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Cu behaviors and effects of mine drainage in Kosaka River, Hokuroku mining district, Northeast Japan

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Abstract: Focusing on the origin of the heavy metal, this study aims to build an imputed method to estimate the heavy metal content in river water by making a distinction between the heavy metal of natural origins and that caused by human activities. Supported by GIS, Kosaka watershed within the Hokuroku basin was divided into several sub-watershed polygons and the outflows of water and Cu were calculated for each polygon. Compared with the natural origin, the dominant Cu emissions affected the river water more significantly in local. Based on the mass balance closure, the heavy metal content of Cu in the Kosaka River was estimated by the conflux accumulation of tributaries and mine drainages. The estimated Cu concentrations were checked by comparing with the actually measured values at monitoring points along the Kosaka River and the results are coincidence with each other in general. It is revealed that the mainstream water quality could be estimated by seizing the water quality of upstream tributaries and human drainages.

Key words: mine drainage; river water; heavy metal; geographic information system (GIS); Kosaka watershed

1 Introduction

Mine drainage is a complex of elements that interact to cause a variety of effects on river water quality. The overall effect of mine drainage may be dependent on the discharge volume, pH, the flow (dilution) rate, the high concentration of dissolved heavy metals, and some other factors [1,2]. Heavy metals concentration can increase the toxicity of river water. For example, zinc, copper and arsenic, which may be present in mine drainage, are toxic at extremely low concentrations [3,4]. Most of the metal mines in Japan had been already closed. However, there is still mine wastewater generating from the depositional remaining or mine waste residue of the abandoned mines. These kinds of drainages need proper treatments before discharge. In the other hand, besides the heavy metal eduction caused by human activities, there are naturally occurring rocks or soils with heavy metals enrichment which may produce high heavy metal accumulations in the source water to streams when these rocks or soils react with rainwater or groundwater. This kind of natural water itself might be toxic and needs proper treatment. The heavy metal contents or water quality in one river can be regarded as the results affected synthetically by both precipitation under the natural geological environment and emissions by the human activity.

Works on the elements concentration distribution on the ground surface, carried out with the attention given to the geochemical maps dealing with the riverbed deposits which represent the geological features existing in the basin, have suggested that the concentration distributions of many elements have close ties to the geological environment [5,6]. The origin of mining and mineral-processing wastes is closely related to the formation of the target resource or minerals [7]. The geological composition in a basin is an important factor to river water quality in natural state. However, for different geological types or rocks, the effects on water quality are different [8]. For the heavy metals present in rocks, the whole-rock contents have relation with the hydrochloric acid dissolution contents while have nothing to do with the water dissolution contents [9]. The previous works have indicated that the river water quality reflects the geological features present in its own basin [10]. WILLIAMS et al [11] studied 270 mine discharges in the Stonycreek River Basin and found

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that the discharges generally had high concentrations of acidity, iron, manganese, aluminum, and sulfate. Water quality is thus severely degraded when mine discharges enter streams and rivers [12]. JAMES et al [13] reported that seven sites within the Allegheny and Monongahela River Basins showed significant trends in sulfate concentration from 1965 to 1995, and these trends appear to be related to the increase or decrease of coal production in the two basins from 1965 to 1995. The water quality cannot be estimated simply by the consideration of geological features alone [14], especially in mining areas where mine drainage from abandoned mines still keeps the major problems in streams and rivers.

The water quality reflects not only its origin but also what it encounters along its flow path. One of the most challenging works of water management is to identify the main anthropogenic pollution sources and to assess their downstream environmental impacts [15]. The primary objectives of the present study were, therefore, to access and understand the primary natural (geological features) and human factors (mine drainages) that affect water-quality conditions, so as to determine proper method for estimating the heavy metal contents in river water, by looking into the sources of heavy metals and making a distinction between the heavy metal of natural origin that caused by human activities.

2 Experimental

2.1 Study area

The Hokuroku basin is one of the most popular mining areas in Japan, located in Northeast Japan (Fig. 1). The basin area is about 178.5 km^2, and the length of the mainstream of the Kosaka River is about 20 km. It is one of the most eminent black ore belts in Japan with hundreds of years' of mining history. There were many mines on the north-shore of the Kosaka River, and more than 10 of them were abandoned mines in which silver, lead, zinc, copper and other minerals had been produced before. At present, the mine wastewater management is still continuing in the Furutobe mine and Ainai mine, and the mine wastewater has been properly treated before going into the Kosaka River. Besides, for the Kosaka refinery still operated as metal recycling factory, the water drainage is also poured into Kosaka River. Six monitoring points (P1−P6) were set from upstream to downstream along the Kosaka mainstream (Fig. 1). For P1, located in the upper reaches of the Kosaka River, the watershed is fully covered by forestry, and it has the least impact on mines or other human activities. The river water here can be regarded as in natural state. The Furutobe mine and Ainai mine locate in the watershed between P1 and P2, and the wastewater drainage mixes into the Kosaka River here. There are households and

