

## Mercury Pollution Related to Artisanal Gold Mining in North Sulawesi Island, Indonesia

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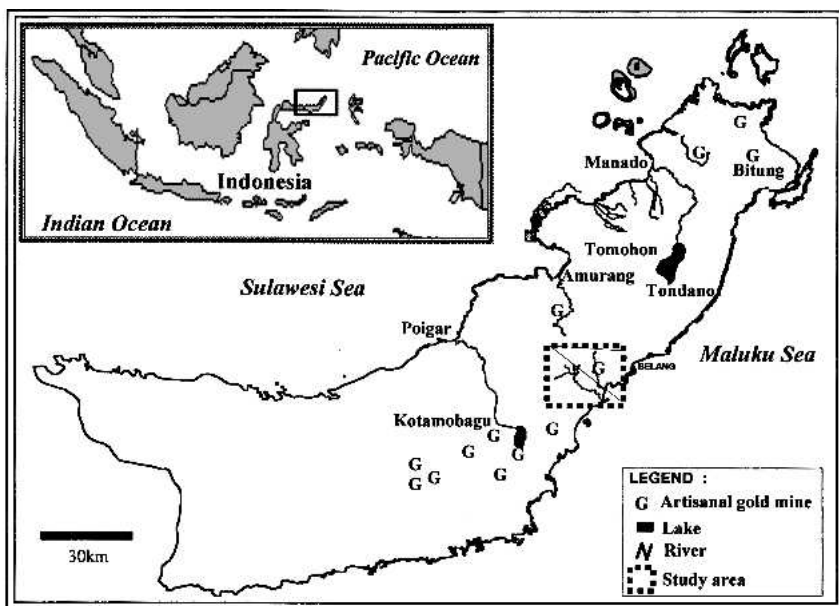
Mercury pollution may have several sources, ranging from those of natural to anthropogenic origins. All types of mercury pollution will concentrate in water pathways consisting of rivers, lakes, coastal waters, with the ocean ultimately becoming the final depository area. In the environment, inorganic mercury may become methylated particularly in soil and sediment. Under normal conditions, only a maximum of 5 % of inorganic mercury is methylated. This rate may increase under certain conditions depending on the organic material content and pH of the soil or sediment. In water systems, methyl mercury is the most important form, which is readily accumulated by biota and magnified through the food chain. Most of the mercury that accumulates in fish tissue is methyl mercury, and fish are probably the most common indirect exposure route by which mercury affects humans. In humans, methyl mercury can cause damage to the neurological, excretory, and reproductive systems. Accordingly, methyl mercury is the mercury form of greatest toxicological concern (Samoiloff 1989, Akagi and Nishimura 1991, Ikingura and Akagi 1996, Clark 1997, Lodenius and Malm 1998).

In order to prevent mercury contamination in early stages, periodic monitoring of the environmental mercury level is necessary. This study has been carried out to provide baseline information regarding mercury contamination of the aquatic environment surrounding the artisanal gold mining in the Ratatotok area located on the southeast coast of North Sulawesi peninsula, Indonesia. The study was carried out in late 2000 and involved observations of the artisanal mining operations, demonstration of the retort used in the gold-mercury amalgam burning, and the determination of the total amount of mercury in soil taken in samples from the gold processing units, in water and sediment samples taken along the impacted Totok River, and from fish and bivalves collected from Totok Bay.

