Research Article

Investigation of the spread of mercury in the land around the waste storage ponds in the Kulon Progo traditional gold mine

Ranno Marlany Rachman1*, Edward Ngii1, Abdul Rahim Sya’ban2

1 Department of Civil Engineering, Halu Oleo University, Kampus Hijau Bumi Tridharma, Kendari, 93232, Indonesia
2 Department of Science and Technology, Mandala Waluya University Jl. Jend. AH. Nasution, Kota Kendari, Sulawesi Tenggara 93561, Indonesia

*corresponding author: rannorachman@uho.ac.id

Abstract

This study was conducted to determine mercury contamination contained in soils in Kalirejo sub-district, Kulon Progo, Indonesia. This contamination occurred as a result of traditional gold mining activities that used the amalgamation method of mercury. Soil samples were collected from 6 sites; one site was located in an uncontaminated location, and 5 sites in contaminated soil were taken from 10 meters distance from the tailing ponds. Samples were collected from each site at 30, 60, and 90 cm depths. Mercury concentrations in each sample were measured according to the US EPA method, using Mercury analyzer type VM-3000. Mercury concentrations in uncontaminated area at 30, 60, 90 depths were 0.19 mg/kg, 0.02 mg/kg, and <0.0001 mg/kg respectively. These values did not exceed the quality standard according to Government Regulation No 101 The Year 2014 concerning Hazardous Waste Management of 0.3 mg/kg. Meanwhile, mercury concentrations around the tailing ponds were 0.30 to 22.51 mg/kg, which exceeded the quality standard.

Keywords:
mercury pollution soil traditional gold mining

Introduction

Traditional gold mining is one of the largest sources of mercury pollution (UNEP, 2008). Indonesia has many traditional gold mines, using the amalgamation process of mercury. There are 713 small-scale gold mines in Indonesia spread across Java, Sumatra, Kalimantan, and Sulawesi with 60,000 small-scale miners (Djamaluddin et al., 2012). Mining activities by the community are characterized by simple and relatively inexpensive exploration and exploitation techniques. Local people generally use simple traditional tools such as hoes, crowbars, hammers, and other traditional tools for their excavation or mining process (Rangkuti, 2013).

Mercury pollution from gold mining also occurs due to residual tailings spilled out during transportation and from a fully-fledged pond (Vidniarizki, 2014). Mercury becomes an important issue after the incident cases of Minamata, Japan in 1957 (Samson et al., 2013).

The amalgamation process in traditional gold mining is done by adding all the material (soil, water, mercury) simultaneously into a drum machine (Schimedt, 2012). Rocks containing gold ore mined soil are mashed to 1-2 cm size. The drum also reduces the ore grain size of coarse-grained ore (1 cm) to fine-grained (80-200 mesh) with its grinding machine in the form of iron bars. The refined soil is put in a drum containing water (1:1) and mercury. However, miners tend to add excessive water mercury mixture to extract more gold particles.

The addition of mercury is dependent on the content of the gold contained in the ore (Widodo, 2008). The drum is rotated with propulsion power or dynamo (Hasriyanty et al., 2018). The drum is closed tightly and rotated for 4-6 hours. After rotation, gold
amalgam is separated from the rock, water, and residual mercury by filtration. At the time of screening, large amounts of mercury are wasted in the surrounding area, which results in environmental pollution. The resulting amalgam of gold is roasted/burned to separate mercury from gold (Drace et al., 2012). In the amalgamation process, about 25-30% of the mercury is wasted in the environment (Veiga et al., 2009). Mercury released into the atmosphere can last for 6 up to 24 months before being deposited back into the ground (Dastoor and Larocque, 2004). Mercury added into the amalgamation tube (drum) is reduced up to 10% at the end of the process. This was carried over into the tailings at the leaching stage (Rohmana et al., 2006). Gold processing by amalgamation can potentially cause harm to the community’s health living near the mine site (Bansah et al., 2016). Other impacts of traditional gold mining are soil and water pollutants caused by chemicals that are used for separating gold from rock (Kpan et al., 2014). Gold mine waste may contain other hazardous toxic substances such as arsenic, cadmium, lead, cyanide, and others (Appel et al., 2014).

Traditional gold mining in the Kalirejo sub-district of Kulon Progo has been conducted since 1995. Traditional mining activities are included in the public mining area covering up to 25 hectares (Rozani, 2013). Gold in the Sangon Region is unevenly distributed. Gold and silver are distributed randomly with the concentration of 1 to 13.8 ppm Au and 5.4 to 63.2 ppm Ag (Gunawan et al., 2001). According to Wiriosudarmo (1999), Small Scale Mining (SSM) is defined as mining operations that are operated by a few sets of people or by a community (collective).

