The Effect of Flood on the Water Quality and the Epilithic Diatom Assemblage in the River Affected by Heavy Metals Produced by Closed Copper Mines

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Abstract

The River Godani, a tributary of the River Kakehashi which joins with main stream of the River Kakehashi about 5km downstream, is known for its pollution due to the mining water. Typhoon No.7 passed through Ishikawa Prefecture on September 22 in 1998 with localized torrential down pour.

In this study, We examined the effect of the Typhoon No.7 on those Rivers from the viewpoint environmental factors of the river water and the ecological viewpoint based on the change of the epilithic diatom assemblages on the river bed.

Each of the four surveys (1998–1999) showed a tendency, in its measurement of EC values decreasing from upperstream to downstream in order.

The concentration of heavy metals increased in all sites after 6 months. At each sampling site 6 months after flood, the concentration of Cu was 1.3–2.0 higher, and that of Zn was 1.3–3.3 times greater than those obtained before flood, while the concentration of Cd showed no district difference from those obtained before flood.

The concentration of heavy metals in attached materials was far higher than that of heavy metals in the mud of river bed. This may be due to the fact that the greatest part of attached material consist of epilithic algae (organic material) and the rest is silt (inorganic material), while in the case of mud the volume of silt is much greater, than that of residue of epilithic algae. After all weight of epilithic algae per unit volume is small compared with silt, but they absorbs heavy metals. On the other hand, inorganic silt only absorbs heavy metals but no process of concentration occurs.

Three surveys, B (November 2, 1988), C (December 21, 1988) and D (March 24, 1999) were after flood.

In the survey B, *Achnanthes minutissima* which has strong tolerance to heavy metals was the most abundant taxon in almost all sampling sites.

It is noticeable that in all sites of D (150 days after flood), the most abundant taxon was *A. minutissima*. The temporary change of the most abundant taxon at Sites 3, 4 (cf. Fig. 2) is considered to be due to the fact that materials on the river bed easily came off and were carried downstream and piled up there temporarily.

Key words : epilithic diatom, the most abundant taxon, electric conductivity, flood, heavy metals

INTRODUCTION

Typhoon No.7 through Ishikawa Prefecture on September 22, 1998 with localized torrential down pour. The flood gave the greatest damage to the two basins of the River Kakehashi and its tributary, the River Godani since the flood in 1896.

The River Godani, a tributary of the River Kakehashi about 5 km downstream is known for its pollution due to the mining waste.

Before and after the closing of Ogoya copper mines, many studies were made on the recovery of organisms due to the decrease of the inflow of heavy metals (Cu, Zn and Cd), using biological indicators¹⁻⁵⁾.

Ito (1975) considered the pollution of Godani and Kakehashi basins as an inorganic pollution due to the still remaining mine-effluent of the closed Ogoya mines, and proved it using aquatic insects and epilithic algae as biological indicators⁶⁾.

In this study we also make use of these biological indicators. We made, however special effort trying to find out the sequence of change of the content of heavy metals before and after the greatest increase and decrease of river water caused by the Typhoon No.7(Sep., 1998).

Our research covered the 150 days from Sept., to March 1999 through 4 periods (A: Before flood, B: 40 days after flood, C: 80 days after flood, D: 150 days after flood) following the sequence of changes of the composition of epilithic diatoms.

MATERIALS AND METHODS

As shown Fig.1 samples were collected from 4 sampling sites shown in Fig. 1. Sites 1 and 2 in the River Godani were Kurataniguchi and Sawa. Sites 3 and 4 in the River Kakehashi were Hanazaka Shorenji.

In Site 1, percolate water from abandoned Kuratani mine and flowing water from the setting basin, flow together to the River Omodani, which joins the River Godani. Upperstream district of Sites 3 and 4 had several abandoned mines, but their polluting influence on the River Kakehashi was very little.

The material were collected in four

different times, namely before flood(A) and temporary repairs of road and river walls after the flood caused by Typhoon No.7(B-D). B was done about 40 days after the flood on November 2, 1998. C was about 80 days the flood December 21, 1998. D was carried out 150 days after the flood March 24, 1999.

