

Environmental Emission of Mercury During Gold Mining by Amalgamation Process and its Impact on Soils of Gympie, Australia

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Abstract—The aims of this study were to estimate the total amount of mercury released to the environment during 60 years of gold mining (1867–1926) at Gympie, Queensland, Australia and to measure the mercury levels in soil samples surrounding the mining activity. We estimated that 1902 tonnes of mercury was released to the environment and about 1236 tonnes of which was released to the air. The mean mercury in the soil samples in the vicinity of the Scottish battery varied from 1.07 to 99.26 $\mu\text{g g}^{-1}$ as compared to 0.075 $\mu\text{g g}^{-1}$ as background mercury concentrations. The maximum mercury concentration measured in sediments of the Langton Gully was 6.12 $\mu\text{g g}^{-1}$. These results show that large amount of mercury was used in this area during gold mining. Since mining is active in the area and Langton Gully flows into Mary River, we therefore, recommend that mercury concentration in air and fish should be monitored.

Key words: Mercury, soil, sediment, cold vapour atomic absorption spectrometry, environment.

Introduction

Mercury contamination is considered one of the worst hazards among anthropogenic impacts upon the global environment. Mercury is one of the few metal pollutants that causes human health problems; even death due to inhalation of vapour and ingestion of contaminated food has been reported. It has been well documented that bacterial methylation process can convert inorganic mercury to methyl mercury which accumulates in fish (MASON and MOREL, 1993). The consumption of mercury contaminated fish was associated with Minamata disease in Japan and Felt hat disease in England was associated with inhalation of mercury vapour that caused many deaths (MITRA, 1986). It is estimated that worldwide more than 1400 human have died and over 20,000 have suffered from mercury poisoning over the last 40 years, giving an illness with mortality rate range of 7–11% (D'ITRI, 1992).

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Mercury emission to environment is either from natural sources (estimated to be 2500 tonnes per year (tyr^{-1}), NRIAGU and PACYNA, 1988) or from anthropogenic sources (estimated to be 4000 tyr^{-1} , PORCELA *et al.*, 1996 cited in LACERDA, 1997). The use of mercury in gold and silver mining has been one of the major sources of anthropogenic emission of mercury to the environment before cyanidation process replaced amalgamation process. A recent study shows that in Brazil, 77.9 tyr^{-1} of mercury was released to the environment through amalgamation process used in gold mining in the Amazon region, which was 67% of the total mercury emission to the atmosphere in the country (LACERDA and MARINS, 1997).

The amount of mercury released to the environment during gold mining from mines that are presently not in operation is difficult to calculate accurately. However, data from mining areas where amalgamation is still in use have been used to calculate the mercury Emission Factor (EF) for the amalgamation process. The Emission Factor is the amount of mercury emitted to produce 1.0 kg of gold. Although EF values are influenced by the quality of gold ore and the climate of the mining area, they are useful for providing reasonable estimates of the total mercury emitted to the environment during gold mining in the past if the total amount of gold recovered is known. MALLAS and BENECITO (1986) interviewed miners in Para State in the northeastern Amazon region and reported that EF values could range from 2.0 to $4.0 \text{ kg Hg kg}^{-1}$ of gold. LACERDA and SOLOMONS (1991) reported EF to be 1.7 based on calculations by actually determining the Hg balance throughout the entire gold production process. WISE (1966) estimated that the mass of mercury consumed in the amalgamation process is of the same order as the mass of gold recovered ($\text{EF} = 1.0$) for 19th century mines in Victoria, Australia and MELLOR (1952) assumed EF higher than 2.0 for this technique. LACERDA (1997) used EF value of 1.5 to compare the rate of input of mercury in tyr^{-1} to the environment from different countries. There is overall agreement among researchers that 65–87% of the total emission of mercury is released to the atmosphere (LACERDA, 1997; MITRA, 1986).