According to Setiabudi 2005, soil in the gold mining area in Sangon, Kulon Progo contains a high mercury level >50 ppm. The quality standard for mercury-uncontaminated soil according to Government Regulation No. 101/2014 is 0.3 mg/kg. In The USA, the total permitted amount of Hg in soils is 2.0 mg/kg (USEPA, 2007). Mercury content above 2 mg/kg in the soil is considered to be toxic (Mahbub et al., 2017). The Priority Substance List (PSL) puts mercury in third place of hazardous substances after arsenic and lead (ATSDR, 2013). Although previous publications have reported that mercury concentrations in the soil around the mining area exceed natural levels of mercury concentrations in soil, investigations around the gold mines in Kulon Progo area are still very limited. The purpose of this study was to determine the level of mercury contamination to soil in traditional gold mining areas in Kulon Progo. Data from this study will be used for contaminations control management efforts around the gold mining area.

Materials and Methods
Soil samples from the gold mining site in Kalirejo District, Kulon Progo, were collected in October 2015. Control soil samples (C1) were taken from one uncontaminated location, meanwhile, the soil was also sampled from contaminated areas in 5 specific locations (T1, T2, T3, T4, and T5) about 10 meters distance from the tailing ponds. Sampling was done by using a core sampler which refers to the US EPA method. Location coordinates of the sampling points were measured using a Global Positioning System instrument. All soil samples were taken at 30, 60, and 90 cm depths. Each sample was inserted into PET (PolyEthylene Terephthalate) plastic and labeled, then inserted into an icebox with a temperature of 4°C and brought to the laboratory for analysis of total mercury concentrations and tailing characteristics. The tailings sample location can be seen in Figure 1. All samples were analyzed for soil physical characterization based on particle size distribution analysis according to ASTM D422 (Withdrawn, 2016). The test for chemical characteristics, i.e. water content test, was done by a gravimetric method and the pH was measured by a pH meter. Particle size analysis methods and chemical characteristics of soil samples were performed at the Soil Mechanics Laboratory and Stone in Civil Engineering FTSP ITS. Soil classification was done according to (USDA, 1987) soil texture class system. Mercury level test was conducted at LPPT UGM Yogjakarta by using Mercury Analyzer Type VM-3000. All samples were analyzed twice.

Results and Discussion

Characteristics of soil samples
Samples of the contaminated soil at different depths showed different compositions of sand, silt, and clay (Table 1). The soil is dominated by sand, followed by silt. Clay exists in a low percentage. The distribution of soil particles varies in each soil sample, with an increasing percentage of clay in the deeper zone. The majority of soil texture is classified as loamy sand and loam in the upper parts and silty loam to clay in the lower parts of the soils. Sandy textured soils have relatively few adsorption sites if low in organic matter, phyllosilicate mineral, and hydrous oxide content (Aksakal et al., 2012). Types and textures of soils affected the heavy metal concentrations and heavy metals were relatively enriched in saline wet sand and silt soils and clayey soil (Hu et al., 2017). Soils that are dominated by sand can drain water quite high. Otherwise, soil that is dominated by clay can drain water very low. That is why the concentration of Hg in clay is high (Orhue and Frank, 2011).

Soil pH
pH values at each location were not much different (Table 2). The pH ranged from 6.74 to 8.56. Natural soils in the area are moderately normal with a pH of 6.74, while the pH of the contaminated soil ranged from 7.48 to 8.56, meaning that the soil was alkaline.
Figure 1. The tailings sample location (Rachman et al., 2017).

Table 1. Distribution of soil particles and soil classes in different depths.