1. Environmental factors

The water samples were taken from the surface water in each site. The determination of water temperature, pH and EC(electric conductivity) was conducted on the spot. A pH meter(model B-212, HORIBA, Japan) and an electric conductivity meter(model B-173, HORIBA, Japan) were used.

1) Determination of heavy metals(Cu, Zn and Cd) in river waters

The water samples were taken from the depth of 20-30 cm from river surface and were filtered through a 0.45 μ m membrane

Fig. 1 Sampling sites

- 1 : Kurataniguchi
- 2 : Sawa
- 3 : Hanazaka
- 4 : Shorenji

- ✕ 1 : Kuratani (ogoya) former mining area
- ✕ 2 : Kanahira former mining area
- ✕ 3 : Gokouji former mining area

filter(Millipore Corps, Type HA) with an aspiration pump soon after the sampling, as the treatment of spa⁷. 1ml of concentration nitric acid was added to 100 ml of the filtrate. The water samples were stored in the glass bottles.

On the basis of calibration curve established by series of the standard solutions, the concentration of metals(Cu, Zn and Cd) was determined by FAAS(flame atomic absorption spectrometer) for Cu and Zn or GFAAS(graphite furnace atomic absorption spectrometer) for Cd.

All the working solution of metals were prepared to be 0.1M HNO₃ solution. In the case of determination of Ca, a 1000mg/l was added to the standard and the sample solution an interference suppressant. The determination was replicated at least three times, and the abnormal data were eliminated by using the Grubb's test.

2) Determination of heavy metals(Cu, Zn and Cd) in attached materials

Sampling of attached materials was carried out with the same criteria as in the case of mud on the river. The attached materials consisted of algae and silt(clayey particles of nearly 10 μ m in diameters).

At the river bed in the depth of 30-40 cm, the stones with almost parallel surface to that of water were selected. The attached materials on the upper surface were brushed down by using a nylon brush and were fixed in a formaldehyde solution.

The samples were centrifuged(2000 rpm $\times$ 10 min.) in order to remove bulk waters. The supernatant materials were discarded and the precipitate was dried over-night at 80°C. Each of the dried samples(10-20 mg) was transferred into a glass test tube. A small amount of water was added so as to wet the sample and 1 ml of concentrated nitric acid water was then added. The tube was set in the quick digesters, and was treated at half power mode, i.e.350W for 30min. Then 1 ml of the mixture of concentrated nitric acid and perchloric acid (3:1v/v) was added to the sample⁹.

It was treated in the digester at 350 W for 30 min and continually at 700W for 20 min again. Most of the sample was dried up under this condition, but the heating treatment of

700W was continued until all the content was dried up. After cooling the tube, 1 ml of 1M nitric acid was added to residue in the tube to leach out metals by ultrasonic treatment. In order to remove the white crystals of silicate the suspension was filtered through a 0.45 μ m membrane filter. The filtrate was transferred to a 25 ml or 50 ml, volumetric flask. A proper amount of nitric acid was added so that final solution showed be 0.1M nitric acid solution, and another 1000mg/l of nitric acid was added as an interference suppressed. Then, the solution was diluted up to the line with the distilled deionized water.

The determination was replicated at least three times, and the abnormal data were rejected by using the Grubb's test.

3) Determination of heavy metals(Cu, Zn and Cd) in the mud of river bed

The sediment on the river bed mud was taken, and the relatively large stones and psephites in it were removed. Then, the residual mud in the river waters was transferred to a polyethylene bottle and was brought back to the laboratory. The samples were stored in a refrigerator prior to analysis. Each of the samples was transferred to 500 ml beaker. Water was added to it and was mixed with a glass rod. After leaving it for 3 min, psephites and fine sands were precipitated down quickly, while the floating particles were suspended in the mixture. The suspension was taken out by centrifugation (2000 rpm $\times$ 10 min) and was over-night at 110°C.

Each of the dried samples was digested in the case of attached materials, and the concentration of metals was determined by FAAS(Cu, Zn) or GFAAS(Pb, Cd).

