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Study of mercury concentration in plants in Traditional Buladu Gold Mining

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Abstract. The objective of the study was to examine the levels of mercury concentrations in plants due to the traditional gold mining of Buladu, Sumalata District, Gorontalo Regency. Buladu gold mining was located in Sumalata District, North Gorontalo Regency, Gorontalo Province. Plant samples were taken in 3 locations, namely plants living along the Hulawa River, living near to the tailings and those in residential areas. The number of plants that became the samples along the Hulawa River were 13 samples, 5 samples in the tailings, and 16 samples in residential areas. The total number of plants were as many as 34 samples. The samplings were taken from the roots and leaves so that there were a total of 68 samples. Plant analysis was carried out using mercury analyzer at UGM Integrated Research and Testing Laboratory. The quality standard used as a reference for plants employed the Decree of the Director General of National Agency of Drug and Food Control No: 03725 / B / SK / VII / 89, where mercury levels could not exceed 0.5 mg / kg. Data analysis used tables and graphs and was subsequently interpreted. The results showed that the concentration of mercury in the leaves of plants living along the Hulawa River ranged from 0.00142 mg / kg to 0.41617 mg / l and that in the root ranged from <0.00014 - 14.890 mg / kg. Mercury concentrations in plants living around the tailings in leaves ranged between <0.00014 - 1.30822 and at roots ranging from 0.01058 - 12.59366. In residential areas, the concentration of mercury in leaves ranged from <0.00014-1.44368 mg / kg and in the root ranged from <0.00014-1.94505. Based on these results, the concentration of mercury in plants along the river, around the tailings and in the people's resident area tended to be above the quality standards set by the Decree of the Director General of National Agency of Drug and Food Control No: 03725 / B / SK / VII / 89.

1. Introduction

Buladu traditional gold mining is located in Sumalata District, North Gorontalo Regency. Gold exploration activities in Buladu area by the Dutch Government which began in the Dutch Colonialization era (18th century). There is historical evidence in this area including 3 Dutch graves on Buladu Beach died in 1899, mine pits with rails and lorries, gold ore processing equipment in the form of large sized pots, and solid tailings found around the mine site. Around the 1970s, the exploitation activity was continued with the traditional mining model. The location of the mining was reopened by the local community. At that time gold search activities were carried out traditionally by drilling sand and rock deposits along the Buladu River [1].



The impact of the mining activities can be positive for the mining business area. However, mining activities can be negative towards the local ecosystem. The emergence of positive and negative impacts from mining business occurs during the exploration and exploitation phase including the processing and sale of mining products and post-mining. Mining business known as small-scale gold mining is considered to be the cause of environmental damage. Ore processing is carried out with an amalgamation process in which mercury (Hg) is used as a binding medium for the gold [2].

In this area, the livelihoods of many people depend on mining. The results showed that 70% of people living in locations around mining were living as miners [3]. The people who lived as miners show that 75% graduated from elementary school, 5% from junior high school and 20% from high school. The low level of education caused people in the Buladu gold mine did not manage waste residues properly. A low level of education also triggers people to depend on their life in traditional mining. A low level of education causes them not to be aware of the dangers of mercury poisoning for their bodies [3]. This triggers the public to throw away the waste carelessly. Communities in the mining location are not aware of the dangers of wastewater being discharged into the environment which will be absorbed into the soil and eventually be absorbed by plant roots.

The first plant's main structure is the root, known by its scientific name radix. Roots have less important role than leaves and stems. The function of the root is to absorb water and nutrients, which will then be transmitted to the stem and leaves, resulting in a metabolic process. The nature of the roots is generally contrary to the nature of the stem, among others, is to grow away from the center of the earth, which is known as the term positive geostrophic or go to a water source and grow away from the light [4]. The nature of the roots goes to the water source, causing the roots to absorb liquid waste that is absorbed into the soil. This will cause the roots to accumulate heavy metals from mining waste. The process of gold mining at Buladu gold mining used mercury in the processing steps. The remaining mercury waste was wasted into the environment, especially the soil. It entered to the soil and could eventually be absorbed by plants. This would be dangerous because if the plants accumulated mercury, it could enter humans through food chain relationships. The purpose of this study was to identify mercury concentrations in various plants that lived in the traditional mining area of Buladu, Sumalata District, North Gorontalo Regency.