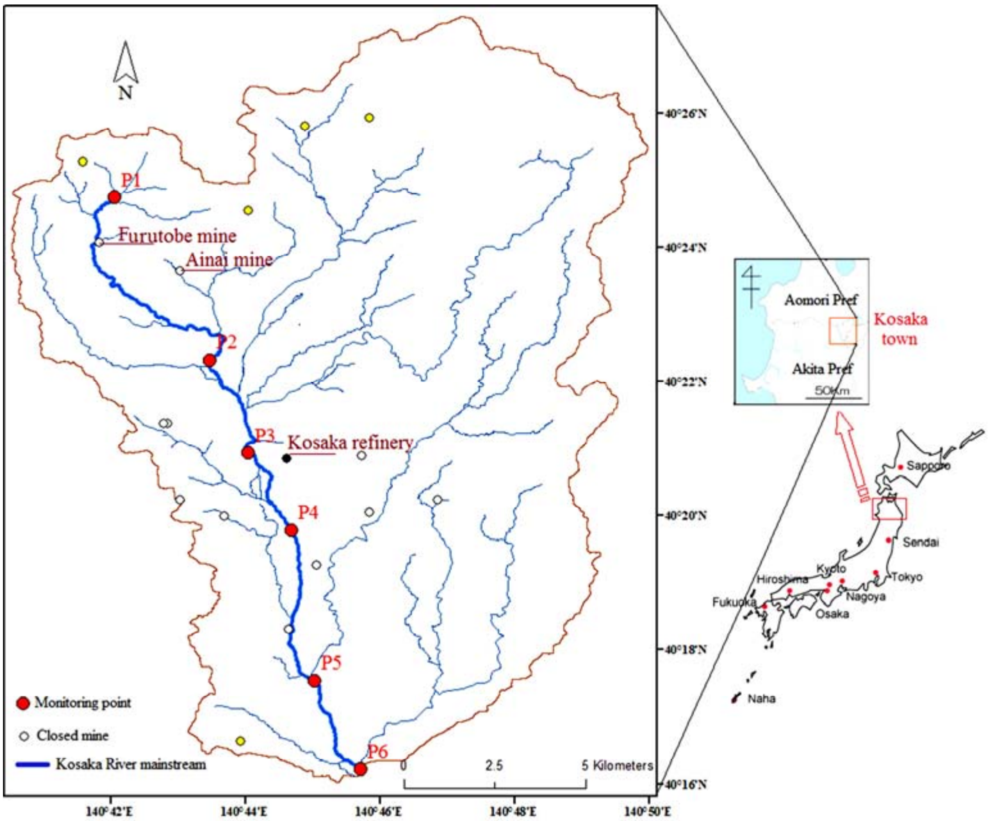

Fig. 1 Scope of study area

paddy fields along downstream from P3. The wastewater drainage from the Kosaka refinery flows into river segment between P4 and P5. The heavy metals in the mainstream are derived from both the anthropogenic origin and natural origin. It is possible to capture the effects of the geological features or the abandoned mines for each tributary watershed by the investigation carried out in this kind of area.

2.2 Watershed segmentation

Supported by ArcGIS 10 (ESRI), the method of hydrology modeling based on digital elevation models was applied to generating stream network, and to dividing sub-watershed and generating basin boundary automatically. The appropriate adjustments for the generated stream network and watershed polygons were done to make them fit well with the real present rivers and the current field situation (Fig. 2). Topology check and modifications were then carried out to make sure that there is no topology error in watershed polygon layer. The 6 monitoring points were set up at the confluences (Fig. 1). The small tributary polygons were merged into the large conform to the watershed of each monitoring point in the mainstream. The tributary watershed polygons labeled 101 and 102 are the area for the water gathering of P1; the tributary watershed polygons named by the labels beginning with the number 2, together with the watershed area of P1, compose the river basin of P2; and the watershed area of P3 consists of the river basin of P2 and the tributary watershed polygons named by the labels beginning with the number 3, so on for the other downstream monitoring points (Fig. 2).

