Bull Environ Contam Toxicol (2011) 87:669–673
DOI 10.1007/s00128-011-0411-z

Metal Concentrations of River Water and Sediments in West Java, Indonesia

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Received: 27 April 2011 / Accepted: 9 September 2011 / Published online: 7 October 2011

Abstract To determine the water environment and pollutants in West Java, the contents of metals and general water quality of the Ciliwung River in the Jakarta area were measured. High Escherichia coli number (116–149/mL) was detected downstream in the Ciliwung River. In addition to evaluate mercury pollution caused by gold mining, mercury contents of water and sediment samples from the Cikaniki River, and from paddy samples were determined. The water was not badly polluted. However, toxic metals such as mercury were detected at levels close to the baseline environmental standard of Indonesia (0.83–1.07 μg/g of sediments in the Cikaniki River). From analyses of the paddy samples (0.08 μg/g), it is considered that there is a health risk caused by mercury.

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Keywords Mercury contamination · Paddy · Water quality · West Java

River water is used for drinking, irrigation for agriculture and fish culture. However, water pollution caused by chemical substances such as metals is a serious problem for the inland water environment in many countries, especially developing nations. Southeast Asia is industrializing but this process affects tropical rain forests, rivers, and mangroves. As a result of various developments such as the "green revolution" and use of chemical fertilizers, income has increased (Estudillo and Otsuka 1999). However, unemployment and destruction of the ecosystem have also increased (Shiva 1991). Therefore, in developing countries, the idea of "sustainable development" is gaining currency. Forty years ago in Japan, as a negative effect of industrialization, there were severe problems with Cd and Hg pollution in Toyama and Minamata (Kimura 1988). Recently, similar phenomena have occurred in Southeast Asia (Hutagalung 1987). This study focused on Indonesia because, in West Java, the rivers also play important roles as traffic arteries and in economic activities. Studies of the quality of water and sediments are needed to evaluate environmental conditions. In the Jakarta gulf, Hg contamination has been reported (Hutagalung 1987; Mahbub and Kuslan 1997).

The Ciliwung River is one of the major sources for water in the capital city, Jakarta. One of the most serious problems in Jakarta is the lack of sewage systems in urban areas; less than 3% of Jakarta's population is connected to such a system. For this reason, domestic waste-water including human waste penetrates underground or flows out directly into rivers whose water is used directly by many persons for cooking, washing, bathing and even drinking.

Therefore, to examine the water environment and pollutants in the Ciliwung River of West Java, Indonesia, the metal contents and general water quality in the river were measured. In addition, to evaluate the health of the inhabitants in an area polluted by Hg due to gold mining, Hg contents of rice and a paddy field obtained from a site near an amalgamation plant beside the Cikaniki River, and of water and sediment from the river, were determined.

Materials and Methods

This study was conducted at ten sampling sites in the Ciliwung River and Cikaniki River in West Java, Indonesia. These sampling sites are shown in Fig. 1. Samples of water and sediments were taken 3 times from September 12–24, 2006, and June 12–19 and September 8–15, 2007. In addition, paddies and soil samples in a rice paddy were obtained near a gold amalgamation plant in the upstream area of the Cikaniki River (Cisarua Village, site [St.] 9).

The pH, conductivity, chemical oxygen demand (COD), NO_2, NO_3 and PO_4 levels in the water samples were measured immediately at each sampling point with a pH meter (Shindengen, model pH boy-KS701, Japan), a specific conductivity meter (Iuchi model TDS-can3, Japan), and simple water quality test packages (Kyoritsu Chemical-Check, WAK-Cl, Japan) according to their instruction manuals. *E. coli* were counted using *E. coli* detection paper (Shibata, Japan).