In Australia, the amalgamation process for the extraction of gold was used from late 19th until middle of the 20th century (SMYTH, 1869; BYCROFT *et al.*, 1982). During this period, large amounts of mercury were released to the environment that resulted in contamination of many sites in Australia. The tailings from an abandoned gold mine on the Thomson River Victoria, Australia, contained $40\text{--}90 \mu\text{g g}^{-1}$ of mercury (MELBOURNE AND METROPOLITAN BOARD OF WORKS, 1975). Moreover tailings contained $88 \mu\text{g g}^{-1}$ of mercury at Wood Point and $120 \mu\text{g g}^{-1}$ of Hg at Blackwood, Victoria, Australia (BYCROFT *et al.*, 1982). There are studies from all over the world about soil contamination from mercury used in gold mining (GLOVER *et al.*, 1975; BYCROFT *et al.*, 1982; EPA Victoria, 1982; PFEIFFER and LACERDA, 1988; LACERDA, 1997). No published data were available for the Gympie gold mining area, Queensland.

According to Gympie Museum and Gympie and District Historical Society information sheets, alluvial gold was first discovered in October 1867 at Gympie,

Queensland, Australia by James Nash. Gympie is about 160 km from Brisbane. It was estimated that all the alluvial gold was exhausted by the end of 1868. Amalgamation was used to recover gold until all mining in Gympie ceased in 1923. A total of 1207.4 tonnes of gold was recovered during 60 years from 1867 to 1926, of which 2.6 tonnes was alluvial gold.

The aims of this study were (i) to estimate the total amount of mercury released to the environment during 60 years of gold mining (1867–1926) at Gympie, Queensland, Australia and (ii) to determine the mercury levels in Gympie soils and sediments from Langton Gully 75 years after the amalgamation process in the area ceased.

Experimental

Total mercury in samples was measured using the method described by ADELOJU *et al.* (1994) The details of the procedure are given below.

Sampling

Samples from 25 sites around the Scottish Gympie Battery were collected. Four sites (1–4) at some distance from the Battery were expected not to be directly contaminated; one site (5) from the shaft down to the above sites; two sites (6 and 7) from tailing heaps between the shaft and the battery; eight sites (8–15) around the Scottish Gympie Battery; two sites (16–17) from a dam close to the battery; six sites (18–23) along the Langton Gully; site one (24) from Old Victoria Battery and one site (25) from Deep Creek fossicking area. The study area is illustrated in Figure 1.

To collect a sample 4–8 holes, depending upon the area, were dug to 20 cm depth and approximately 4 kg of soil was thoroughly mixed and sampled by dividing samples into half each time until the required sample of about 100 g was obtained. Samples were transported to the laboratory in air-tight clean plastic containers at low temperature. Samples were air dried and ground to a fine powder to ensure sample homogeneity. Moisture in soil samples was determined by drying the samples at 80°C for 5 hours and water content in samples varied from 4 to 25%.

Reagents and Standards

All reagents used were of analytical grade and water was deionised. Mercury stock solution was prepared by dissolving 1.3535 g of HgCl_2 in 20% (v/v) HCl. Organic stock solution was prepared by dissolving an appropriate amount of methyl mercury chloride in ethanol (12 mL of 96% v/v) and diluting it to 100 mL with water. Stannous chloride solution (30% w/v) in 20% HCl was prepared daily and stabilised by adding a piece of tin. Working standards were prepared daily in 2% HCl. Hawkesbury River sediment (AGAL-10) reference material high in mercury was purchased from the Australian Government Analytical Laboratories (AGAL).