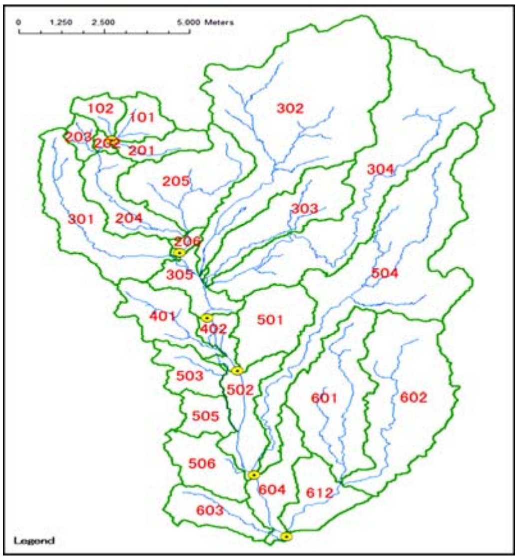

Fig. 2 Sub-watershed division of whole Kosaka watershed

2.3 Water quality analysis

Water samples were collected both in snow-melting season in April and dry season in July in 2009. Six water samples from the 6 monitoring points along the Kosaka mainstream and 18 water samples from the tributaries ahead where they inflow into the mainstream, were filtered on site using a 2 mm sieve to separate out the larger particles. 0.7% HNO_3 was added into each sample to prevent the metal ion from hydrolysis precipitation. The general content of Cu was analyzed by ICP/MS (Perkin Elmer Sciex). The catalytical data quality was guaranteed through the implementation of laboratory quality assurance and quality control methods, including the use of standard operating procedures, calibration with standards, analysis of reagent blanks, recovery of known additions and analysis of replicates.

2.4 Water flow rate

The stream flow rates of the 6 monitoring points along the Kosaka mainstream were actually measured by multiplying river cross section and the average water flow velocity, both in snow-melting season and dry season. Since the river water comes from precipitation in the watershed, the water flow rate of the rivers is considered to be directly proportional to the river basin area. The water-harvesting quantity per unit area between two monitoring points was calculated through dividing the increased water flow rate by the increased watershed area. For a tributary polygon, multiplying the water-harvesting quantity per unit area by the polygon area, the water flow rate could be obtained. The water flow rates at the monitoring points however are affected not only by the precipitation but also by the water intake and drainage. It is necessary to deduct the part that caused by human activity from the actually measured water flow rate between every two monitoring points, so as to acquire an accurate estimate of the relationship between the watershed area and the precipitation. By field survey and the consultation with authorities, the water use of the study area was investigated, including the water intake of paddy fields and the drainages of mines and the Kosaka refinery. Based on the revised water flow considering the man-made effect, the stream flow rate of each tributary was calculated by the weighted average apportionment of interzone water increase between two monitoring points using watershed area of each tributary.

2.5 Heavy metal flow of human drainages

Considering the human-derived heavy metals, the following data were acquired by querying the municipality publishment and gathering information from autonomous bodies: 1) the monthly data of the Furutobe mine and Ainai mine, about the heavy metal concentration in mine waste water and the water discharge quantity before and after treatment, between

April 2007 and March 2009; 2) the year average heavy metal emission data in 2008 gained by the Pollutant Release and Transfer Register (PRTR) of the Kosaka refinery. The waste water discharge of the Kosaka refinery to the Kosaka River was 11×10^6 t/a. For each waste water emitter, unit time discharges of heavy metals and waste water were calculated. The water discharges and their heavy metal concentrations of the human drainages as well as other polygon information are shown in Table 1.

2.6 Heavy metal content estimation in river water

To analyze the river behavior from the viewpoint of its component tributaries in the watershed, the study area was divided into several polygons based on the tributaries that flow into the Kosaka mainstream (tributary polygons) and the watershed boundaries of the monitoring points in the mainstream (mainstream polygons). The water quantity as well as the heavy metals that flow into the mainstream from the tributaries was calculated for each polygon. The dissolved heavy metal flows and accumulates from the upstream to downstream as the river water flows, and the heavy metal content together with the flow rate changes at junctions. The water flow rate and the heavy metal content at one point of the mainstream could be determined as the sum of those come from all the polygons in the upstream. As shown in Fig. 3, polygons A and B are two tributary polygons in the watershed, each with a tributary. Polygon C is a mainstream polygon. The heavy metal content and the water flow rate at point d in the downstream can be calculated as the sum