2. Sampling and treatment of attached algae

In the sampling of attached algae, the following criteria were used⁹ :

- Samples were taken in riffles about 30 cm deep and with about 40 cm/s flow rate.
- Samples were collected only from the flat stone surfaces parallel to the water surface.
- In places where river water did not flow, samples were taken from a

vertical surfaces of any non-living substrate 30 cm deep in water to avoid the effect of sedimentary matters.

- d) To obtain assemblages which had reached maturity in their successional process, only those with large standing crops were collected.

These samples were cleaned with sulfuric acid and potassium permanganate and mounted in cedax.

Diatom taxa were identified using micrographs at 1500 magnification referring to the following descriptions¹⁰⁻¹⁵⁾.

The relative abundance of diatom taxa in each assemblage was obtained by counting the number of individuals.

RESULTS AND DISCUSSION

1. Environmental factors

1) Water temperature, pH and EC

As shown in Table 1, the range of water temperature was from 6.9°C (C, Site 2) to 20.9°C (A, Site 4).

The river water of the Omodani, which joins the River Godani at Kurataniguchi (Site 1), is polluted by the percolated water from the now abandoned Kuratani mines and showed pH values 4.6-5.4 when measured.

In order to neutralize the condition, treatment by $\text{Ca}(\text{OH})_2$ was useful way.

Changing heavy metals (Cu, Zn and Cd) to their hydroxide and letting them precipitated to the bottom of water body. So the range of pH values measured in this survey was 6.8 (D, Site 1) to 8.7 (D, Site 3).

EC values were ranged from 98 $\mu\text{s}/\text{cm}$ (C, Site 3) to 320 $\mu\text{s}/\text{cm}$ (D, Site 1). In each sampling time, similar tendency of measurement values was observed reduction of values from Site 1 to Site 2 in order.

The concentration of heavy metals in the river water was high at Site 1 near the precipitation tank, but the concentration of heavy metals in the river water naturally lower where there is no more river water bringing heavy metals into the middle and lower stream. This tendency remains the same after the flood.

2) Concentration of heavy metals (Cu, Zn and Cd) in the river waters

As shown in Table 2, the analytical results

Table 1 Water temperature, pH and EC

Factor	Sampling Site	A	B	C	D
Water Temperature (°C)	1	19.0	14.7	7.2	10.3
	2	20.0	14.4	6.9	10.0
	3	20.5	13.8	7.2	8.3
	4	20.9	14.7	7.7	8.6
pH	1	6.9	7.1	7.2	6.8
	2	7.3	7.9	8.1	7.4
	3	7.9	7.8	8.0	8.7
	4	8.0	7.8	7.8	7.6
EC ($\mu\text{s}/\text{cm}$)	1	242	280	178	320
	2	171	190	110	170
	3	120	135	98	100
	4	130	134	110	100

A : Before flood, September 4, 1998

B : About 40 days after flood, November 2, 1998

C : About 80 days after flood, December 21, 1998

D : About 150 days after flood, March 24, 1999

Table 2 Concentration of heavy metals (Cu, Zn and Cd) in the river water

Factor	Sampling Site	A	B	C	D
Cu (mg/l)	1	0.15	0.24	0.22	0.25
	2	0.027	0.035	0.054	0.053
	3	0.015	0.015	0.019	0.019
	4	0.013	0.014	0.020	0.020
Zn (mg/l)	1	0.92	1.26	1.39	2.70
	2	0.38	0.37	0.41	0.74
	3	0.19	0.27	0.20	0.25
	4	0.08	0.22	0.19	0.26
Cd (mg/l)	1	0.0043	0.0061	0.0051	0.0074
	2	0.0012	0.0022	0.0016	0.0021
	3	0.0006	0.0015	0.0009	0.0008
	4	0.0006	0.0013	0.0008	0.0008

of the concentration of heavy metals in the river water were as follows; Cu from 0.013 mg/l (A, Site 4) to 0.25 mg/l (D, Site 1); Zn from 0.08 mg/l (A, Site 4) to 2.7 mg/l (D, Site 1); Cd from 0.0006 mg/l (A, Sites 3, 4) to 0.0074 mg/l (D, Site 1).