2. Research Methods

The location of the study was in Buladu gold mining in Sumalata District, North Gorontalo District, Gorontalo Province. Plant samples were taken in 3 locations, namely plants that lived along the Hulawa River, lived close to tailings and lived in residential areas. Sampling location was shown in Figure 1.

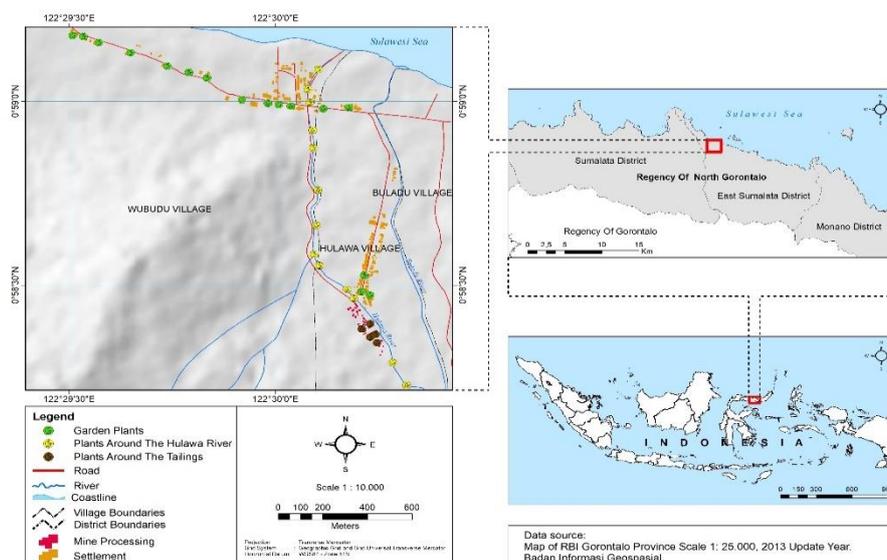


Figure 1. Map of Sampling Locations

The number of sample plants that lived along the Hulawa River were 13 samples, 5 samples in tailings and in 16 samples in residential areas. The total number of plants were as many as 34 samples. Samplings were carried out on the roots and leaves so that it obtained a total of 68 samples. Plant analysis was carried out using mercury analyzer at UGM Integrated Research and Testing Laboratory. The quality standard used as a reference for plants employed the Decree of the Director General of National Agency of Drug and Food Control No: 03725 / B / SK / VII / 89, in which mercury levels cannot exceed 0.5 mg / kg. Data analysis applied tables and graphs and was subsequently analyzed.

3. Result and Discussion

3.1. Mercury concentration in plants along the Hulawa River

3.1.1. *Mercury Concentration at Plant Roots.* The analysis results of the highest mercury concentration in the roots of plants that lived along the river obtained a value of 14.890 mg / kg, on needle grass plants (*andpagon aciculatus*). The lowest value showed <0.00014 mg / kg in taro plants (*Colocasia esculenta*). The average value of mercury concentration in plant roots along the Hulawa River was 2.387 mg / kg. This result was above the quality standard set by the Decree of the Director General of National Agency of Drug and Food Control No: 03725 / B / SK / VII / 89 that should not be exceeding 0.5 mg / kg. Some types of plants such as *suplir* (*adiantum cuneatum*), grass plants (*andpagon aciculatus*) and taro (*Colocasia esculenta L*) had concentrations above the established quality standards. The analysis results of the mercury concentrations in plant roots around the river were shown in Table 1.

Table 1. Concentration of Mercury in the Roots of Plants Living around the Hulawa River

No	Names	Result mg/kg	Quality Standard mg/kg
1	Suplir (<i>Adiantum cuneatum</i>)	0.88752	0.5
2	Taro (<i>Colocasia esculenta</i>)	1.37442	0.5
3	Grass (<i>Andpagon aciculatus</i>)	14.890	0.5
4	Needle grass (<i>Andpagon aciculatus</i>)	5.11985	0.5
5	Tapak Dara (<i>Catharanthus roseus L.G.Don</i>)	0.06701	0.5
6	Suplir (<i>Adiantum cuneatum</i>)	1.34	0.5
7	Taro (<i>Colocasia esculenta L</i>)	3.88	0.5
8	Suplir (<i>Adiantum cuneatum</i>)	1.69	0.5
9	Suplir (<i>Pteridophyta</i>)/ <i>filiciinae</i>)	0.65601	0.5
10	Taro (<i>colocasia esculenta(L) Schott</i>)	0.82258	0.5
11	Tapak Dara (<i>Catharanthus roseus L.G.Don</i>).	0.26817	0.5
12	Taro (<i>Colocasia Esculenta</i>)	<0.00014	0.5
13	Water hyacinth (<i>Eichhornia crassipes</i>)	0.03647	0.5