Table 1 Polygon information

Section	Polygon number	Attribution	Polygon area/ km^2	Rice field area/ 10^5 m^2	Water use for rice field/($t\cdot min^{-1}$)	Anthropogenic emission facility	Drainage quantity/ ($t\cdot min^{-1}$)	Cu concentration of drainage/ ($g\cdot L^{-1}$)
--P1	101	Tributary	2.34	0.00	0.00			
	102	Tributary	1.64	0.00	0.00			
	P1							
P1−P2	201	Tributary	2.23	0.00	0.00			
	202	Mainstream	0.43	0.00	0.00			
	203	Tributary	1.18	0.00	0.00			
	204	Mainstream	4.70	20.52	4.56	Furutobe mine	0.23	8.60
	205	Tributary	8.91	9.22	2.05	Ainai mine	2.51	2.61
	206	Mainstream	0.55	24.00	5.33			
	P2							
P2−P3	301	Tributary	9.67	19.43	4.32			
	302	Tributary	27.76	97.00	21.56			
	303	Tributary	7.01	22.62	5.03			
	304	Tributary	22.03	43.54	9.68			
	305	Mainstream	3.16	56.09	12.46			
	P3							
P3−P4	401	Tributary	4.76	10.33	2.30			
	402	Mainstream	1.85	0.00	0.00			
	P4							
P4−P5	501	Tributary	5.62	0.00	0.00	Kosaka refinery	21.00	48.02
	502	Mainstream	4.71	0.00	0.00			
	503	Tributary	2.94	0.00	0.00			
	504	Tributary	22.59	37.57	8.35			
	505	Tributary	2.28	0.00	0.00			
	506	Tributary	4.20	1.10	0.24			
	P5				0.00			
P5−P6	603	Tributary	3.67	10.00	2.22			
	601+602	Tributary	29.55	152.45	33.88			
	604	Mainstream	4.46	0.00	0.00			
	P6							

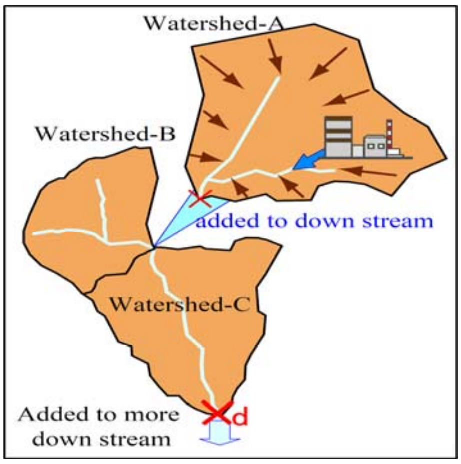

Fig. 3 Sketch of heavy metal flows

of those come from the two tributary polygons A and B, and those come from the mainstream polygon C.

Differentiating the supply sources, the water and the transported heavy metals that move with the river water could be distinguished into natural original part and anthropogenic original part. The natural original heavy metals derived from the dissolved out heavy metals from the soils and rocks while the rainwater flow through and acted with them within the watershed before reaching the rivers. It is considered that the larger the accumulated rainfalls, the more the heavy metals move to the river. The dissolved rate of heavy metals also depends on the geologic features of the soils and rocks that reacted with rainwater, varying with different geologic features.

As a seasonal phenomenon, the water flow rate as well as the heavy metal content of this kind of non-point sources is influenced and changes with the climate in the watershed. However, anthropogenic original heavy metals come from the point sources, such as mine drainages or other industrial drainages, are insusceptible by the climate changes. The heavy metals that outflow from each polygon can be calculated as follows:

$$\begin{cases} C_{\mathrm{f}} = N_{\mathrm{f}} + A \\ N_{\mathrm{f}} = \alpha\beta \end{cases}$$

where C_{f} means the unit-time heavy metal outflow at a river cross section; N_{f} is unit-time heavy metal outflows derived from natural origin; α and β refer to the water-flow rate and the heavy metal concentration in the water, respectively; A is the outflow of heavy metals derived from anthropogenic origin. For tributary polygon, β is the heavy metal concentration of tributary water; for mainstream polygon, β is the average heavy metal concentration dissolved from the representative soil samples sampling within the polygon. For A, it was calculated based on the drainage information and data obtained from authorities. The water and heavy metal flow of anthropogenic origin flow into the mainstream at confluences. It was taken into account by the calculation of the increase and decrease of the water and heavy metal based on the water intakes and drainages along the water use route. The heavy metal concentration in river water was estimated by the heavy metal accumulation of the upstream polygons and drainages.

3 Results and discussion

3.1 Water flow rate analysis

As an essential data for heavy metal content estimation of the Kosaka River in this study, the water flow rates at the 6 monitoring points were measured twice, respectively in the snow-melting season of April and dry season of July in 2009. As a result of the measurement, the water flow rate at P6 in snow-melting season was roughly four times the amount in the dry season (Fig. 4). For the water flooding, the water flow rates at P4 and P5 could not be measured in snow-melting season. Instead, the water flow rates at the 470 m downstream of P4 and at the 590 m upstream of P5 were measured. The water flow rate calculation of tributary polygon involved was then adjusted accordingly. The water quantity of human-induced intakes and discharges was measured along the water use route in August 2010. There should be a difference between the water intakes in snow-melting season and in the dry season. However, since the relationship between the water intake quantity and the water flow rate of the Kosaka River was unknown, the water flow rates of the human-induced water flows were directly employed when it was on the investigation day. Figure 4 shows the relationship between the water flow rate and the watershed area both before and after the revision of human effects.

