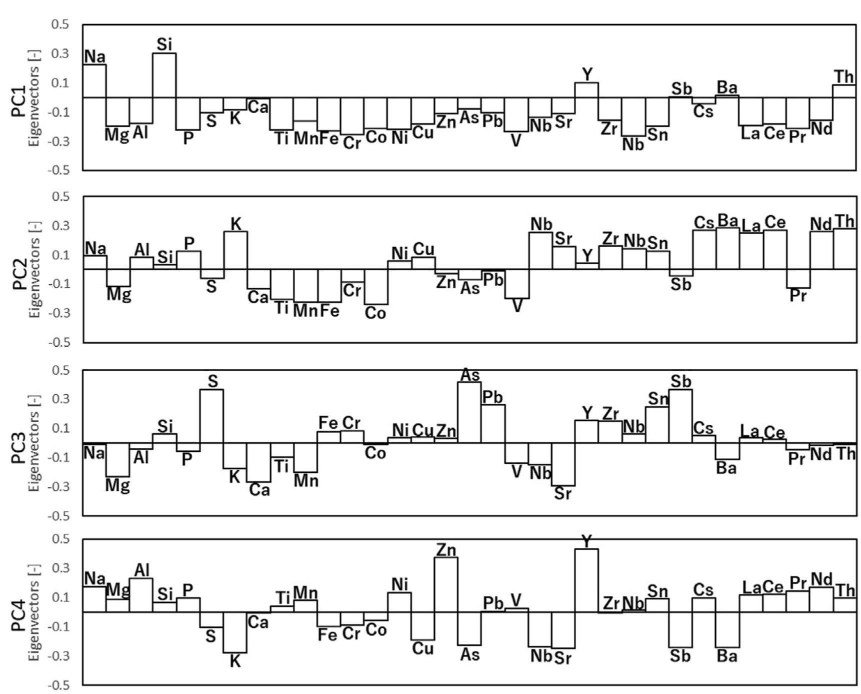

Figure 6. Eigenvectors of the first four PCs for major elements and trace elements. The horizontal axis indicates elements, while the vertical axis indicates the value for each element that constitutes the eigenvector.

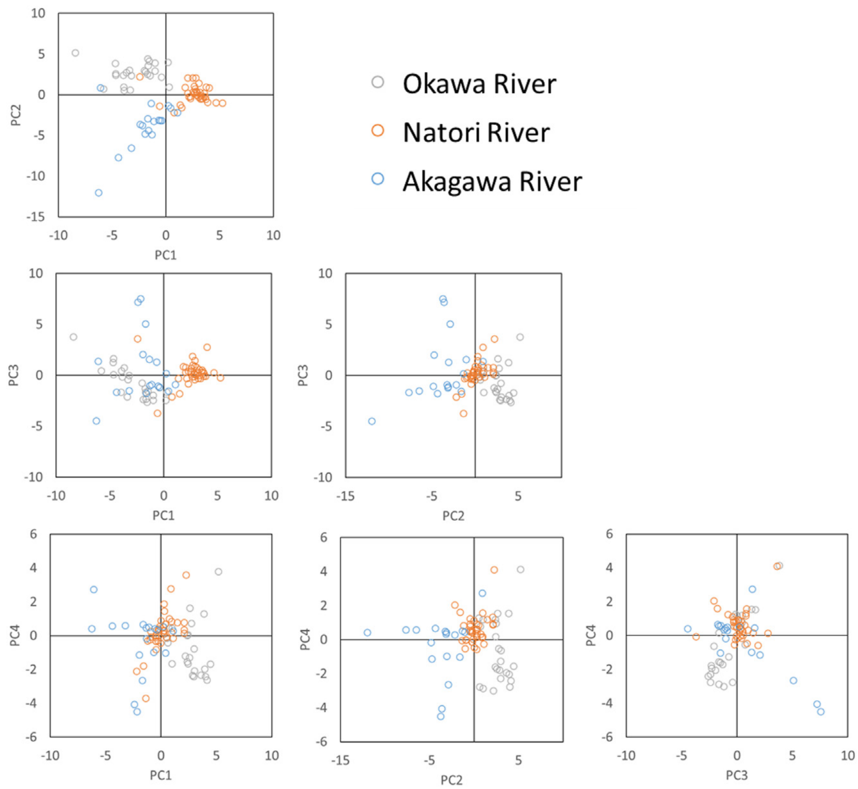

Figure 7. Biplots of first four PCs for all sediments. The sediments are classified by river area.