Each survey showed a similar tendency in the measurement of their concentration to decrease from upperstream to downstream, as in the case of EC values.

3) Concentration of heavy metals in attached materials on the river stones

As shown in Table 3, the analytical result of heavy metals in attached material on the river bed, stones were as follows; Cu was from 1500 $\mu\text{g}/\text{g}$ (A, Site 4) to 82300 $\mu\text{g}/\text{g}$ (D, Site 3); Zn from 2100 $\mu\text{g}/\text{g}$ (C, Site 4) to 40600

$\mu\text{g/g}$ (D, Site 3); Cd from $3.8 \mu\text{g/g}$ (B, Site 1) to $30 \mu\text{g/g}$ (A, Site 3).

The values of concentration of Cu decreased both before flood(A) and after(B, C) from upstream to downstream, but the measurement after flood(D) showed increase in Sites 3 and 4 in the downstream area. These values were even 50.7-51.4 times more than those obtained in the same sites before flood(A). The values of Zn, showing the same tendency, were 6.1-8.1 times more than those obtained before the flood.

The values of Cd unlike those of Cu and Zn showed no distinct change either in different sites or in different periods.

4) Concentration of heavy metals in the mud of river bed

As shown in Table 4, the analytical results

Table 3 Concentration of heavy metals(Cu, Zn and Cd) in the attached materials on the surface of stones on the river bed

Factor	Sampling Site	A	B	C	D
Cu ($\mu\text{g/g}$)	1	24800	21200	32500	20600
	2	10400	16900	8130	24600
	3	1600	8820	2490	82300
	4	1500	6540	2550	76000
Zn ($\mu\text{g/g}$)	1	8900	7260	8070	5750
	2	11400	10200	5580	13600
	3	6680	7150	3540	40600
	4	4450	5330	2100	36000
Cd ($\mu\text{g/g}$)	1	15.0	3.8	13.0	7.4
	2	24.0	12.0	9.3	11.0
	3	30.0	14.0	14.0	9.5
	4	22.0	15.0	8.7	18.0

Table 4 Concentration of heavy metals(Cu, Zn and Cd) in the mud on the river bed

Factor	Sampling Site	A	B	C	D
Cu ($\mu\text{g/g}$)	1	7660	12500	5970	15600
	2	2890	770	1400	2560
	3	1280	500	690	980
	4	1300	670	960	1200
Zn ($\mu\text{g/g}$)	1	2510	3520	1580	3000
	2	1660	833	960	1300
	3	1280	890	917	940
	4	1480	800	881	820
Cd ($\mu\text{g/g}$)	1	5.0	4.1	2.7	4.2
	2	5.2	1.7	2.5	2.8
	3	7.3	2.8	3.7	3.3
	4	7.8	3.0	8.4	2.7

of the heavy metals in mud on the river bed were as follows; Cu was from $500 \mu\text{g/g}$ (B, Site 3) to $15600 \mu\text{g/g}$ (D, Site 1); Zn from $800 \mu\text{g/g}$ (B, Site 4) to $3520 \mu\text{g/g}$ (B, Site 1); Cd from $1.7 \mu\text{g/g}$ (B, Site 2) to $8.4 \mu\text{g/g}$ (C, Site 4).

Cu decreased before flood(A) in Sites 2 and 4. The values of Cu were only 17% of there obtained at Site 1. The same tendency was observed in surveys after flood(B, C, D). But after the flood, the values of Site 4 were a slightly higher than those obtained at Site 3.

Zn showed high values in every sampling time and decreased from Site 1 to Site 4 in order.

Cd showed higher values in Sites 3 and 4 than those obtained at upstream sites, but 40 days after flood(B) showed opposite tendency. As shown in Tables 3 and 4, regardless of survey time and Survey site, the concentration of Cu, Zn and Cd in attached materials give higher values than those of the concentration of Cu, Zn and Cd in the mud.