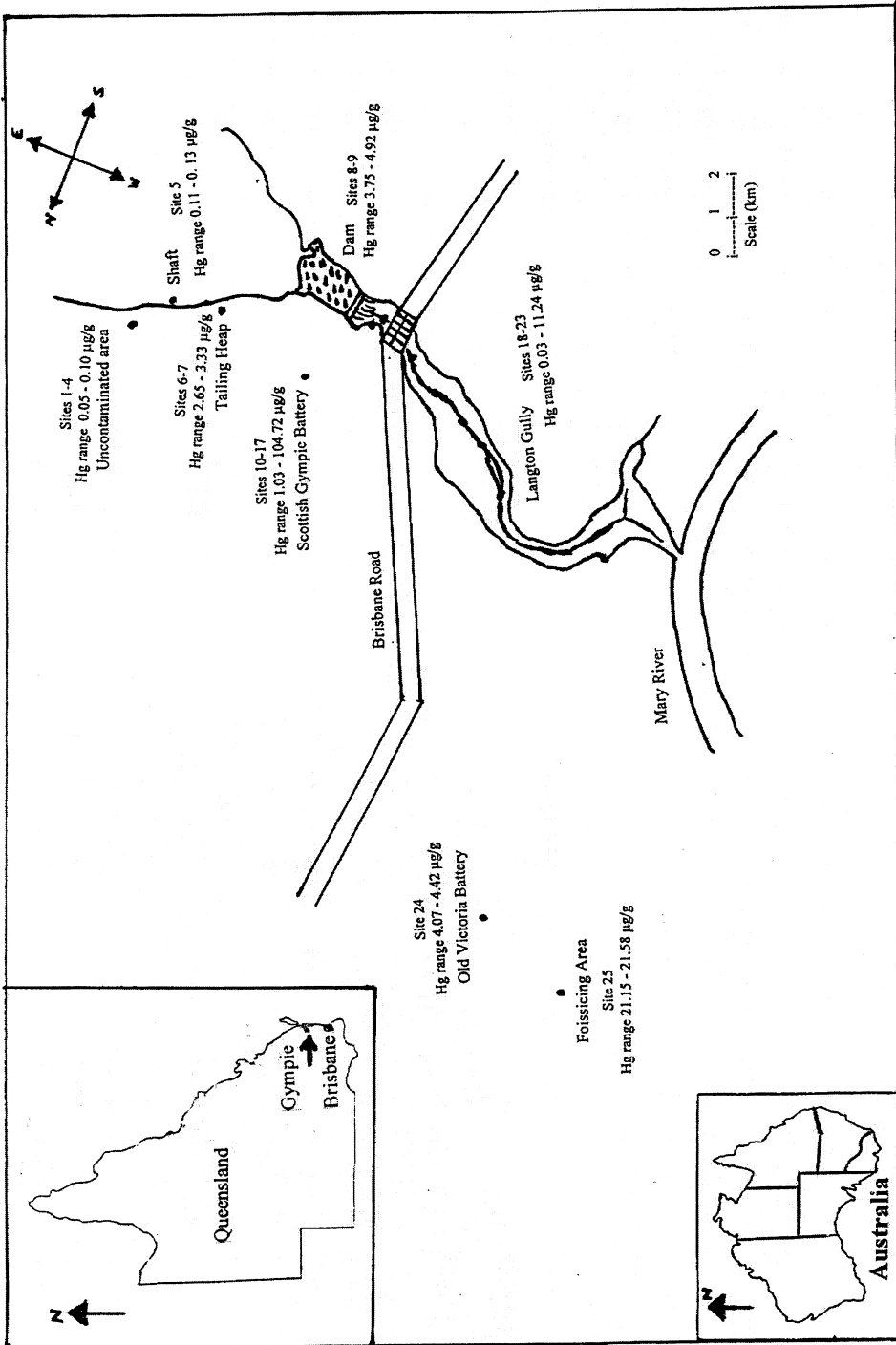


Figure 1
Study and sampling areas.

Sample Preparation

Soil sample 0.2 g of soil sample (after correction for moisture) was digested using 2.5 mL of digestion mixture (1 part of concentrated H_2SO_4 and 2.25 parts of concentrated HNO_3) in a 100 mL Erlenmeyer flask at 90°C in a water bath for one hour. The digest was diluted with water to a known volume by transferring it to a 100 mL volumetric flask. 1.0 mL of the diluted digest was transferred to a 100 mL Erlenmeyer flask containing 2.5 mL of the digestion mixture and 16.5 mL of deionised water in order to make the total volume of 20.00 mL.

CV-AAS Measurement

A Teflon coated small magnet was added to the above Erlenmeyer flask containing 20 mL of the sample solution and it was placed on a magnetic stirrer. The flask was then connected to a water reservoir and the mercury cell using a dreschel head. In all measurements, a total of 20.00 mL volume of the standard/digest and 1.0 mL of 30% (w/v) stannous chloride were stirred using magnetic stirrer for three minutes to concentrate mercury vapour in headspace. The mercury vapour concentrated in headspace was displaced at a rate of 10 mLs^{-1} from the flask into the cell using water displacement method. After recording the peak height signal, the cell was purged with compressed air to clean out any residual mercury in the cell.