| Sample code | Latitude/longitude | Depth (cm) | Proportion of soil fraction (%) | Texture class |
|-------------|--------------------|-----------|---------------------------------|---------------|
|             |                    |           | Sand | Silt | Clay |               |
| C.1         | 110°3’36,704” E   | 30        | 40.93 | 34.46 | 24.61 | Loam          |
|             | 7°49’36,672” S    | 60        | 31.35 | 44.29 | 24.18 | Loam          |
|             |                    | 90        | 21.35 | 47.95 | 30.70 | Clay loam     |
| T.1         | 110°3’44,474” E   | 30        | 76.23 | 8.49  | 15.28 | Loamy sand    |
|             | 7°50’1,820” S     | 60        | 71.37 | 10.23 | 18.40 | Loamy sand    |
|             |                    | 90        | 32.33 | 48.37 | 19.29 | Loam          |
| T.2         | 110°3’45,615” E   | 30        | 53.73 | 28.46 | 17.80 | Loamy sand    |
|             | 7°50’1,628” S     | 60        | 43.92 | 45.74 | 10.34 | Loam          |
|             |                    | 90        | 35.40 | 44.87 | 19.73 | Loam          |
| T.3         | 110°3’56,786” E   | 30        | 32.33 | 48.37 | 19.29 | Loam          |
|             | 7°50’16,648” S    | 60        | 25.53 | 47.29 | 27.18 | Loam          |
|             |                    | 90        | 13.93 | 52.86 | 33.21 | Silty clay loam |
| T.4         | 110°3’57,644” E   | 30        | 57.04 | 30.69 | 12.27 | Loamy sand    |
|             | 7°50’15,453” S    | 60        | 36.24 | 50.72 | 13.04 | Silty Loam    |
|             |                    | 90        | 28.44 | 25.57 | 45.98 | Clay          |
| T.5         | 110°3’49,860” E   | 30        | 71.41 | 10.65 | 17.94 | Loamy sand    |
|             | 7°50’31,336” S    | 60        | 55.99 | 31.35 | 12.66 | Loamy sand    |
|             |                    | 90        | 18.35 | 59.95 | 21.70 | Silty Loam    |
Table 2. Soil pH.

| Sample code | Soil pH |
|-------------|---------|
| C1          | 6.74    |
| T.1         | 8.56    |
| T.2         | 7.76    |
| T.3         | 7.88    |
| T.4         | 8.18    |
| T.5         | 7.48    |

Based on the data in Table 2, it can be seen that the pH value of the tailings at the research sites is generally quite alkaline, i.e. 7.48 - 8.56. High pH values will result in low solubility of heavy metals due to the presence of similar ions that affect soil contamination (Fijałkowski et al., 2012). According to Draszawka (2017), soil pH is an important parameter, directly affecting the absorption/release, precipitation, complex forms, and oxidation-reduction reactions.

**Total mercury concentration in soil**

The result showed (Table 3) the mercury concentrations in the soil sample are mostly higher than those in the soil for control. The amount of mercury in the soil at the site of traditional gold mining ranged from 0.30 to 22.51 mg/kg. Based on Table 3, it can be seen that the concentration of mercury in the control soil at locations and depths are low. The concentration of mercury contained in the control location that occurred by the hydrologic cycle is also influenced by wind speed and direction. The hydrological cycle contributes to the evaporation, condensation, infiltration process supported by wind speed and direction of the natural deposition process. The mercury concentration in the control location is still below the quality standards set by Indonesian government regulations Number 101 the year 2014.

Table 3. Analysis results total mercury concentration in soil.

| Sample code | Depth (cm) | Mercury concentration (mg/kg) | Quality Standard* (mg/kg) |
|-------------|------------|-------------------------------|--------------------------|
| C1          | 30         | 0.19                          | 0.3                      |
|             | 60         | 0.02                          |                          |
|             | 90         | not detected                  |                          |
| T.1         | 30         | 5.34                          |                          |
|             | 60         | 22.51                         |                          |
|             | 90         | 3.22                          |                          |
| T.2         | 30         | 0.42                          |                          |
|             | 60         | 1.01                          |                          |
|             | 90         | 0.78                          |                          |
| T.3         | 30         | 0.75                          |                          |
|             | 60         | 1.06                          |                          |
|             | 90         | 0.88                          |                          |
| T.4         | 30         | 4.81                          |                          |
|             | 60         | 3.49                          |                          |
|             | 90         | 0.89                          |                          |
| T.5         | 30         | 0.74                          |                          |
|             | 60         | 0.30                          |                          |
|             | 90         | 4.22                          |                          |

*Decree of the Government of the Republic of Indonesia No. 101/2014, concerning Hazardous Waste Management

According to Siregar (2006), at a given location, concentration is influenced by environmental factors, including distance from the sources of pollution, topography, altitude, air pollutants, rainfall, solar radiation, and wind speed and direction. Based on the analysis in Table 3, the concentration of mercury in soil was much higher than the threshold of mercury-contaminated soil quality standards of the Government Regulation of Indonesia No 101 of 2014. The amount of mercury in the soil at a depth of 30 cm, the amount of mercury was 0.19 mg/kg. This is because during the combustion process of gold with mercury, which took place in the open area, it led to emissions of mercury vapour carried by the wind, causing the mercury vapour to be discharged into the ground (Dranga et al., 2012). At a depth of 90 cm, the amount of mercury was low by <0.0001 mg/kg. This condition was caused by the volatile nature of mercury when reacting with the surrounding environment (Risher, 2003). The smaller amount of mercury was related to the distance from the ground being farther from polluted sources.
location and depth. In the first sample location, very high amounts of mercury occur at a depth of 60 cm which amounts to 22.51 mg/kg because the soil contains sandy loam that has a fast conductivity of water affecting the seepage speed into the soil (Oyeogbe and Oluwasemire, 2013), whereas the lowest amount of mercury at a depth of 90 cm is equal to 3.22 mg/kg because the soil contains silt clay whose nature has a very fine grain and also low permeability rendering it impenetrable to heavy metals (Rachman et al., 2018).