2. Epilithic diatom assemblages on the river bed

1) The structure of diatom assemblages

Main constituent of attached materials on the river bed is epilithic algal communities. In this research diatom assemblages belonging to a attached algal community, were surveyed four times in all sites; once before the flood and three times after the flood.

73 taxa(20 genera) were identified in this survey. The number of taxa belonging to the genus *Navicula* was 15 and the number of taxa belonging to *Gomphonema* was 12.

Table 5 shows the main diatom taxa and their relative abundance of 4 sites in each sampling time. The name of taxa whose relative abundance does not exceed 1% at any station of sampling-time were omitted from the table.

It is clear that the strongest affect of the flood was observed at site after flood(B). No diatom assemblage was found at this site.

2) The most abundant taxon and their relative abundance

The taxon with the most relative abundance is the specific composition of diatom assemblages are shown in Fig.2. As shown in Fig.2 11 diatom assemblages out of

Table 5 Main diatom taxa and their relative abundance(%) at 4 sites in each sampling time

Species	Date				Date				Date				Date			
	Sep. 4, 1998(A)				Nov. 2, 1998(B)				Dec. 21, 1998(C)				Mar. 24, 1999(D)			
Sampling Site	1	2	3	4	1	2	3	4	1	2	3	4	1	2	3	4
<i>Achnanthes japonica</i>		4.7	9.8	8.2		3.7	1.5		2.3	1.0	1.0		1.7	1.6	4.2	1.5
<i>A. lanceolate</i>	4.2		3.3	1.9				1.1			1.0			1.0	2.1	2.5
<i>A. minutissima</i>	93.2	12.6	45.2	13.4	92.6	73.3	11.3	84.2	79.1	28.2	76.2	95.0	85.9	52.9	48.0	
<i>Amphora pediculus</i>		3.5	3.3	1.2					1.3							
<i>Cocconeis plascentula</i> var. <i>lineata</i>																1.2
<i>Cymbella minuta</i>		1.9	1.3	2.4				2.4		1.2	2.6	1.5			2.4	2.2
<i>C. turgidula</i> var. <i>nipponica</i>								1.3			1.0					
<i>Diatoma hiemale</i> var. <i>mesodon</i>								1.1	3.5							5.5
<i>Eunotia minor</i>			1.3													
<i>Gomphonema angustatum</i>								2.7				1.1				1.5
<i>G. clevei</i>			1.3													
<i>G. helveticum</i>		1.6	3.6													
<i>G. minutum</i>			1.0	1.6				3.3			2.6	3.3				
<i>G. parvulum</i>			1.3	3.8				1.1			1.4	1.3				
<i>Melosira varians</i>		1.9	1.3	2.1						4.2	4.8				2.7	1.8
<i>Navicula pupula</i>			2.6													
<i>N. ventralis</i>			3.0	4.9			1.4	1.8			1.8				2.1	2.2
<i>Nitzscha parvula</i>			1.0													
<i>Surirella angusta</i>	2.6	66.9	16.4	52.0	1.7	12.9	54.3	4.2	7.8	13.3	9.7	2.2	4.8	4.2	4.0	
<i>S. biseriata</i>				1.2												
<i>S. ovata</i> var. <i>pinnata</i>				1.9			4.9	13.7			32.2	2.9		1.0	7.7	18.5
<i>Synedra rumpens</i>			1.0								1.0				4.7	2.2
<i>S. ulna</i>											2.2					
<i>S. ulna</i> var. <i>ramesi</i>																1.2
Total number of taxa occurred	3	19	25	27	0	10	20	22	15	24	33	18	6	22	41	31
Total number of valves counted	309	317	305	425	0	462	588	451	311	502	503	546	534	505	337	325

Fig. 2 The most abundant taxon in the diatom assemblages

□ : *Achnanthes minutissima*
 ▨ : *Surirella angusta*
 ■ : *S. ovata* var. *pinnata*

A: Before flood
 B: 40 days after flood
 C: 80 days after flood
 D: 150 days after flood

15, *Achnanthes minutissima* was the most abundant taxon and the range of its relative abundance was from 45.9-95.2%.