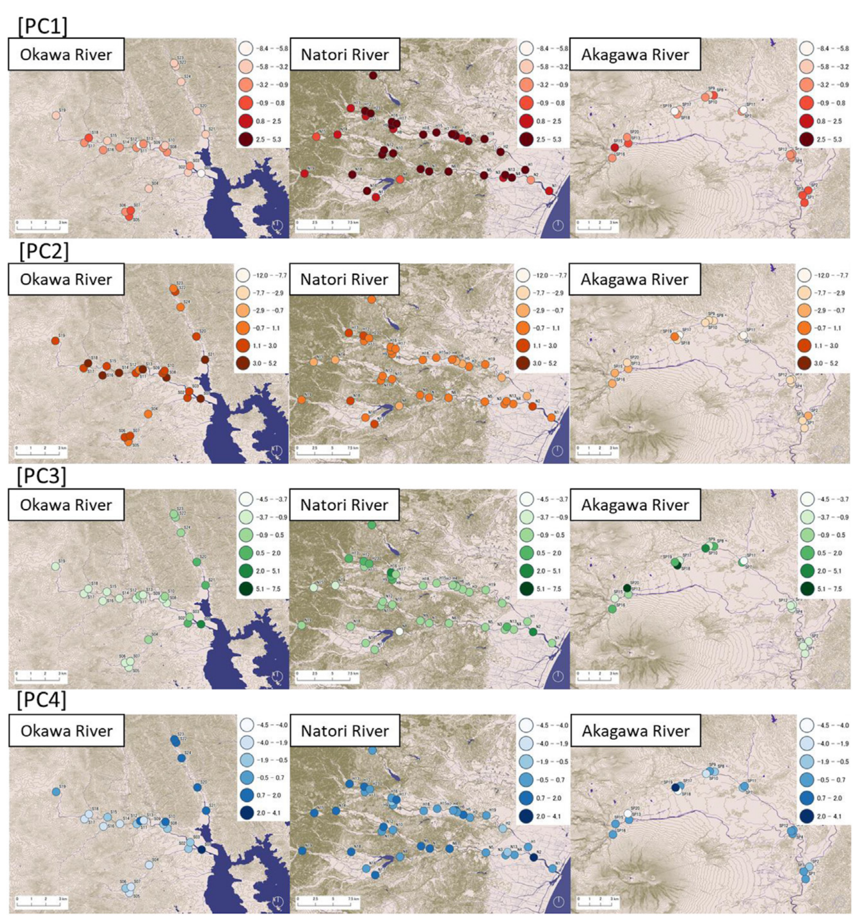

Figure 8. Map of eigenvalues for PC1 to PC4. The strength of the color depends on the score by PCA.

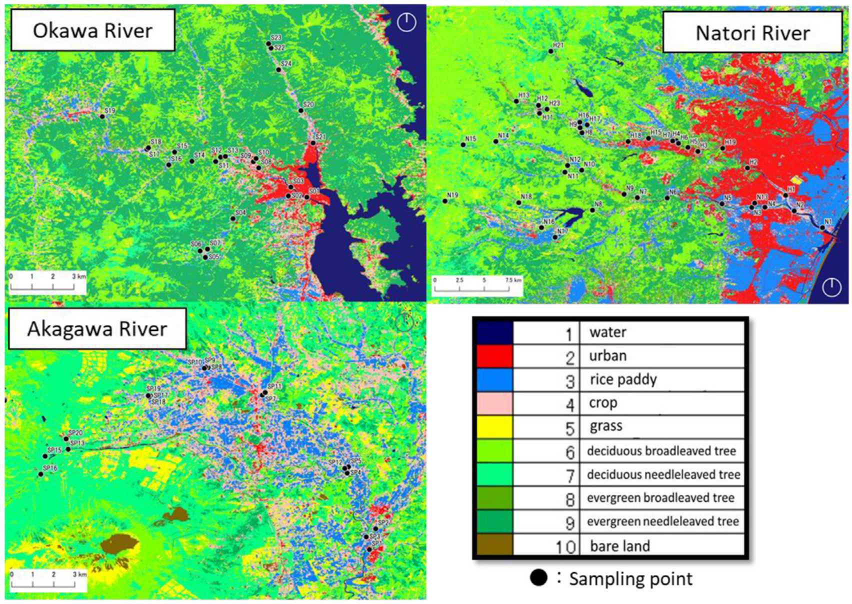

Figure 9. Map of land use in the catchments of the study area.

rivers from mountainous areas and paddy fields in runoff. These areas have relatively high concentrations of other major elements. Areas with many paddy fields adjacent to the Natori River had low scores, which was similar to the findings obtained from similar areas along the Okawa River and the Akagawa River. This trend of low scores also suggested that PC1 captured information on land use.