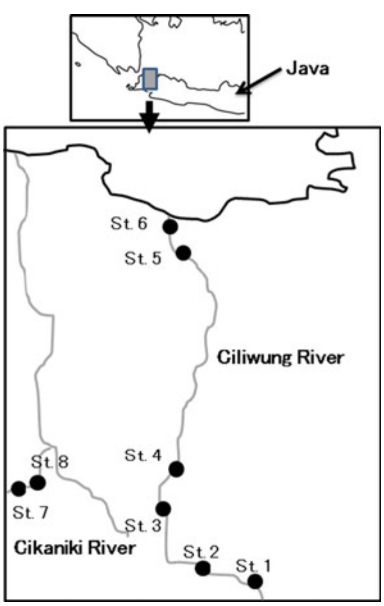

Fig. 1 Sampling sites in West Java, Indonesia

Before the determination of metal contents in water samples, 5 ml of ultrapure concentrated (conc.) HNO_3 was added to 5 mL aliquots of samples for measurement of toxic metals (Sigma–Ardrich). The contents of Mg, Mn, Al, Co, and Pb in the samples were analyzed with an inductively coupled plasma mass spectrometer (ICP-MS, Seiko SPQ-6500, Tokyo, Japan). The detection limit of each metal is around 0.1–1 ng/mL. To extract the metals from the sediments, 50 mL of 0.1 M HNO_3 were added to 10 g of dried sediment, and subsequently the mixture was agitated for 24 h. The supernatants were collected after centrifugation at 3,000 rpm for 15 min. Twenty paddy samples (about 0.4 g each) from ickers were completely digested with 10 mL of conc. HNO_3, and supplemented up to 100 mL with distilled water. To remove the insoluble materials, the digested solution was filtered with a 0.45 µm Millipore filter (USA). The contents of Mg, Mn, Al, Co, Cd, Pb, Fe, Cu and Zn were then measured with the ICP-MS. In addition, inorganic Hg contents in the same samples were measured using a Hiranuma HG 300 Mercury Analyzer (Hiranuma Sangyo Co., Ltd., Japan: detection limit 0.1 ng/mL). Total Hg contents were determined as follows: to degrade the organic compounds, 1 mL of conc. H_2SO_4, 1 mL of conc. HNO_3 and 2 mL of $KMnO_4$ (50 g/L) were added to 30 mL of the above-mentioned acidified sample. The mixture was shaken for 15 min, and heated at 95°C for 2 h. After cooling, 1 mL of hydroxylamine chloride was added to neutralize the excess $KMnO_4$. The neutralized solution was filtered with a 0.45 µm Millipore filter. The filtered solution was diluted up to 100 mL with distilled water and the Hg contents were measured using a Hiranuma HG 300 Mercury Analyzer.

Results and Discussion

The water quality in the samples from the Ciliwung and Cikaniki Rivers is listed in Table 1. Each number is the mean value for the 3 sampling times. The pH values ranged from 7.1 to 7.5. This pH range was considered to be normal. The conductivity values ranged from 58 to 375. The highest value was 375 downstream in the Ciliwung River at St. 5 and the lowest was 58 upstream in the Ciliwung River, at St. 1. The highest COD value in the samples was 130 mg/L downstream in the Ciliwung River, at St. 6. The ranges of NO_2 and NO_3 concentrations were from 0–0.63 mg/L to 0.1–15 mg/L, respectively. The middle reach of the Ciliwung River at St. 3 had the highest values for them. It was interesting that a low concentration of NO_x was observed downstream in the Ciliwung River in the city of Jakarta, at Sts. 5 and 6. In addition, the highest phosphate concentrations, 2 mg/L were detected in the downstream area of the Ciliwung River Sts. 5 and 6. The number

Table 1 General water quality of the rivers in West Java, Indonesia

Site no.	Altitude (m)	Conductivity (μS/cm)	COD (mg/L)	NO_2 (mg/L)	NO_3 (mg/L)	PO_4 (mg/L)	*E. coli* (no./mL)	pH
1 Ciliwung River	1,178	58	5.5	n.d.	0.1	0.05	13	7.3
2 Ciliwung River	467	113	14	0.01	1.2	0.35	31	7.5
3 Ciliwung River	252	183	35	0.63	15.0	1.07	67	7.1
4 Ciliwung River	131	193	55	0.30	15.0	0.73	116	7.1
5 Ciliwung River	26	375	80	0.05	2.0	2.00	117	7.1
6 Ciliwung River	16	320	130	0.02	1.0	2.00	149	7.2
7 Cikaniki River	405	60	15	0.04	1.0	0.13	55	7.2
8 Cikaniki River	341	103	10	0.18	6.0	0.63	57	7.2

n.d. means not detected

of *E. coli* in the river water was relatively high because domestic waste-water was discarded directly into the river. As expected, *E. coli* were found at all sampling points. In the downstream of the Ciliwung River, the level of *E. coli* was extremely high. These results indicated that contaminants due to domestic waste-water, for example, PO4 and *E. coli* , increased in a distance-dependent manner from the upstream of the river. On the other hand, only NOx, which is an indicator for organic materials and pesticides, was increased in the middle reach of the river. From St. 2, the number of rice paddies and dairy farms increased.