In three diatom assemblages(A, Site 2 and 4, B, Site 4) *Surirella angusta* was the most abundant taxon and the relative abundance was from 1.7-66.9%.

In one diatom assemblage(C, Site 3), *S. ovata* var. *pinnata* was the most abundant taxon and the relative abundance was 32.2%.

3) The number of taxa and Shannon's diversity index

Shannon's diversity index(Di) was obtained in all epilithic diatom assemblages. As shown in Table 6, the range of Di was from 0.3 to 1.3.

The fact that both in the upstream area(sites 1, 2) and in the downstream area (sites 3, 4) the Di showed comparaively small values indicates that the main cause of the river water pollution was the heavy metals(Cu, Zn and Cd).

SYNTHETIC CONSIDERATION

1. EC in river water

Each survey showed a tendency in its measurement of EC values, which reduced from Site 1 to Site 4 in order. The concentration of heavy metal ions in the river water was naturally diluted. There is no inflow of rivers containing heavy metal ions. This situation remained the same after the flood.

2. Concentration of heavy metals(Cu, Zn and Cd) in the river water

As shown in Table 2, the values of heavy metals increased in each site after the flood, and this tendency continued about 6 months. Concentration of ions heavy metals in the river water was highest near, Site 1. But it was no diluted naturally on middle and

lowstream.

Also in this case, as in the case of EC values each survey showed a tendency in the measurement of concentration which reduced from upstream to downstream.

At each site, 6 months after the flood, the values of Cu, Zn and Cd were greater than those obtained before the flood(Table 2).

This fact may be considered as the result of the process in which the attached materials on the river bed easily came off and were carried downstream, and piled up there. But more detailed survey must be undertaken before a definite conclusion can be reached.

3. The concentration of heavy metals in the attached materials and mud on river bed

It is obvious from Tables 3 and 4, that the concentration of heavy metals in attached materials was far higher than in river mud.

This may be due to the fact that the great part of attached material consisted of epilithic algae(organic material). After all the weight of epilithic algae per unit volume is small compared with silt, but they absorb and concentrate heavy metals. On the other hand, inorganic silt only absorbs heavy metals but no process of concentration occurs.

So as shown in Tables 3 and 4, the concentration of heavy metals in attached materials is much higher than in silt.

4. The most abundant taxon in epilithic diatom assemblages

28 years have passed since the Ogoya mine was closed, but the specific structure of epilithic diatom assemblage sample taken at each sampling site was still affected by heavy metals.

In these water areas, *Achnanthes minutissima* which has strong tolerance,

Table 6 Number of taxa and Shannnon's diversity index(Di) of diatom assenblage

Sampling Site	A		B		C		D	
	Total number of taxa	Shannon's diversity index(bit)	Total number of taxa	Shannon's diversity index(bit)	Total number of taxa	Shannon's diversity index(bit)	Total number of taxa	Shannon's diversity index(bit)
1	3	0.29			15	0.76	6	0.26
2	19	1.31	10	0.35	24	0.95	22	0.76
3	25	2.03	20	1.08	33	2.85	41	2.13
4	27	1.83	22	1.70	18	2.13	31	1.88

occupies its position as the most abundant taxon, among taxon, diatom assemblage.

In almost all sites at the period B(40 days after flood), the abundant taxon was *A. minutissima* except at Site 4.

It is worth noting that in all sites at the period D(150 days after flood), the most abundant taxon was *A. minutissima*.

The changes of the abundant taxon in Sites 3 and 4, signifying the increase of pollution materials, were considered to be due to the fact that materials on the river bed easily came off and were carried downstream and piled up there temporarily.

The changes, however, might also be considered to be due to a crop-out of a copper vein caused by the flood.

In this case the effects of heavy metals in the river water are expected to extend over a considerable length of time. In order to investigate and clarify this situation, we intend to focus our attention on the changes of epilithic diatom assemblages for sometime in the future in the same area.

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