Source: Primary data (2016)

Kosegeran, [5] conducted research on plants that lived in Talawaan Gold Mining. Samples were taken in 3 locations as many as 9 samples of ferns and 3 samples of the soil. The results showed that ferns did not contain mercury, while the soil from 3 locations contained 0.6 ppm mercury. This result was lower compared to research conducted at Buladu gold mining. Plants in the Buladu gold mining showed that the *suplir* type (*adiantum cuneatum*) had mercury concentrations of 0.88752 - 1.69 mg / kg, grasses (*andpagon aciculatus*) ranging from 5.11985-14.890 mg / kg, and taro (*colocasia esculenta*) ranging from 0.00014-3.88 mg / kg. This result was already very high because it was already above the established quality standard. The increase of mercury concentrations in plants was caused by high concentrations of mercury in sediments. The concentration of mercury in the sediments along the Hulawa River at 3 sampling ranged between 10.8731-55.0680 mg / kg; 0.08995 - 136.70 mg / kg and 0.55255 - 244.16 mg / kg [3]. There was a relationship between elevation of mercury in plants and high concentrations of mercury in sediments. The higher the concentration of mercury in the growing medium, the higher it will be in plants. This result was same as the research conducted by Lona, et al [6]. Research conducted by Lona et al (6) showed that mercury absorbed by roots ranged from 0.112 to

0.2997 mg / g and leaves ranged from 0.0221 to 0.0287 mg / g. The higher the addition of metals to the growing medium, the higher the concentration of mercury accumulated by the organ of the *seruni* plant. Plants can absorb pollutants from a medium so that they accumulate around the roots of plants. Plant roots will absorb pollutants together with the absorption of nutrients and minerals in the media and then be translocated to the leaves [7,8].

Based on these results, mercury pollution in plants at the Buladu gold mining site was already very high and endangering the community because mercury could enter to human body through food chain. Plants get nutrients for the growth needs from the surrounding environment by absorption of the roots. The process of absorbing nutrients and minerals into plants through an active transport mechanism occurs between cells, where nutrients are absorbed in the form of elements or compounds.

The results of research conducted on plants showed that the highest accumulation of pollutants was in locations 0-500 m from pollutant sources [9]. The amount of heavy metal taken by plants from the soil is determined by the availability of pollutant materials. The more pollutant content in the soil, the more easily it is absorbed by the roots. The results of the analysis of mercury concentration in the leaves of plants that live around the river are shown in Table 2.

Table 2. Analysis Results of Mercury Concentration on Leaves around the Hulawa River

No	Plant's Name	Results mg/kg	Quality Standard mg/kg
1	Fern plants (<i>Pterydophyta</i>) / <i>filiciinae</i>)	0.03550	0.5
2	Taro (<i>colocasia esculenta</i> (L) Schott)	0.09744	0.5
3	Needle grass (<i>Andropagon aciculatus</i>)	0.20938	0.5
4	Needle grass (<i>Andpagon aciculatus</i>)	0.41617	0.5
5	Tapak Dara (<i>Catharanthus roseus</i> L.G.Don)	0.05923	0.5
6	Fern plants (<i>Pterydophyta</i>)/ <i>filiciinae</i>)	0.02168	0.5
7	Taro (<i>Colocasia Esculenta</i>)	0.12209	0.5
8	Suplir (<i>Pterydophyta</i>)/ <i>filiciinae</i>)	0.06983	0.5
9	Suplir (<i>Pterydophyta</i>)/ <i>filiciinae</i>)	0.01194	0.5
10	Taro (<i>colocasia esculenta</i> (L) Schott)	0.431	0.5
11	Tapak dara (<i>Catharanthus roseus</i> L.G.Don)	0.02917	0.5
12	Taro (<i>Colocasia Esculenta</i>)	0.06377	0.5
13	Water hyacinth (<i>Eichhornia crassipes</i>)	0.00142	0.5

Source: Primary data (2016)

The analysis results showed that mercury concentrations were higher in roots than in leaves. The concentration of mercury was very high in plants due to the residual processing of gold being disposed of in the treatment tank. If it is full, it will overflow in the surrounding soil and eventually flow and enter the river. It caused the plants around to be contaminated with mercury. The tendency of mercury accumulation in the roots was higher than the leaves because the root was a part that functions to absorb nutrients from functioning plants and organs that had a direct contact with the planting medium. The absorption results were obtained in the form of elements of mercury to be translocated to other parts which are leaves.