Concentrations of metals such as Pb, Mg, Mn, Al and Co in river water are shown in Table 2. The concentration of Mn in the Ciliwung River was about 10 times higher than that at the other points. Many miners search for alluvial gold in the Bogor area. In the purification process of the alluvial gold, Hg is widely used as in the gold-amalgam method. In this study, the Hg concentration was determined in the Ciliwung River and the upstream area of the Cikaniki River where there is a gold-amalgam refinery. Hg was hardly detected in the Ciliwung River. However from 0.119 to 0.218 ppb Hg was found in the Cikaniki River, and about 80% of the total Hg was inorganic (Table 2).

To study the relationship between metal concentrations of river water and sediment samples, sediment samples were taken at each sampling site except St. 5 (which was too deep). The metal concentrations in sediment samples are summarized in Table 3.

Metal concentrations in the sediments were more than 10-fold those in river water. At St. 4, concentrations of all metals were lower than at other sampling sites. The reason for this is assumed to be the presence of a filtration plant near St. 4. In the downstream of the Ciliwung River, at St. 6, a high concentration of Cd was detected. The Hg concentrations in sediments at 3 sites of the Cikaniki River (St. 7, St. 8 and the paddy field) and rice are shown in Table 4. The total Hg concentrations ranged from 0.63 to 1.07 μg/g, and the inorganic Hg concentration ranged from 0.23 to 0.333 μg/g. Unlike in the water samples, organic Hg accounted for 50% to 70% of the Hg in the sediments. Metal concentrations in the paddy samples are shown in Table 3. These concentrations were not so high. However, the total and inorganic Hg concentrations in the paddy samples were 0.08 and 0.065 μg/g, respectively (Table 4).

As shown in Table 1, COD levels and the number of *E. coli* were quite high. The results for general water quality in the present study were in good agreement with the results reported by Kido et al. (2009) and Kurasaki et al. (2000). It was thought that river water in West Java contained high levels of organic compounds and agricultural chemicals because of the limited availability of sewage systems in the area. Typical evidence of this was the detection of high levels of *E. coli* in the water samples, as described by Kido et al. (2009). The high microbial contamination in the river water is a health risk to the inhabitants. Thus, to enhance the river water environment in West Java, sewage and drainage systems need to be established as soon as possible.

To examine whether metal pollution occurred, metal concentrations were measured in water and sediment samples. As a result, the concentration of Mn in the Ciliwung River at Sts. 5 and 6 was found to be about 10 times higher than at the other sites. This value exceeded the environmental standard value. Although Mn is an essential trace element, excess uptake of it may result in some nerve disorders (Benedetto et al. 2010). Toxic metals such as Pb were scarcely observed in the water samples. As shown in Table 3, relatively high Cd contents were detected in sediments at all the sampling sites. Since Cd was also detected in the upstream area, it was considered that this metal was naturally present in the environment rather than due to contamination in West Java. In Japan, a serious illness due to Cd pollution called “Itai-itai disease” is well known. The disease is considered to be due to Cd contained in rice paddies. In 2006, the international baseline value for paddies growing rice was fixed at less than 0.4 ppm (Yu et al. 2006). The Cd content in the paddy in this study was lower than this baseline value (Table 3). However, the value was about 3 times higher than that in Japan (Environmental White Paper of Yamaguchi Prefecture

Table 2 Metal concentrations (μg/L) in river water in West Java, Indonesia

Site No.	Mg	Mn	Co	Al	Pb	Total Hg	Inorganic Hg
1 Ciliwung River	3,150	34.0	26.5	36.6	2.47	n.d.	–
2 Ciliwung River	4,035	32.5	25.9	51.7	2.55	n.d.	–
3 Ciliwung River	3,930	32.5	25.7	35.9	3.22	n.d.	–
4 Ciliwung River	3,960	36.8	26.1	33.8	3.19	n.d.	–
5 Ciliwung River	4,670	200.8	25.6	44.1	3.55	n.d.	–
6 Ciliwung River	4,520	395.8	25.3	35.6	3.88	n.d.	–
7 Cikaniki River	1,380	36.2	25.8	160.9	3.28	0.218	0.185
8 Cikaniki River	1,660	34.8	25.8	139.8	3.34	0.119	0.096