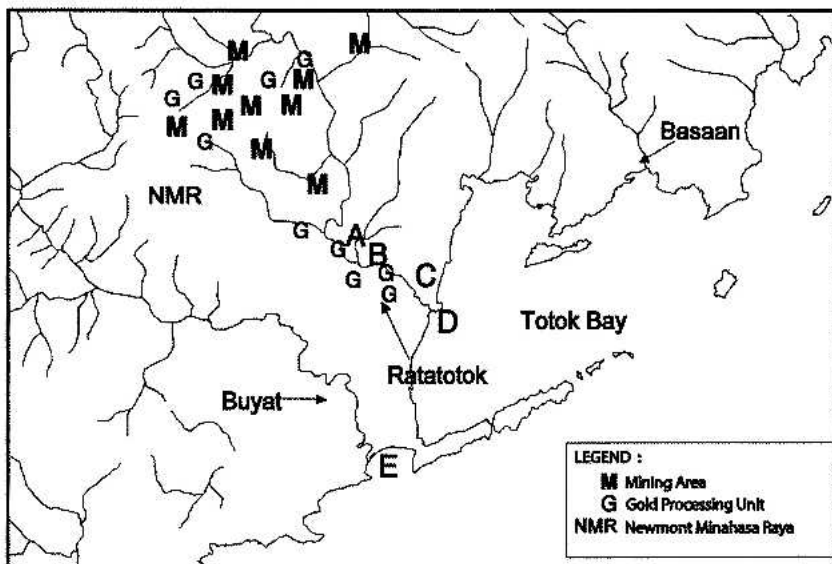
## MATERIALS AND METHODS

The present study was conducted in the Ratatotok area located on the southeast coast of the North Sulawesi peninsula of Indonesia facing the Maluku Sea (Fig. 1). Its topography consist of plains at the coastal area and becomes hilly several

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**Figure 1.** Study area in the North Sulawesi Peninsula, Indonesia.



**Figure 2.** Sampling sites in the Ratatotok area.

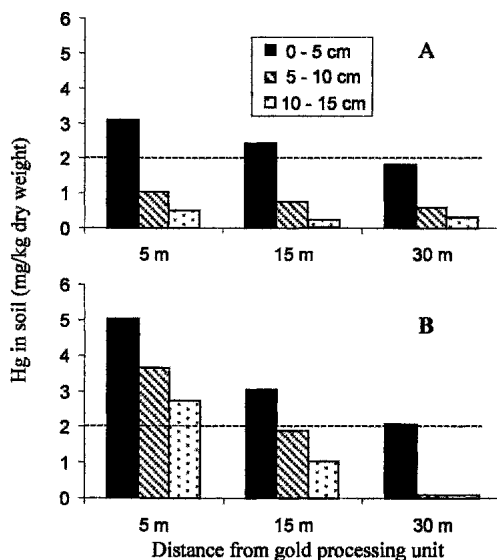
hundreds meters from the shoreline. Mangrove forests and coral reefs are spread along the shoreline. The gold mining area is located about 500 m from sea level on hill sites. Artisanal mining takes place on the upper reaches of the Totok River which runs into Totok Bay, and in the area mined by the Newmont Minahasa Raya Company which is located on the upper reaches of the Buyat River that runs into Buyat Bay (Fig. 2).

In order to investigate mercury contamination of the water system, water and sediment samples were collected from four sampling sites along the Totok River and one additional site at the estuary of the Buyat River (Fig. 2). Due to the difficulties in access to the up-stream sides of the gold processing units, all of the sampling sites in Totok River were located downstream of the processing units. Fish and bivalve samples were caught within Totok Bay by selected local fishermen. Totok Bay is thought to be a reservoir as an end point of all drains from the mining area. Water samples were taken using pre-labeled and clean 250 ml Nalgene polyethylene bottles. Sediment samples were taken by a sediment core sampler down to 30 cm from the sediment surface, and packed in three different layers 0-10 cm, 10-20 cm, 20-30 cm in pre-labeled 250 ml plastic bottles. The muscles of scad *Decapterus* sp. (four specimens; 31.0-31.6 cm in TL), grouper *Epinephelus merra* (two specimens; 15.7-22.0 cm in TL), barracuda *Sphyrna* sp. (one specimen; 23.3-42.9 cm in TL), parrot *Scaridae* sp. (two specimens; 15.8-18.1 cm in TL), snapper *Lutjanus* sp. (three specimens; 18.0-20.6 cm in TL) and giant clam *Tridacna* sp. (three specimens; 17.4-23.5 cm in shell length) were dissected for mercury analysis. All water samples were immediately cooled, while both sediment and biological samples were frozen -20 °C until chemical analysis.

