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Title	Metal Concentrations of River Water and Sediments in West Java, Indonesia
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Citation	Bulletin of Environmental Contamination and Toxicology, 87(6), 669-673 https://doi.org/10.1007/s00128-011-0411-z
Issue Date	2011-12
Doc URL	http://hdl.handle.net/2115/47657
Rights	The final publication is available at www.springerlink.com
Туре	article (author version)
File Information	YsudaBECTHUSCAP.pdf



1	Metal Concentrations of River Water and Sediments in West Java, Indonesia
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20	Abstract. To determine the water environment and pollutants in West Java, the contents of metals and
21	general water quality of the Ciliwung River in the Jakarta area were measured. High Escherichia coli
22	number (116-149/ml) was detected downstream in the Ciliwung River. In addition to evaluate mercury
23	pollution caused by gold mining, mercury contents of water and sediment samples from the Cikaniki
24	River, and from paddy samples were determined. The water was not badly polluted. However, toxic
25	metals such as mercury were detected at levels close to the baseline environmental standard of Indonesia
26	(0.83 to 1.07 $\mu$ g/g of sediments in the Cikaniki River). From analyses of the paddy samples (0.08 $\mu$ g/g), it
27	is considered that there is a health risk caused by mercury.
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29	Key Words. mercury contamination; paddy; water quality; West Java

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31River water is used for drinking, irrigation for agriculture and fish culture. However, water pollution 32caused by chemical substances such as metals is a serious problem for the inland water environment in 33 many countries, especially developing nations. Southeast Asia is industrializing but this process affects 34tropical rain forests, rivers, and mangroves. As a result of various developments such as the "green 35revolution" and use of chemical fertilizers, income has increased (Estudillo and Otsuka 1999). However, 36 unemployment and destruction of the ecosystem have also increased (Shiva 1991). Therefore, in 37developing countries, the idea of "sustainable development" is gaining currency. Forty years ago in Japan, 38 as a negative effect of industrialization, there were severe problems with Cd and Hg pollution in Toyama 39 and Minamata (Kimura 1988). Recently, similar phenomena have occurred in Southeast Asia (Hutagalung 40 1987). This study focused on Indonesia because, in West Java, the rivers also play important roles as 41traffic arteries and in economic activities. Studies of the quality of water and sediments are needed to 42evaluate environmental conditions. In the Jakarta gulf, Hg contamination has been reported (Hutagalung 431987; 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.

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## 55 Materials and Methods

56 This study was conducted at ten sampling sites in the Ciliwung River and Cikaniki River in West Java,

57 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

59 samples in a rice paddy were obtained near a gold amalgamation plant in the upstream area of the

- 60 Cikaniki River (Cisarua Village, site [St.] 9).
- 61 The pH, conductivity, chemical oxygen demand (COD), NO<sub>2</sub>, NO<sub>3</sub> and PO<sub>4</sub> levels in the water samples

62 were measured immediately at each sampling point with a pH meter (Shindengen, model pH boy-KS701,

- 63 Japan), a specific conductivity meter (Iuchi model TDS-can3, Japan), and simple water quality test
- 64 packages (Kyoritsu Chemical-Check, WAK-Cl, Japan) according to their instruction manuals. E. coli
- 65 were counted using E. coli detection paper (Shibata, Japan).
- 66 Before the determination of metal contents in water samples, 5 ml of ultrapure concentrated (conc.)

67	HNO <sub>3</sub> was added to 5 ml aliquots of samples for measurement of toxic metals (Sigma-Ardrich). The
68	contents of Mg, Mn, Al, Co, and Pb in the samples were analyzed with an inductively coupled plasma
69	mass spectrometer (ICP-MS, Seiko SPQ-6500, Tokyo, Japan). The detection limit of each metal is around
70	0.1 to 1 ng/ml. To extract the metals from the sediments, 50 ml of 0.1 M $HNO_3$ were added to 10 g of
71	dried sediment, and subsequently the mixture was agitated for 24 hr. The supernatants were collected after
72	centrifugation at 3,000 rpm for 15 min. Twenty paddy samples (about 0.4 g each) from ickers were
73	completely digested with 10 ml of conc. HNO3, and supplemented up to 100 ml with distilled water. To
74	remove the insoluble materials, the digested solution was filtered with a 0.45 $\mu m$ Millipore filter (USA).
75	The contents of Mg, Mn, Al, Co, Cd, Pb, Fe, Cu and Zn were then measured with the ICP-MS. In addition,
76	inorganic Hg contents in the same samples were measured using a Hiranuma HG 300 Mercury Analyzer
77	(Hiranuma Sangyo Co., Ltd., Japan: detection limit 0.1 ng/ml). Total Hg contents were determined as
78	follows: to degrade the organic compounds, 1 ml of conc. $\mathrm{H}_2\mathrm{SO}_4,$ 1 ml of conc. $\mathrm{HNO}_3$ and 2 ml of
79	$KMnO_4$ (50 g/L) were added to 30 ml of the above-mentioned acidified sample. The mixture was shaken
80	for 15 min, and heated at 95°C for 2 hr. After cooling, 1 ml of hydroxylamine chloride was added to
81	neutralize the excess KMnO4. The neutralized solution was filtered with a 0.45 $\mu m$ Millipore filter. The
82	filtered solution was diluted up to 100 ml with distilled water and the Hg contents were measured using a
83	Hiranuma HG 300 Mercury Analyzer.