n.d. and – means not detected and not determined, respectively

Table 3 Metal concentrations (μg/g) in sediment and paddy samples

Site No.	Al	Cd	Co	Cu	Mg	Mn	Pb	Zn
1 Ciliwung River	590	0.34	1.68	1.68	222	104	0.301	6.4
2 Ciliwung River	376	0.83	1.78	3.98	165	224	0.615	13.7
3 Ciliwung River	560	0.69	1.21	2.49	191	49.8	1.43	8.7
4 Ciliwung River	0.406	0.62	1.00	0.169	168	148	n.d.	1.71
5 Ciliwung River	–	–	–	–	–	–	–	–
6 Ciliwung River	307	1.11	1.90	6.15	125	115	1.23	29.9
7 Cikaniki River	301	0.91	1.03	2.02	211	210	1.21	3.33
8 Cikaniki River	520	0.83	1.15	2.87	52	462	0.232	4.39
Paddy field	279	0.78	1.59	2.10	225	191	1.88	4.55
Paddy sample	0.181	0.207	0.229	0.268	11.5	0.518	0.154	0.578

n.d. and – means not detected and not determined, respectively

Table 4 Total and inorganic Hg contents (μg/g) in sediments and paddy around the Cikaniki River

Site	Total Hg	Inorganic Hg
7 Cikaniki River	1.07	0.28
8 Cikaniki River	0.83	0.23
Paddy field	0.63	0.33
Paddy	0.08	0.065

2006). Thus, further monitoring of Cd in paddies should be continued because the detected Cd may have chronic toxicity in humans.

In West Java, there are several rivers contaminated by Hg which is used to extract gold from gold ore. Hg waste, especially organic Hg, causes Minamata disease, one of the most serious pollution-triggered diseases. Lots of babies were born with abnormalities owing to Hg pollution in the area of Minamata Bay in Japan (Kudo et al. 1998). In the Cikaniki area, it is estimated that about 4.8 tons of Hg is dumped into the river per month. The river water then flows to the mouth of river and passes many villages along the way. However, as shown in Table 2, Hg contents detected in the river water were relatively low as compared with the environmental baseline for total Hg in Indonesia (below 1 ppb). On the other hand, although the baseline value of the sediments was below 1 ppm, total Hg concentrations in sediments were from 0.63 to 1.03 ppm. These values mean that the mining area was not so safe from the viewpoint of health. Generally, organic Hg is more toxic than inorganic Hg. We found that organic Hg was present at low levels (Tables 2 and 3). However, in the sediments, the organic Hg accounted for 50% to 70% of total Hg (Table 4). Moreover, Hg is concentrated in the food chain, and inorganic Hg may change to organic Hg in living organisms (Sanfeliu et al. 2003). In addition, in other rivers in West Java, total Hg contents were reported to be from 0.14 to 2.35 ppb in river water, 4.98 to 6.13 ppm in sediments and 441 to 642 ppb in fish and shrimp in the downstream of the Cisadani River (Yustiawati et al. 2006). Thus, the accumulated Hg in fish and shellfish should be monitored.

In this study, total Hg content in the paddy was 0.08 ppm (Table 4). This value was about 16 times higher than that in the basin of the Agano River which became known as the second Minamata disease area in 1974 (Nakagawa and Yumita 1998). It was reported that the Hg contents in the paddy field and rice were about 0.15 and 0.005 ppm, respectively. The Hg intake from rice is well below the safe guideline level (0.036 mg Hg/day) in Japan (200 g rice/day per person). In Indonesia, consumption of rice is 2–3 time higher than in Japan. Therefore the total Hg intake from rice in Indonesia (500 g/day per person) is calculated to be around 0.040 mg Hg/day. This value exceeds the safe guideline level, and thus the rice from paddies beside the Cikaniki River may affect human health. Of course, since unpolished rice will be polished, the uptake of Hg will be low. However, consumption of Hg polluted rice must be studied in more detail.

Acknowledgments This research was supported by Grants-in-Aid from the Japan Society for the Promotion of Science (No. 18404004 and No. 23406021 for Kurasaki), and by the JST-JICA Project: Wild Fire and Carbon Management in Peat-Forest in Indonesia.

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