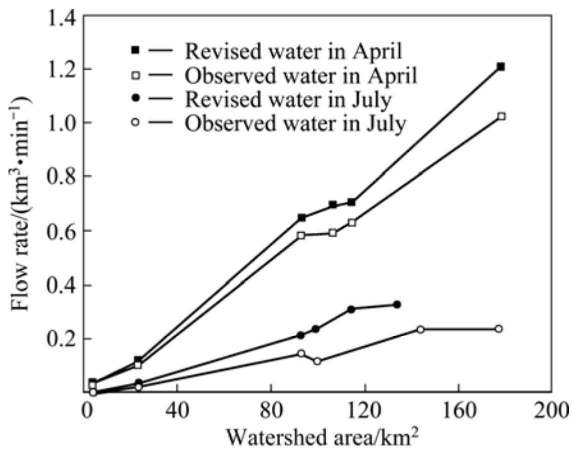

Fig. 4 Water flow rate before and after revision

In dry season, no water flows to the Kosaka River were observed in some of the polygons, such as polygons 504, 601 and 604 , in which the tributaries maybe dry up, or the water flow rates were too small to provide the water use. Accordingly, these kinds of polygons were ignored when calculating the water flow rate accumulation in the downstream. Consequentially, the watershed areas of P5 and P6 were smaller in July after being revised (Fig. 4). The revised water flow rates both in April and July were higher than the actually measured ones. The difference was regarded as the result of human activities. As the water flow rate estimation has significant effects on the estimation accuracy of heavy metal concentration in river water, if the water quantity of the intakes and drainages has not been correctly considered, estimation errors might be produced in the downstream of P3 from where the water use route was involved. Considering that the investigation data in August 2010 were directly used for calculation, there might be an overestimation in dry season and an underestimation in snow-melting season for water flow rate.

3.2 Mainstream water quality analysis

The experimental data of Cu concentration in the Kosaka River at monitoring points are summarized in Fig. 5. Seeing from the upstream to the downstream, the Cu concentration between P1 and P2 increases while decreases between P2 and P3 and increases again between P4 and P5. There is no obvious change between P3 and P4, that is to say, there are two peaks respectively at P2 and P5. It can be considered that the first peak at P2 might be caused by the Furutobe mine and Ainai mine, and the Cu concentration increased between P4 and P5 was mainly influenced by the Kosaka refinery drainage. The water quality was bound up with the location of the monitoring points both in the snow-melting season and dry season.

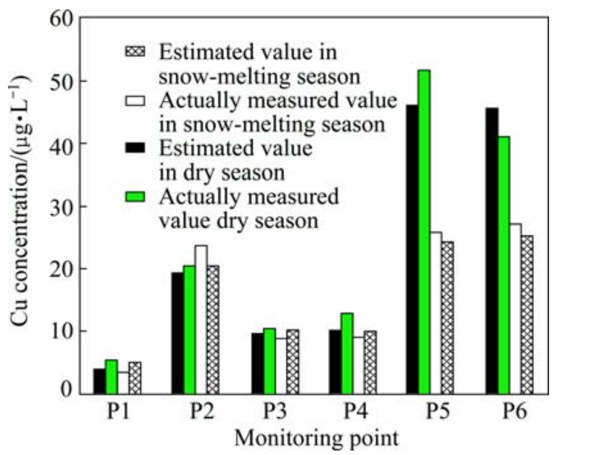

Fig. 5 Cu concentration in Kosaka River at monitoring points

Viewing from the seasonal variation, there were almost no differences of Cu concentration between the snow-melting season and the dry season at P1 and P3. The upstream watershed of P1 and the watershed between P2 and P3 were more natural than the other section of the whole Kosaka watershed area since there are no much human activity effect while no mine drainages and also no paddy field. Besides P1 and P3, the Cu concentrations in snow-melting season were quite different from those in the dry season at the other monitoring points, especially at the points in the downstream of P3 where they were influenced by both the Kosaka refinery drainage and paddy fields. At most of the monitoring points, the Cu concentrations were lower in snow-melting season than in dry season. This phenomenon was especially clear in downstream. The larger water flow rate in snow-melting season can effectively dilute the pollutants. Therefore, reducing the mine drainages in dry season should be proposed.

3.3 Estimation of Cu concentration in Kosaka River

3.3.1 Estimated Cu concentration at monitoring points

Thinking about the natural outflow of the tributaries and the human emissions in the Kosaka watershed, the Cu concentrations at the monitoring points along the Kosaka mainstream were estimated based on the water flow rate and Cu contents in tributaries as well as that in mine drainages. The results are shown in Fig. 5. The estimated Cu concentrations at most of the monitoring points were coincident with actually measured ones, especially for the points in the upstream. Considering that the situation in the downstream was more complex as there were water use routes with more human activities, the estimated error occurred easily. However, the results revealed that the mainstream river quality can be estimated from the water quality of their tributaries and human drainages.