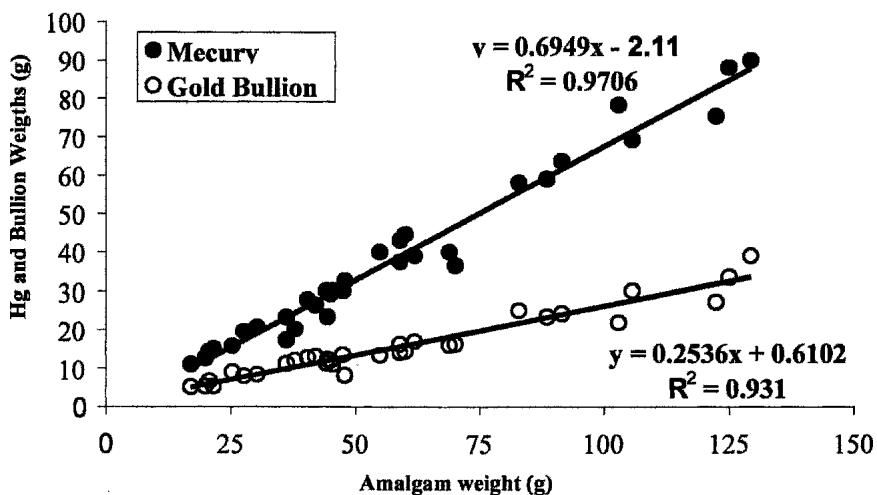
Water samples were filtered through a disposable 0.45 µm portable polycarbonate filtration unit. After filtration, water samples were treated by the cold-oxidation of acidified samples using bromine monochloride prior to the reduction of the samples with stannous chloride. Sediment samples were digested using a 1:1 ratio of nitric acid and hydrochloric acid, along with hotplate or microwave heating. Biological samples were digested using nitric acid and hydrogen peroxide along with hotplate heating. Total Hg for all samples were determined by the cold vapor technique using a Sansou Automatic Mercury Analyzer Model HG-3000, according to the method suggested by Akagi and Nishimura (1991). Detection limits for water, sediment, and fish samples were 10<sup>-2</sup> µg/l, 1 µg/kg dry weight, and 1 µg/kg wet weight, respectively. Quality control was monitored for all chemical analyses. Instrument calibration was verified by analyzing certified calibration solutions during each instrumental run. These external reference standards were generally within 92 % to 103 % of the nominal concentrations. All of the sample spikes for water, sediment, and fish samples were within 90 % to 113 % recovery. Preparation blanks were prepared to detect potential contamination during the digestion procedure. These preparation blanks measured below the detection limit.

## RESULTS AND DISCUSSION

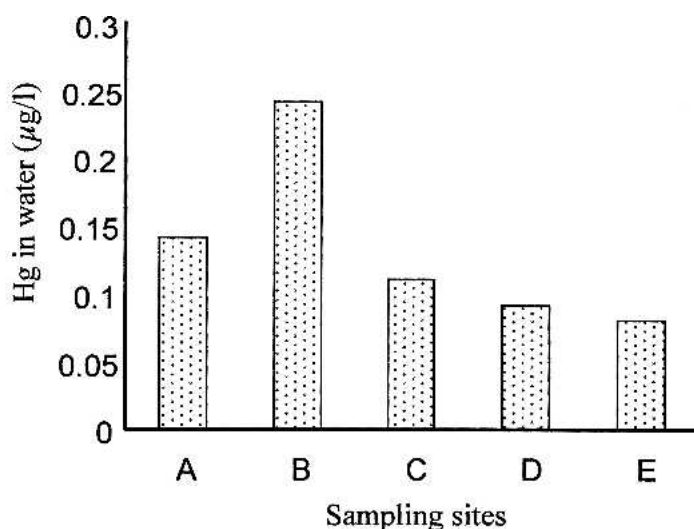
The results from soil sample analysis and the tests regarding the retort used for burning of the gold-mercury amalgam provide strong evidence that a significant amount of mercury used in the processing unit is released into the environment by the current processing practices. As depicted in Fig. 3, the soil surrounding the gold processing units is contaminated by mercury. The concentration of mercury in soil is high at sites close to the processing units, and gradually decreases towards beyond them. The vertical gradient of mercury level found in the soil samples also confirmed that the processing units are the primary source of mercury emissions.