At the second sample location, the amount of mercury tends to a lower value than the first sampling location. This is due to the condition of the soil texture that is dominated by clay content so which is impenetrable to heavy metals through the pores of the soil (Rani et al., 2012). In the third sample location, the amount of mercury there was a very small difference between the depths of 30 cm, 60 cm, and 90 cm. This condition is caused by the nature of mercury which is volatile when it is on the ground, as a result of the reaction between mercury and the outside environment, and is influenced by the texture of the local soil-containing clay. The small pores in the soil affect the low uptake of heavy metals in the soil.

At samples of location 4, the amount of mercury was found extremely high at a depth of 30 cm which was equal to 4.81 mg/kg and the lowest (0.89 mg/kg) was at a depth of 90 cm. The high amount of mercury at a depth of 30 cm resulted from the heavy use of mercury in the processing of gold. The low amount of mercury at a depth of 90 cm was alleged because of the type of clay soil with low permeability. Besides, it was affected by the soil texture classified as clay tending to be sticky when wet with a strong fusion between adjacent soil grains contributing to the low uptake of heavy metals into the soil.

At location 5 relatively high amounts of mercury were present at a depth of 90 cm, 4.22 mg/kg. This condition is caused by the excessive use of mercury in processing gold. In addition, the soil depth of 30 cm and 60 cm is dominated by loamy sand, which can drain water containing heavy metals such as mercury, which is easy to carry because it has a poor soil permeability value. Water containing heavy metals is retained at a depth of 90 cm because at that depth it is dominated by silty loams which have better permeability values. (Haddad et al., 2019).

**Conclusion**

Based on the test results, the amount of mercury in the soil ranged from 0.30 to 22.51 mg/kg, so it can be concluded they exceeded the quality standard of mercury-contaminated soil according to Government Regulation No. 101 which is 0.3 mg/kg. However, a more detailed investigation of Hg speciation in the contaminated soils is required further in any case, and that changes in soil climatic, physical, biological, atmospheric, and chemical properties should be considered. These factors may lead to the variability of total Hg concentrations in the short and long term.

**Acknowledgement**

The authors gratefully acknowledge the Education Fund Management Institute of the Republic of Indonesia for providing the research grant.

**References**

Aksakal, E.L., Angin, I. and Oztas, T. 2012. Effects of diatomite on soil physical properties. *Catena* 88(1): 1–5.

Appel, P.W.U. and Na Oy, L.D. 2014. Mercury-free gold extraction using borax for small scale gold miner. *Journal of Environmental Protection* 5: 493-499.

ATSDR (Agency for Toxic Substances and Disease Registry), (2013). Substance Priority List.

Bansah, K.J., Yalley, A.B. and Duper, N.D. 2016. The hazardous nature of small-scale underground mining in Ghana. *Journal of Sustainable Mining* 15: 8-25.

Dastoor, A.P. and Laroque, Y. 2004. The global circulation of atmospheric mercury: A modeling study. *Atmospheric Environment* 38: 147–161.

Djimaluddin, H., Thamrin, M. and Achmad, A. 2012. Potential and prospects added value metals in Indonesia; a study on mineral conservation efforts. *Proceedings of the Research Faculty of Engineering, Makassar* 6: 2-14.

Drace, K., Kiefer, A.M., Veiga, M.M., Williams, M.K., Ascar, B., Knapper, K.A., Logan, K.M., Breslin, V.M., Skidmore, A., Bolt, D.A., Geist, G., Reidy, L. and Czrdziel, J.V. 2012. Mercury-free, small-scale artisanal gold mining in Mozambique: utilization of magnets to isolate gold at clean tech mine. *Journal of Cleaner Production* 32: 88-95.

Dranga, B.A., Lazar, L. and Koeser, H. 2012. Oxidation catalysts for elemental mercury in flue gases: a review. *Catalysts* 2(1): 139-170.

Draszawka-Bolzan, B. 2017. Effect of pH and soil environment. *World News of Natural Sciences* 8: 600-60.