3.3.2 Cu flow in Kosaka River

Cu flow moving along in a current of water in the river per unit time (hereafter called accumulated Cu flow), was analyzed by both the actually measured data and the estimation approach. For the actually measured value (solid lines in Fig. 6), the accumulated Cu flow was calculated by multiplying the actually measured Cu concentration and the water flow rate at the monitoring points together. For the estimated value (dotted lines in Fig. 6), it was the sum of all the outflows from the polygons and drainages locating upstream. The accumulated Cu flows of estimation well coincide with the actually measured ones in addition to slight errors in the most downstream area.

In general, the accumulated Cu flow in the Kosaka River gradually increaseed from upstream to downstream as the watershed area increased, excluding a decrease between P5 and P6 in dry season consistent with the change of the Cu concentration. The accumulated Cu flow in snow-melting season is much larger than that in the dry season. The excessive output between them ought to derive from the natural origin, and was mainly caused by the increase of rainwater. Because, anthropogenic original Cu flow which comes from the point sources is insusceptible by the climate changes.

3.3.3 Analyses of Cu sources in Kosaka River

The sources of Cu in the Kosaka River were analyzed by clarifying the outflow from each polygons and drainages in the whole watershed. The results are presented in Fig. 7. It can be seen that the green bars

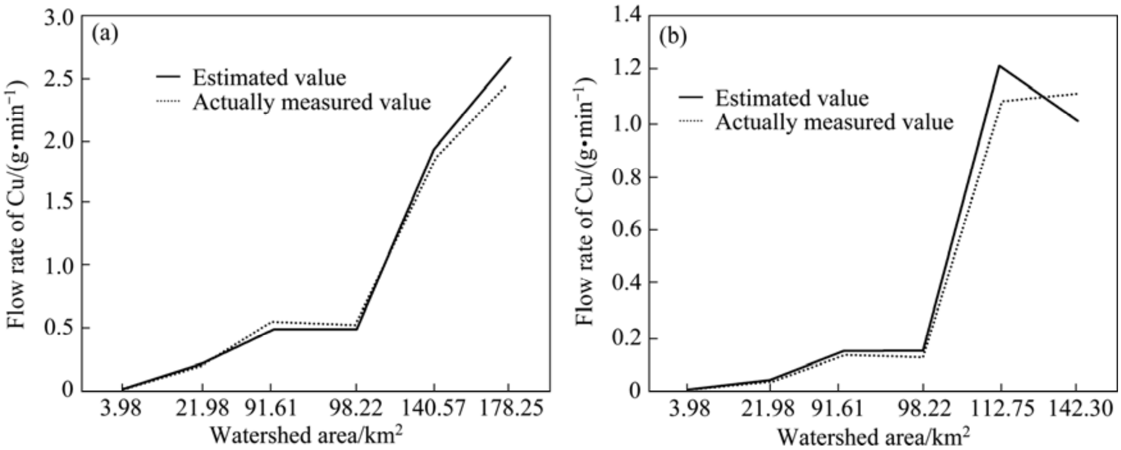

Fig. 6 Cu flows in Kosaka River: (a) In snow-melting season; (b) In dry season

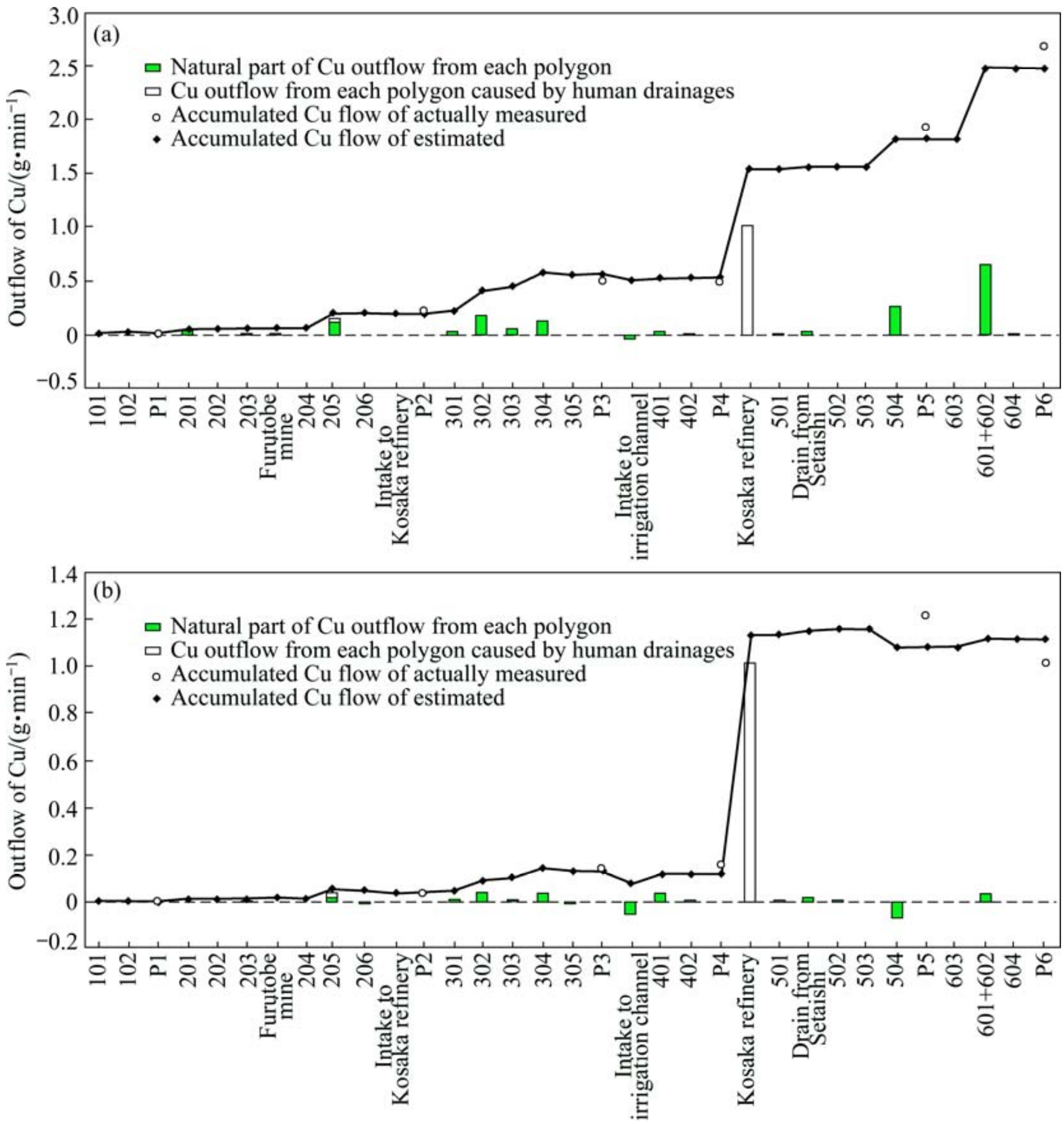

Fig. 7 Cu sources analysis for Kosaka River: (a) In snow-melting season; (b) In dry season

stand for the natural part of the Cu outflow from each polygon, which came from the non-point sources caused by the dissolution of geological environment elements, while the blank bars express outflows coming from the point sources that caused by human drainages. All of the outflows interflow into the mainstream, and the confluence effects were checked at the monitoring points, by comparing the estimated Cu concentration that calculated by accumulating all the outflows of the upstream polygons and drainages with the actually measured Cu concentration. They are roughly in accordance with each other.

The Cu outflow of polygon 205 included both the non-points source coming from the geological elements dissolution and the point source caused by Ainai mine drainag because the two outflows join together before flowing into the mainstream. Figure 7 shows that the drainage of the Kosaka refinery has significant effects on the Kosaka River quality. The Cu outflows vary a lot among different polygons, and have different influence on the Kosaka River water. The reason could be that the geological environment features are different among the polygons, thus resulting in different dissolution of Cu. There might be some polygons that are rich in Cu, and in which the natural dissolution quantity is large. Moreover, it is possible that some other non-point sources exist in the polygons with high heavy metal outflows. The increasing human interference in the downstream that reflected in the complex human water use (see Fig. 8), might also be one possible reason for Cu accumulation.