3.3.2. Characteristics of PC2

As shown in Figure 7, PC2 (Eigenvalues: 25%) was considered to separate the characteristics of the Okawa River and the Akagawa River. The map of PC2 scores in Figure 8 shows that scores were high for the Okawa River and low for the Akagawa River. The scores for the Natori River are between those for the Okawa River and the Akagawa River. PC2 is considered to have captured the influence of the geological background (Figure 1). The geology of the Okawa River is dominated by sedimentary rocks, and that of the Akagawa River is dominated by volcanic rocks. The Natori River has a mixture of these geological backgrounds (mountainous areas: volcanic rocks, coastal areas: sedimentary rocks). As for the geological characteristics of PC2, based on the relationship between each element obtained by PCA, it is considered that trace elements and major elements such as Mn and Fe are important. This relationship was the same trend as the content of each element in Figure 2.

The median value for Mn was 0.1 wt% in the Okawa River, 0.09 wt% for the Natori River and 0.16 wt% in the Akagawa River, respectively. The median value for Fe was 6.8 wt% for the Okawa river, 3.8 wt% for the Natori River and 10 wt% for the Akagawa River. Also, the trace elements of Sr and Ba in the Okawa River are characterized potential of PC2 and their contents are higher than those in other river areas. The increase in the trends for these specific trace elements is considered to be due to mineralization caused by geological effects mineralization is a phenomenon in which trace elements aggregate to form rocks. This phenomenon occurs under natural conditions and on a large scale results in deposit formation (Neiva et al., 2019). In the actual environment, there are cases where the concentration of trace elements in the river basin around the mine increased due to the mineralization, which had an environmental impact over a wide area too (Kosai et al., 2021, Peter and Patrick, 1994). Willy at al. 2012 suggested that it can provide information on the spatial distribution patterns of heavy minerals in stream sediments, playing an important role in determining their likely geographical origin in Japan.

Gold and sulfur were mined in the catchments of the Okawa River and Akagawa River, respectively. It is therefore considered that mineralization affects the distribution of trace elements in each river area. Further, it is considered that the geological characteristics of both regions were captured by PC2.

3.3.3. Characteristics of PC3

PC3 is characterized by positive vectors for S, As and Sb, and negative vectors for Ca and Sr. These five elements do not show the same trend in characteristics potential of this PC depicted in the frequency distribution in Figure 4 and the biplots of the PCA in Figure 7. In Figure 8, for the map of PC3, the score increases from upstream to downstream reaches for the Okawa River, and decreases from upstream to downstream reaches for the Akagawa River. In the Natori River, the score increased in some of the upstream areas and some of the downstream areas. It is considered that these increases were affected by influents from hot springs and seawater in the Natori River.

Various factors affect element concentrations in river sediments from the upstream and downstream river reaches. In addition, dilution due to the confluence of rivers also affects these concentrations. The change of the score from the upstream to the downstream river reaches is considered to indicate the mixing-diluting action that occurs in these river areas. These rivers areas are canalized. It is unlikely that geological rocks, sediments, soil, etc., that can occur in the natural environment, are eroded and transported downstream in the rivers. In addition, the Natori River has dams for hydraulic control of runoff for Sendai City and other cities. The dams in the upstream reaches have been shown to affect the concentration and distribution of major and trace elements in river sediments (Neiav et al., 2016). Dams also have the effect of trapping river sediments, which can become agitated due to inflows from the surrounding environment. The mixed river sediments in dams can have a similar composition, and water and sediments from these dams are released regularly. The distribution of the elements contained in river sediments below dams is thus unlikely to change significantly, which may explain why the score was uniform for the Natori River.

When hot spring water is released into a river, mixing of river water and hot spring water will occur at the discharge point (Nakamura et al., 2016; He et al., 2019). At sampling points where the score increased relative to other sampling points, it is considered that river water is mixed with hot spring water and seawater.

3.3.4. Characteristics of PC4

PC4 was similar to PC3 in terms of the distribution of the PCA scores in Figure 8. The characteristic of PC3 is that there are places where the score is high for some rivers or some reaches in the same river area. On the Okawa River, the scores obtained at sampling sites in the northern part of the map are higher than those for other rivers reaches on the same map. On the Natori River, the score is relatively high compared to other rivers, and the score is also high at the confluence of rivers. On the Akagawa River, the score is high at one point in the north.