Instrumentation and Glassware

A GBC Model 902 atomic absorption spectrometer (GBC, Australia) operated in a double-beam mode was used for mercury determination for all samples under the following conditions: hollow cathode lamp current 3 mA, slit width 0.5 nm, wavelength 253.7, signal integration time 10 s, with an open ended quartz cell at room temperature.

All glassware and plastic containers were soaked in nitric acid (2 M) for at least 24 hours and rinsed three times with distilled water and two times with deionised water before use, then dried in an oven at 100°C to remove all traces of water.

Results and Discussion

Amount of Mercury Emitted to the Environment

Table 1 shows that about 1902 tons of mercury was emitted to the environment at a rate of about 30 tyr^{-1} for sixty years from 1876–1926. The EF proposed by LACERDA (1997) was preferred over EF of 1.0 proposed by WISE (1966) for Victoria, Australia because climatically Gympie (Latitude 22°) Queensland is

Table 1
Amount of mercury emitted to environment

Gold Mined (t)	Emission Factor (EF)	Estimated Hg loss to environment (t)	Rate of Hg loss to environment (tyr^{-1})	Estimated Hg emitted to air (t)
1267.9	1.0 (WISE, 1982)	1267.9	21.2	824–1078
1267.9	1.5 (LACERDA, 1997)	1901.8	31.7	1236–1617

hotter by $7 \pm 2^\circ\text{C}$ than Victoria (Latitude 37°). Vaporisation of mercury to air increases exponentially with temperature and the vaporisation rate doubles for every 10°C increase of temperature. EF of 1.5 used in this study was lower than 1.7 calculated by LACERDA and SOLOMONS (1991) by actually determining the Hg balance throughout the entire gold production process. The rate 31.8 tyr^{-1} was 2–3 times the rates calculated for Bendigo (11 tyr^{-1}) and 4–6 times the rate for Victoria (5 tyr^{-1}), both in Australia (LACERDA, 1997). Moreover over 67% (1236.3 tons) of it was released into air inhaled by miners and populations in the surrounding areas. However, one should keep in mind that these estimates have been based on the amount of gold recovered from the area, which was obtained from official records. There is a strong possibility that all the gold metal mined from this area might have not been officially recorded. Based on this limitation, the total amount of mercury released into the environment is expected to be more than the estimated amount in this study. Moreover, the value of EF used in this study is an estimate based on other studies. The actual value was not known. The EF value is influenced by many factors such as the climatic conditions, management of the mining operations, quality of ore, etc. The EF value estimates might have influenced (increased or decreased) the total amount of mercury released into the environment.

Analysis Technique Variables: Detection Limit, Mercury in Reagents, and Recoveries

The absolute detection limit of 1 ng was obtained for the method. The mean background mercury concentration in reagents used for a measurement was $1.5 \pm 0.5 \text{ ng}$. The reliability and accuracy of the method was further evaluated by doing recovery studies on the reference material and a soil sample. The recovery of inorganic mercury from the reference material was $101.2 \pm 4.3\%$ ($n = 3$) and of organic mercury was $96.9 \pm 5.6\%$ ($n = 3$). The amount of mercury recovered from the reference material $12.68 \pm 0.75 \mu\text{g g}^{-1}$ was close to the certified value of $11.79 \pm 1.1 \mu\text{g g}^{-1}$. From the soil sample, the recovery for inorganic mercury was $93.9 \pm 2.3\%$ whereas for organic mercury was $81.2 \pm 0.2\%$. The recovery for organic mercury, although low, was within the acceptable range. Moreover, 98% of mercury in soils is in inorganic form (BARNETT *et al.*, 1995 and DAVIS *et al.*, 1997).

Background Mercury Levels in Soil Samples

Table 2 shows background mercury levels at four sites (1–4) which were at a considerable distance from the mining activity, therefore were considered to be less prone to mercury contamination. Mean mercury level of $0.075 \mu\text{g g}^{-1}$ of soil was considered as background mercury in soil samples. The background mercury concentrations in the samples were consistent with the results reported by DAVIS *et al.* (1997). According to them mercury levels in nonmercuriferous crustal soils and sediments in background areas, not directly impacted by anthropogenic discharges or volcanic emissions, range from 0.05 to $0.20 \mu\text{g g}^{-1}$. However, it is believed that mercury levels at sites 1–4 might also have been slightly elevated due to atmospheric precipitation of passively dispersed mercury in air from surrounding mining activity that involved amalgamation process. Nevertheless, the mean value of $0.075 \mu\text{g g}^{-1}$ of soil provided an excellent standard for comparing mercury contamination in the area.