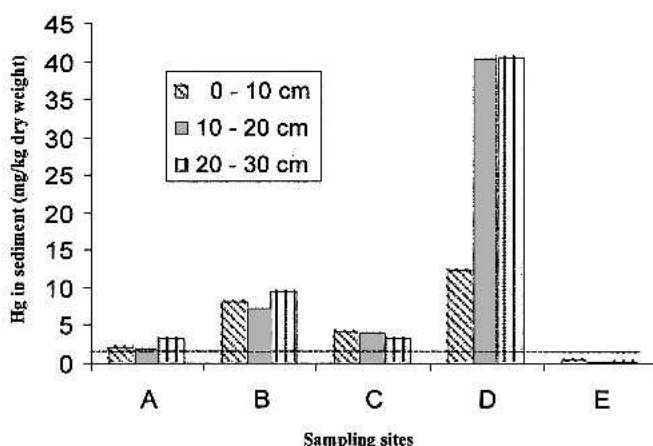
**Figure 3.** Mercury content in soil from the Ratatotok artisanal gold mining area. A and B indicate the sampling sites correspond to Fig. 2. The dotted lines indicate the recommended safety level (WHO/ICPS 1990).



**Figure 4.** Relation between recapture mercury and gold bullion and amalgam weight.

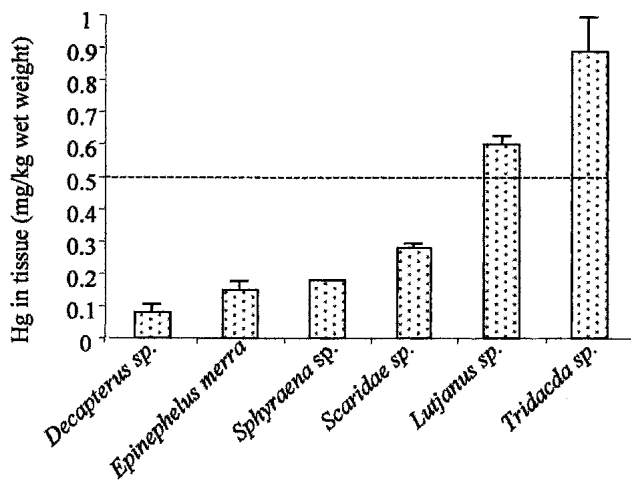


**Figure 5.** Mercury content in water from the Totok River and the Buyat River. Sampling sites correspond to Fig. 2.



**Figure 6.** Mercury content in sediment from the Totok River and the Buyat River. Sampling sites correspond to Fig. 2. The dotted line indicates the recommended safety level (WHO/ICPS 1990).

Tests for burning of the gold-mercury amalgam used retort were successfully conducted 33 times at 15 processing units. Data recorded from those tests provide a clear explanation about the amount of mercury vapor lost to the environment during amalgam burning. Some of the owners of the gold processing units were convinced by the results of the retort tests. They were encouraged to use retort for amalgam burn process because it is not only improves work safety but also provides an economic advantage. The regression line between weights of recaptured mercury and amalgam



**Figure 7.** Mercury content in fish and bivalves from Totok Bay. The dotted line indicates the recommended safety level (WHO/ICPS 1990).

(Fig. 4) with a high regression coefficient revealed that the amalgam practices maintain a constant proportion of the amount of mercury vapor released to surrounding atmosphere which is approximately 70 % of the amalgam weight.

Mercury released into the environment can either remain close to its source for long period, or widely disperse (Glass et al. 1990, Watts 1998). In the case of the Ratatotok artisanal mining area, the concentrations of mercury found in the soil surrounding the processing units (Fig. 3) appear relatively low when considering the estimated amount of mercury lost from the processing units that had already operated for nearly two years. This suggested that the mercury-enriched sediment from gold processing units apparently remains nearby for a while, and then is released into the watershed during rainy periods. The steep hill topography and wet climate of the Ratatotok area (JICA 2002) are expected to generate a strong flow even during a light rain. This in turn will concentrate the mercury-enriched sediment at the river mouth. As revealed in Fig. 6, the mercury levels in the sediment from the mouth of the Totok River are approximately 20-fold greater than the recommended safety level of 2 mg/kg for sediment (WHO/ICPS 1990).