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## 85 **Results and Discussion**

86	Table 1	General	water	quality	of the	rivers	in	West	Java,	Indonesia	a
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Site No.	Altitude (m)	Conductivity (µS/cm)	COD (mg/L)	NO <sub>2</sub> (mg/L)	NO <sub>3</sub> (mg/L)	PO <sub>4</sub> (mg/L)	E.coli (no./ml)	рН
1 Ciliwung River	1178	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.

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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

94ranges of NO<sub>2</sub> and NO<sub>3</sub> concentrations were from 0-0.63 mg/L and 0.1-15 mg/L, respectively. The middle 95 reach of the Ciliwung River at St. 3 had the highest values for them. It was interesting that a low 96 concentration of NO<sub>x</sub> was observed downstream in the Ciliwung River in the city of Jakarta, at Sts. 5 and 97 6. In addition, the highest phosphate concentrations, 2 mg/L were detected in the downstream area of the 98 Ciliwung River Sts. 5 and 6. The number of E. coli in the river water was relatively high because 99 domestic waste-water was discarded directly into the river. As expected, E. coli were found at all 100sampling points. In the downstream of the Ciliwung River, the level of E. coli was extremely high. These 101results indicated that contaminants due to domestic waste-water, for example, PO4 and E. coli, increased 102 in a distance-dependent manner from the upstream of the river. On the other hand, only NOx, which is an 103indicator for organic materials and pesticides, was increased in the middle reach of the river. From St. 2, 104the number of rice paddies and dairy farms increased.

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106	Table 2 Metal	concentrations	$(\mu g/L)$	ın	river wat	ter in	west Ja	iva,	Indonesia

Site No.	Mg	Mn	Co	Al	Pb	Total Hg	Inorganic Hg
1 Ciliwung River	3150	34.0	26.5	36.6	2.47	n.d.	—
2 Ciliwung River	4035	32.5	25.9	51.7	2.55	n.d.	_
3 Ciliwung River	3930	32.5	25.7	35.9	3.22	n.d.	_
4 Ciliwung River	3960	36.8	26.1	33.8	3.19	n.d.	_
5 Ciliwung River	4670	200.8	25.6	44.1	3.55	n.d.	_
6 Ciliwung River	4520	395.8	25.3	35.6	3.88	n.d.	_
7 Cikaniki River	1380	36.2	25.8	160.9	3.28	0.218	0.185
8 Cikaniki River	1660	34.8	25.8	139.8	3.34	0.119	0.096

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"n.d." and "-" mean not detected and not determined, respectively.

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 121 concentration of Cd was detected. The Hg concentrations in sediments at 3 sites of the Cikaniki River (St. 122 7, St. 8 and the paddy field) and rice are shown in Table 4. The total Hg concentrations ranged from 0.63 123 to 1.07  $\mu$ g/g, and the inorganic Hg concentration ranged from 0.23 to 0.333  $\mu$ g/g. Unlike in the water 124 samples, organic Hg accounted for 50 to 70% of the Hg in the sediments. Metal concentrations in the 125 paddy samples are shown in Table 3. These concentrations were not so high. However, the total and 126 inorganic Hg concentrations in the paddy samples were 0.08 and 0.065  $\mu$ g/g, respectively (Table 4).

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128 Table 3 Metal concentrations ( $\mu g/g$ ) 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

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"n.d." and "-" mean not detected and not determined, respectively.

131As shown in Table 1, COD levels and the number of E. coli were quite high. The results for general 132water quality in the present study were in good agreement with the results reported by Kido et al. (2007) 133 and Kurasaki et al. (2000). It was thought that river water in West Java contained high levels of organic 134compounds 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 135136Kido et al. (2009). The high microbial contamination in the river water is a health risk to the inhabitants. 137Thus, to enhance the river water environment in West Java, sewage and drainage systems need to be 138established as soon as possible.

139

140 Table 4Total and inorganic Hg contents ( $\mu$ g/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

141To examine whether metal pollution occurred, metal concentrations were measured in water and 142sediment samples. As a result, the concentration of Mn in the Ciliwung River at Sts. 5 and 6 was found to 143be about 10 times higher than at the other sites. This value exceeded the environmental standard value. 144Although Mn is an essential trace element, excess uptake of it may result in some nerve disorders 145(Benedetto et al. 2010). Toxic metals such as Pb were scarcely observed in the water samples. As shown 146in Table 3, relatively high Cd contents were detected in sediments at all the sampling sites. Since Cd was 147also detected in the upstream area, it was considered that this metal was naturally present in the 148environment rather than due to contamination in West Java. In Japan, a serious illness due to Cd pollution 149called "Itai-itai disease" is well known. The disease is considered to be due to Cd contained in rice 150paddies. In 2006, the international baseline value for paddies growing rice was fixed at less than 0.4 ppm 151(Yu et al. 2006). The Cd content in the paddy in this study was lower than this baseline value (Table 3). 152However, the value was about 3 times higher than that in Japan (Politic White Paper of Yamaguchi 153Prefecture, 2006). Thus, further monitoring of Cd in paddies should be continued because the detected Cd 154may have chronic toxicity in humans.