Mercury in Soil Samples

The mean mercury concentration at site 5, Oriental Console Shaft, $0.12 \mu\text{g g}^{-1}$ of soil was slightly higher than the background, but was still low and within the acceptable background level. A slight increase in mercury concentration at this site over the background concentration ($0.075 \mu\text{g g}^{-1}$) might be the result of transport activity involved in moving tailings from the shaft to the battery. The samples collected from sites 6 and 7 were sand samples from tailing heap after cyaniding. Mercury levels in samples from these sites were much higher than the background levels. The mercury concentrations in these tailings were lower than reported by WISE

Table 2

Background mercury in Gympie soils

Site	1	2	3	4
No. of analyses	3	3	3	3
Mercury ($\mu\text{g g}^{-1}$)	0.10	0.07	0.08	0.05
Mean Deviation ($\mu\text{g g}^{-1}$)	0.01	0.01	0.00	0.00

Table 3

Mercury levels between the background and the battery sites

Site	5	6	7
No. of analyses	4	4	4
Mercury ($\mu\text{g g}^{-1}$)	0.12	2.80	3.16
Mean Deviation ($\mu\text{g g}^{-1}$)	0.01	0.13	0.18

(1966) in Victoria because some mercury from tailings is expected to be removed during the cyanidation process.

Mercury levels in the vicinity of Scottish Gympie Battery are reported in Table 4. The mean mercury levels in this area ranged from 1.07 to 99.26 $\mu\text{g g}^{-1}$ of soil. Extremely high levels were found around the battery indicating that large quantities of mercury were used in the direct vicinity of the battery. Site 8 was at some distance from the battery, therefore, it contains lower concentration of mercury than sites (9–11) adjacent to the battery. Moreover, at sites 8, 9 and 12, it appeared to have some land filling done that might have happened afterwards. Despite land filling, mercury levels were much higher than the background levels. Samples collected from sites 13 and 14, close to retort house were also high in mercury 12.82 $\mu\text{g g}^{-1}$ and 4.17 $\mu\text{g g}^{-1}$, respectively. There was no vegetation around this area which may be the result of mercury and/or cyanide contamination.

Mercury in Sediments

Mercury levels in the vicinity of water dam are reported in Table 5. Samples from site 15 consisted of sediments from a huge water tank above the battery, used for water supply for crushing ore at the battery. Higher levels of mercury in sediments from the water tank than in sediments from the dam indicate the accumulation of mercury in the water tank due to recycling of the water used for crushing the ore. Sites 16 and 17 were directly from the dam and the samples collected were muddy. A mean mercury level 4.3 $\mu\text{g g}^{-1}$ was found in samples collected from the dam. This concentration of mercury is about 57 times the background concentrations. Mean mercury concentrations in soil around the water reservoir were lower than those measured around the Scottish Gympie Battery (Table 5).

Table 4

Mercury levels around Scottish Gympie battery

Site	8	9	10	11	12	13	14
No. of analyses	4	4	4	4	4	4	4
Mercury ($\mu\text{g g}^{-1}$)	8.50	19.14	99.26	87.04	1.07	12.82	4.17
Mean Deviation ($\mu\text{g g}^{-1}$)	0.34	1.17	4.62	3.55	0.04	0.77	0.25

Table 5

Mercury in sediments of water reservoir used during mining

Site	15	16	17
No. of analyses	3	4	4
Mercury ($\mu\text{g g}^{-1}$)	10.79	4.72	3.93
Mean Deviation ($\mu\text{g g}^{-1}$)	0.41	0.21	0.23

Table 6
Mercury in sediments of Langton Gully

Site	18	19	20	21	22	23
No. of analyses	4	4	3	4	3	3
Mercury ($\mu\text{g g}^{-1}$)	4.04	3.47	2.68	6.12	0.13	0.04
Mean Deviation ($\mu\text{g g}^{-1}$)	0.26	0.05	0.09	0.11	0.01	0.01