The mercury level in water at all sampling sites of the Totok River were quite far below the recommended safety level of 1 µg/l (Fig. 5). A comparison of the high levels of mercury in the sediment samples (Fig. 6) suggested that if mercury-enriched sediment is dominant in the elemental and amalgam phases, then the particles are easily transported into the watershed because the ore handled by the artisanal miners is mostly silica-carbonate, since the hilly area of Ratatotok is composed of clay and rocky soil (JICA, 2002). In addition, due to technical limitations, the depths of nearly all of the mine shafts were less than 50 m, while sulfate deposits are more abundant at deeper layers, as indicated from the ore taken by the Newmont Minahasa Raya Company. If the mine wastes contain small amounts of sulfide deposits, such wastes



form mercury-enriched acid mine drainage. In this form, mercury is initially transported in a dissolved state and as colloids. As the acidity of this stream is buffered by country rocks, the adsorption of mercury species on iron oxide phases, and clays removes the dissolved mercury species, and then particle transport becomes dominant. Mercury-enriched sediment accumulated at the mouth of the Totok River will be agitated by small to medium size sea waves and currents in Totok Bay. Besides of horizontal dispersion, this will lead to the vertical dispersion of heavier mercury particles into the sediment as indicated in Fig. 6 (Kramer 1988). Mercury levels in water and sediment from the mouth of the Buyat River are significantly small, agreeing closely with the field observation data indicating that no artisanal gold mining activities take place within the catchments area of this river.

The biological samples collected from Totok Bay consisted of fish (12 specimens) and mollusk (3 specimens). Fish samples were composed of five species while only one species of mollusk was collected. The results shown Fig. 7 vividly demonstrate that the long term accumulation of mercury-enriched sediment at mouth of the Totok River has been transferred into biological system, since 80 – 90 % of the total amount of mercury in fish tissue has been found to be organic mercury or methyl mercury (Samoiloff 1989). Taking into account that the tridacna is a filter feeder organism, mean value of the mercury level in tridacna samples, which is almost two-fold of the recommended safety level, provides an indication of the occurrence of mercury bio-accumulation on the bay. Moreover, a quite high level of mercury in carnivorous fish such as snapper must be a noticeable sign of the bio-magnification process through the aquatic food chain (Viswanathan et al. 1988, Akagi and Nishimura 1991).

## REFERENCES

- Akagi H, Nishimura H (1991) Speciation of mercury in the environment. In: Suzuki T, Imura N, Clarkson TW (eds) *Advances in Mercury Toxicology*, Plenum Press, New York, p 512
- Clark RB (1997) *Marine Pollution*. Fourth Edition. Oxford University Press, UK.
- Glass GE, Sorensen JA, Schmidt KW, Rapp GR (1990) New source identification of mercury contamination in the Great Lakes. *Environ Sci Technol* 24:1059-1069
- Harada M, Nakachi S, Cheu T, Hamada H, Ono Y, Tsuda T, Yanagida K, Kizaki T, Ohno H (1999) Monitoring of mercury pollution in Tanzania: relation between head hair mercury and health. *Sci Total Environ* 227:249-256
- Ikingura JR, Akagi H (1996) Monitoring of fish and human exposure to mercury due to gold mining in the Lake Victoria Goldfield, Tanzania. *Sci Total Environ* 191:59-68
- Kramer CJM (1988) Physical aspects of the aquatic environment. In: De Kruijf HA, De Zwart D, Ray PK, Viswanathan PN (eds) *Manual on Aquatic Ecotoxicology*, Kluwer Academic Publishers, Dordrecht, p 332
- Limbong D, Kumampung J, Rimper J, Arai T, Miyazaki N (2003) Emission and environmental implications of mercury from artisanal gold mining in North Sulawesi, Indonesia. *Sci Total Environ* 302: 227-236.
- Lodenius M, Malm O (1998) Mercury in the Amazon. *Environ Contam Toxicol* 157:25-52.
- Samoiloff MR (1989) Toxicity testing of sediments: Problems, trends and solutions. In: Nriagu JO, Llakshminarayana JSS (eds) *Aquatic Toxicology and Water*

- Quality Management, John Wiley and Sons Inc., New York, p292
- Viswanathan PN, Jaffery FN, Misra V, Chawla G (1988) Biomonitoring and water ecotoxicology: An over view. In: De Kruijf HA, De Zwart D, Ray PK, Viswanathan PN (eds) Manual on Aquatic Ecotoxicology, Kluwer Academic Publishers, Dortrecht, p 332
- Watts RJ (1998) Hazardous waste: source, pathways, receptors. John Wiley and Sons Inc., New York
- WHO (1990) Methylmercury (Environmental health criteria 101), International Program on Chemical Safety, WHO, Geneva