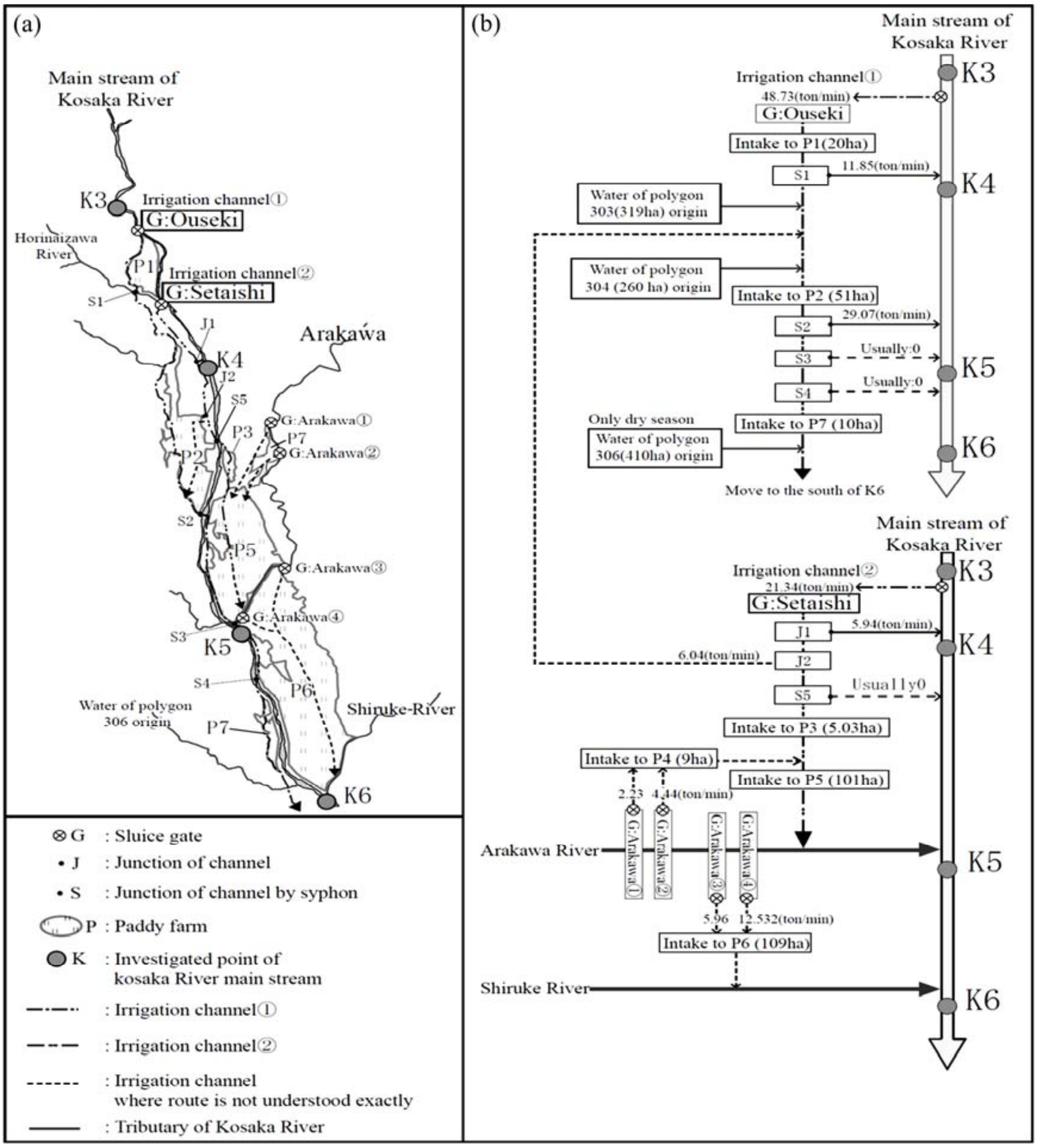

Fig. 8 Water use route for water intakes and outtakes to main stream: (a) Location; (b) Flow chart (K refers to the P in the text, and K1−K6 respectively refer to P1−P6.)

On account of that, mining district shows relatively high amount of heavy metal concentration in geological circumstances due to regional mineralization and/or related geological processes [16−18]. Such as, there are the remains of the abolished Kosaka Uchinotai mine and Toki mine respectively lying in the polygon 504 and the polygon 612 (601+602), without drainages.

4 Conclusions

1) The heavy metal content in river water could be estimated by the accumulation of all the outflows of the tributaries and human drainages upstream, that is to say, it is possible to estimate the heavy metal content in river water by getting the right water quantity and heavy metal concentration coming from each tributary watershed, and the confluence effects resulting in the river water quality.

2) This estimation method effectively distinguishes the anthropogenic origin heavy metals from the natural origin ones. The effects of mine drainages or other human drainages on river water quality could be clearly revealed.

3) There are spatial differences between the Cu outflows of the non-point sources, the natural outflows from the watershed polygons in Kosaka River area. Compared with the natural origin, the dominant emission sources affect the river water more significantly in local.

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日本东北北麓矿区小坂河中的铜行为及矿废水对其的影响

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摘　要：以重金属来源为出发点，通过区分自然来源和由人类活动造成的重金属污染以探究新的推算方法来估算河水中的重金属含量。以 GIS 为依托，北麓矿区的小坂流域被分割为若干子流域多边形，并计算了每个多边形单位时间内流出的水量和水溶态铜。与自然来源相比，人为排放的铜对局部河流水质的影响更为显著。综合考虑小坂流域中支流的自然析出和人为排放的质量平衡闭合，以上游各支流和矿废水排放的汇流累积效应来估算小坂河中的铜含量。估测的铜含量通过与主流各监测点的实测铜浓度比较来检验其估算的准确性，结果显示两者大体一致。研究表明，河流水质可以通过其上游支流的水质及人为水系的水质进行估算。

关键词：矿废水；河水；重金属；GIS；小坂流域

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