Fijałkowski, K., Kacprzak, M., Grobelak, A. and Placek, A. 2012. The influence of selected soil parameters on the mobility of heavy metals in soils. *Inżynieria i Ochrona Środowiska* 15: 81-92.

Gunnawan, A., Setiawan, L., Subarna, A.W. and Suhendi, E. 2001. Pilot of Gold Mining in Kokap, Kulon Progo, Yogyakarta. Research and Development Center for Mineral and Coal Technology (Tekmira), Bandung (in Indonesian).

Haddad, S.A., Lemanowicz, J. and Abd El-Azeim, M.M. 2019. Cellulose decomposition in clay and sandy soils contaminated with heavy metals. *International Journal of Environmental Science and Technology* 16(7): 3275-3290.

Hasriyanto, A.A., Saleh, S., Basir-Cyio, M. and Yunus, M. 2018. Effect of mercury contamination on the diversity of soil Arthropods in Poboya gold mining. *Journal of Environmental Science and Technology* 11(5): 254-261.

Hu, B., Zhou, J., Liu, L., Meng, W. and Wang, Z. 2017. Assessment of heavy metal pollution and potential ecological risk in soils of Tianjin sewage irrigation region, North China. *Journal of Environmental & Analytical Toxicology* 7: 425-525.

Indonesian Government Regulations Number 101 the year 2014. Waste Management of Hazardous and Toxic
Materials. The Government of the Republic of Indonesia.
Kpan, J.D.A., Opoku, B.K. and Gloria, A. 2014. Heavy metal pollution in soil and water in some selected towns in Dunkwa-on-Offin District in the central region of Ghana as a result of small scale gold mining. Journal of Agricultural Chemistry and Environment 3: 40-47.
Mahbub, K.R., Kriahman, K., Naidu, R. and Megharaj, M. 2017. Mercury toxicity to Eisenia fetida in three different soils, Environmental Science and Pollution Research 24(2):1261-1269.
Orhue, E.I. and Frank, U.O. 2011. The fate of some heavy metals in soils: a review. Journal of Applied and Natural Science 3: 131-138.
Rachman, R.M., Bahri, A.S. and Trihadiningrum, Y. 2018. Mercuric contamination in artisanal gold mining area of Manyera River, Niger State Nigeria. E3 Journal of Environmental Research and Management 4: 0326-0333.
Schimdt, C.W. 2012. Quicksilver and gold: mercury pollution from artisanal and small-scale gold mining. Environmental Health Perspectives 120(11): 425-429.
Setiabadi, B. 2005. Spreading Mercury as a result of the Gold Mining Business in Regional Sangon, Kulon Progo, Special Region Yogyakarta. Colloquium of Field Results, Ministry of Energy and Mineral Resources of Indonesia, DIM:1-17 (in Indonesian).
Siregar, E.B.M. 2006. Air Pollution, Plant Response, and Effect on Humans. USU Repository, Medan (in Indonesian).
UNEP. 2008. The Global Atmospheric Mercury Assessment: Sources, Emission and Transport. Geneva: UNEP-Chemicals.
United States Department of Agriculture (USDA). 1987. Textural Soil Classification System, 1987.
USEPA. 2007. Treatment Technologies for Mercury in Soil, Waste, and Water. Washington D.C., USA.
Veiga, M.M., Nunes, D., Klein, B., Shandoro, J.A., Velasquez, P.C. and Sousa, R.N. 2009. Mill leaching: a viable substitute for mercury amalgamation in the artisanal gold mining sector. Journal of Cleaner Production 17: 1373-1381.
Vidnairizki, N.P. 2014. Phytoaccumulation of Mercury on Cyclosorus interruptus H. Ito., Desmodium gangeticum DC., and Photomorphone subpeltata Mig., in Traditional Gold Mining Sangon village, Kulon Progo. Thesis, Faculty of Biology, Gajah Mada University (in Indonesian).
Wahab, S.A.A. and Marikar, F.A. 2012. The environmental impact of gold mines: pollution by heavy metals. Central European Journal of Engineering 2: 304-313.
Widodo. 2008. Pollution of mercury (Hg) as the impact of the gold ore processing on Cilunggunung river, Waluran, Sukabumi Regency. Jurnal Geologi Indonesia 12(11): 139-149 (in Indonesian).
Wiriosudarmo, R. 1999. Economic Empowerment through Small-Scale Mining. Ecomine NL Foundation, Policy, and Management Seminar on Small-Scale Mining, Jakarta (in Indonesian).
Withdrawn. 2016. ASTM D422-63(2007)e2, Standard Test Method for Particle-Size Analysis of Soils, ASTM International, West Conshohocken, PA, 2007, www.astm.org.