Table 6 shows the mercury levels in the sediments collected from the Langton Gully. Langton Gully samples were collected from sites downstream from the dam behind the bridge on the Brisbane Road. Mean mercury levels in sediments from sites 18 to 21 ranged from $2.68 \mu\text{g g}^{-1}$ to $6.12 \mu\text{g g}^{-1}$. The mercury concentrations in the sediments suggest that the gully is contaminated. These concentrations are considerably higher than those reported in sediments of the Lerdererg River next to the Blackwood gold mine in Victoria, Australia (BYCROFT *et al.*, 1982). Sites 20 and 21 contained less mercury because the samples were clay samples and sampling at site 21 close to site 20 was done at a deeper level. Mean mercury concentrations in Langton gully sediments and soil samples from the water reservoir were comparable but lower than the samples from Scottish Gympie Battery (Table 4).

Mercury at Another Location

There were 600 heads of batteries in the Gympie region that used mercury coated copper plates to trap gold. Mercury concentrations in soils around these batteries are also expected to be high. To ensure the use of mercury at other sites we included samples from sites 24 and 25. Site 24 covered Deep Creek bank mud accumulated on ground bar below the Old Victoria Battery. The presence of $4.22 \mu\text{g g}^{-1}$ of Hg in the mud indicated the movement of mercury from the Old Victoria Battery. Site 25 was a fossicking area immediately below the Old Victoria battery. The mean mercury concentration at site 25 was greater than $21 \mu\text{g g}^{-1}$. The mercury levels at this site may further increase because the area is still open to the general public for gold fossicking, and some amateur fossickers have been reported to use mercury.

Implications

In general, soils in the vicinity of Scottish Gympie Battery were found to be contaminated with mercury even after 75 years since the use of mercury in the area ceased. The natural decontamination process for mercury is very slow (MITRA, 1986), since most of the mercury in the soil is elemental and in an inorganic form, it

Table 7
Mercury from Deep Creek fossicking area and from Old Victoria Battery

Site	24	25
No. of analyses	4	4
Mercury ($\mu\text{g g}^{-1}$)	4.22	21.37
Mean Deviation ($\mu\text{g g}^{-1}$)	0.17	0.24

vaporises and disperses in air. Therefore, the mercury concentration in air might increase as compared to the background mercury concentration in ambient air, which is 1.5 to 1.8 ng m^{-3} (BEAUCHAMP and TORDON, 1998). The concentrations of mercury vapour in air over contaminated soils and precious metal mines have been reported to reach 1500 ng m^{-3} at ground level and dust particles are also known to contain high concentrations of mercury (MITRA, 1986). Mercury is a neurotoxin. It is known that inhaled mercury accumulates in brain. The rate of accumulation of mercury in the brain is much higher than the disposal rate from the brain by the natural body mechanism (MITRA, 1986). Since mining is active in this area, therefore there is need to evaluate the concentration of mercury in air in the vicinity of the mining area.

Langton Gully directly downstream from the Scottish Gympie Battery was found to contain high mercury levels. Of particular concern, however, is the proximity of Langton Gully to Mary River. Several mercury-contaminated sites including shafts, tailing sites and Old Victoria Battery exist downstream of the Scottish Gympie Battery. Langton Gully is a permanent watercourse, it could be expected to carry mercury downstream to Mary River, where it may be transformed into organic form and accumulate in fish. The consumption of fish from the river may indeed increase mercury levels in humans. Research into mercury levels in the river sediments and fish is, therefore, warranted.

Conclusions

About 1902 tons of mercury were released to the environment in the vicinity of Gympie during 60 years of gold mining and 65–87% of this (< 1236 tons) was released into the air. High mercury levels were found in the soil around Scottish Gympie Battery. Mercury concentrations in contaminated soils in the area ranged from 0.11 to 104.72 $\mu\text{g g}^{-1}$ (Mean $12.00 \pm 21.14 \mu\text{g g}^{-1}$). High mercury concentrations in samples from Victoria battery indicate that soils around all the 600 batteries could also expected to be contaminated with mercury. Analyses of sediment samples show that Langton Gully was also contaminated as result of considerable amount of washed from tailings into the Gully. As the Gully is a permanent water-course to

Mary River, the mercury contamination is expected to extend to the river via a race. Moreover, it is believed that tailings were also directly dumped into the river. Therefore, there is a strong need to investigate mercury levels in air around the mining areas as well as sediments and fish from the river.

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