155In West Java, there are several rivers contaminated by Hg which is used to extract gold from gold ore. 156Hg waste, especially organic Hg, causes Minamata disease, one of the most serious pollution-triggered 157diseases. Lots of babies were born with abnormalities owing to Hg pollution in the area of Minamata Bay 158in Japan (Kudo et al. 1999). In the Cikaniki area, it is estimated that about 4.8 tons of Hg is dumped into 159the river per month. The river water then flows to the mouth of river and passes many villages along the 160way. However, as shown in Table 2, Hg contents detected in the river water were relatively low as 161compared with the environmental baseline for total Hg in Indonesia (below 1 ppb). On the other hand, 162although the baseline value of the sediments was below 1 ppm, total Hg concentrations in sediments were 163from 0.63 to 1.03 ppm. These values mean that the mining area was not so safe from the viewpoint of 164health. Generally, organic Hg is more toxic than inorganic Hg. We found that organic Hg was present at 165low levels (Tables 2 and 3). However, in the sediments, the organic Hg accounted for 50% to 70% of 166 total Hg (Table 4). Moreover, Hg is concentrated in the food chain, and inorganic Hg may change to 167organic Hg in living organisms (Sanfeliu et al. 2003). In addition, in other rivers in West Java, total Hg 168contents were reported to be from 0.14 to 2.35 ppb in river water, 4.98 to 6.13 ppm in sediments and 441 169 to 642 ppb in fish and shrimp in the downstream of the Cisadani River (Yustiawati et al. 2003). Thus, the 170accumulated 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 to 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.

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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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## 186 References

- Benedetto A, Au C, Avila DS, Milatovic D, Aschner M. (2010) Extracellular dopamine potentiates
  Mn-induced oxidative stress, lifespan reduction, and dopaminergic neurodegeneration in a
  BLI-3-dependent manner in Caenorhabditis elegans. PLoS Genet 26:e1001084
- Estudillo JP, Otsuka, K. (1999) Green revolution, human capital, off-farm employment: changing sources
- of income among farm households in central Luzon, 1966-94. Economic Develop Cultural Change
  47:162-168
- Hutagalung HP. (1987) Mercury content in the water in marine organisms in Angke Estuary, Jakarta Bay.
  Bul Env Contam Toxicol 39:406–11
- 195 Kido M, Yustiawati, Syawal MS, Sulastri, Hosokawa T, Tanaka S, Saito T, Iwakuma T, Kurasaki M.
- (2009) Comparison of general water quality of rivers in Indonesia and Japan. Environ Monit Assess197 156:317-329
- 198 Kimura I. (1988) Aquatic pollution problems in Japan. Aquat Toxicol 11, 287-301
- 199 Kudo A, Fujikawa K, Miyahara S, Zheng J, Takigami H, Sugahara M, Muramatsu T. (1998) Lessons from
- minamata mercury pollution, Japan After a continuous 22 years of observation, Water Sci Tech
   38:187-193
- Kurasaki M, Hartoto DI, Saito T, Suzuki-Kurasaki M, Iwakuma T. (2000) Metals in water in the Central
  Kalimantan, Indonesia. Bull Environ Contam Toxicol 65:591-597.
- 204 Mahbub T, Kuslan M. (1997) In Pemda DKI Jakarta, Studi Potensi Kawasan Perairan Teluk Jakarta.
- 205 Paper presented on Pemantauan Kualitas Lingkungan Jakarta tahun 1996 (workshop on Jakarta
- 206 environmental quality monitoring), held in Jakarta.

207 Nakagawa R, Yumita Y. (1994) Change and behavior of residual mercury in paddy soils and rice of Japan.

- 208J Applie Crystallograph 17:263-8522
- Sanfeliu C, Sebastià J, Cristòfol R, Rodríguez-Farré E. (2003) Neurotoxicity of organomercurial
   compounds. Neurotox Res 5:283-305
- 211 Shiva V. (1991) The violence of the green revolution ecological degradation and political conflict.
- Ecologist 21:57-60.

- 213 Yu H, Wang J, Fang W, Yuan J, Yang Z. (2006) Cadmium accumulation in different rice cultivars and
- screening for pollution-safe cultivars of rice Sci Total Environ 370:302-309.
- 215 Yustiawati, Syawal MS, Terashima M, Tanaka S. (2006) Speciation analysis of mercury in river water in
- 216 West Java, Indonesia. TROPICS 15:425-429
- 217
- 218 Figure Caption
- 219 Fig. 1 Sampling sites in West Java, Indonesia



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