In Japan, a drainage water from mines is acidic and contains heavy metals. Even after the mine is closed, the flow rate of heavy metals is regarded as a problem caused by the pit waste water, sludge and slag (Ishiyama et al., 2008; Mongi et al., 2007a,b). Also, the environmental impact of mineral wastewater has become a significant problem worldwide (Neiav et al., 2016).

Large amounts of acidic mine wastewater are typically released into areas that surround active and closed mines. This is because, in the case of metal mines, minerals such as chalcopyrite, pyrite, sphalerite and galena remain inside and outside the mine. Heavy metals contained in ore wastewater are oxidatively dissolved by sulfide minerals due to the influence of water and air that have permeated deep into the mine from the ground surface. For example, sulfuric acid is a strongly acidic waste that leaches from mines or mining areas (He et al., 2019; Montes-Avila et al., 2019). Mine wastewater often continues to leach from mining areas, even after the mine is closed, and treatment must be continued semi-permanently. In addition, minerals remaining inside and outside the mine may become dissolved in runoff from rainwater, which may then affect the surrounding environment (Mine Pollution Control, Japan Oil, Gas and Metals National Corporation, 2020). In fact, mine drainage and various elements from closed mines have been known to enter rivers in runoff, requiring environmental monitoring and earth observation techniques to mitigate against their effects on the environment (Osán et al., 2002).

Also, it is necessary to consider the impact of domestic wastewater on rivers. From the land use data shown in Figure 9, it can be seen that many of the sampling points along the Natori River are in urban areas. In Japan, domestic wastewater is properly treated and discharged by local governments (Tsuzuki et al., 2010). However, some domestic wastewater is discharged into rivers from urban areas. The domestic wastewater contains various chemicals (Magowo et al., 2020), and it is considered that this effect was captured by PC4 in the Natori River. In other river areas, the influence of domestic wastewater was small, and it is considered that it did not affect the PCA. PC4 is therefore suggested to be an indicator of inflows from closed mines and domestic wastewater in river areas.

4. Conclusion

This study examined high-dimensional data for 33 elements in river sediments from the Okawa River, the Natori River and the Akagawa River in Japan. These rivers are characterized by the presence or absence of closed mines. Using the data, high-dimensional geochemical environmental information was extracted from frequency distribution and PCA. The characteristics of each river area and the environmental impact of closed mines were then considered based on potential elemental relationships and PC scores depicted on maps.

For the Okawa River, the concentrations of the trace elements Sr and Ba were higher than they were in other areas. Si of the Natori River and Mn and Fe of the Akagawa River were also widespread in river sediments.

PCA was used to examine the relationships among, and contributions of, four PC elements of river sediments. These PC elements were low-dimensional and captured 73% of geochemical and environmental information. PC1 was mainly influenced by the land use in the river area. PC2 captured the geological background. In addition, it was possible to empirically estimate and discriminate among the characteristics of the sediments from the three river catchments using the plots of the PC1 and PC2 scores. PC3 captured the mixing-diluting effect that occurs in rivers. PC4 effectively captured the effects of domestic wastewater from urban areas and the effects of closed mines on element concentrations in sediments.

The effects of the closed mines, inferred by extracting high-dimensional geochemical information, could be confirmed using the PC4 score for the Okawa River and the Akagawa River. Therefore, the data-driven analysis performed in this study successfully extracted the fundamental relationships that exist among the 33 elements contained in river sediments. By examining the elemental relationships obtained using these mathematical methods, it is possible to infer the effect of geological features and mines on sediment physiochemistry using existing data on river sediments.

In order to propose measures necessary for the rehabilitation of closed mines, it is necessary to clarify which factors related to closed mines are captured by PC4. In the future, by using Zn and Y as index elements that had the most marked effect on PC4, we intend to evaluate both the range of effects of closed mines and the changes in the surrounding environment over time.

Declarations

Author contribution statement

Kengo Nakamura: Conceived and designed the experiments; Performed the experiments; Analyzed and interpreted the data; Contributed reagents, materials, analysis tools or data; Wrote the paper.

Yoshishige Kawabe: Performed the experiments; Wrote the paper.

Komai Takeshi: Conceived and designed the experiments; Wrote the paper.

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Data availability statement

Data included in article/supplementary material/referenced in article.

Declaration of interests statement

The authors declare no conflict of interest.

Additional information

No additional information is available for this paper.

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