3.1.2. Analysis results of mercury concentration on plant roots around tailings. The analysis results of mercury concentration in plant roots that lived around the tailings showed that the highest concentration was 12.59366 mg / kg in green taro plants (*Colocasia Esculenta*) and the lowest was 0.01058 mg / kg in needle grass plants (*Andpagon aciculatus*). Based on these results, mercury concentration in plants had exceeded the limit set by the Director General of National Agency of Drug and Food Control No. 03725 / B / SK / VII / 89. The analysis results of mercury concentrations in plant roots that lived around tailings can be seen in Table 3.

Table 3. Analysis Results of Mercury Concentration in Roots of Plants that Lived Near Tailings

No	Names	Analysis Result mg/kg	Quality Standard mg/kg
1	<i>Suplir (Adiantum cuneatum)</i>	2.37107	0.5
2	Taro (<i>Colocasia Esculenta</i>)	12.59366	0.5
3	Needle grass (<i>Andpagon aciculatus</i>)	7.35	0.5
4	Soft <i>suplir (Pteridium aquallinum)</i>	1.73	0.5
5	Needle grass (<i>Andpagon aciculatus</i>)	0.01058	0.5

Source: Primary data (2016)

The high concentration of mercury in plants around tailings is related to the high concentration of mercury in sediments. Mercury concentrations in tailings sediments in sampling I, II and III ranged from 10.0643 - 36.4008 mg / kg, 3.31 - 135.55 mg / kg and 104.36 - 236 mg / kg [3]. One of the functions of the root is to absorb water and nutrients from the soil, so the concentration of mercury will be high in the roots.

Direct tailing disposal to the soil without any treatment caused the soil to be contaminated with mercury so that there was a possibility of mercury accumulation in the surrounding food plants. In addition, it could also cause the infiltration of Hg to ground water used by residents as a source of clean water. Mercury is a cumulative poison, in the sense that a small amount of mercury is absorbed in the body for a long time will cause danger. The dangers of diseases caused by mercury compounds include damage to hair and teeth, loss of memory and disruption of the system requirements [2].

3.1.3. Analysis results of mercury concentration on plant leaves around tailings. The analysis results of mercury concentration in the leaves of plants that lived around the tailings showed that the highest concentration was 1.30822 mg / kg on needle grass (*Andpagon aciculatus*) and the lowest was 0.00014 mg / kg in taro (*Colocasia Esculenta*). This result was above the quality standard set by the Decree of the Director General of National Agency of Drug and Food Control No: 03725 / B / SK / VII / 89. Based on the decree of the Director General of National Agency of Drug and Food Control No. 03725 / B / SK / VII / 89, the concentration of plants cannot exceed 0.5 mg / l. The analysis results of mercury concentration in leaves living around the tailings can be seen in Table 4.

Table 4. Analysis Results of Mercury Concentration on Leaves of Plants that Lived Near Tailings

No	Names	Analysis Result mg/kg	Quality Standard mg/kg
1	<i>Suplir ((Adiantum cuneatum)</i>	0.51563	0.5
2	Taro (<i>Colocasia Esculenta</i>)	<0.00014	0.5
3	Needle grass (<i>Andpagon aciculatus</i>)	1.30822	0.5
4	<i>Suplir (Pteridium aquallinum)</i>	0.17754	0.5
5	Needle grass (<i>Andpagon aciculatus</i>)	<0.00014	0.5

The analysis results of mercury concentrations in plant roots that lived around the tailings ranged from 0.01058-12.59366 mg / kg higher than the mercury concentration in the leaves of plants ranging from 0.00014-1.30822 mg / kg. Eckenfelder [10] stated that the ability of absorption and accumulation of heavy metals by plants is divided into 3 processes, namely the absorption of heavy metal precipitates by the roots, precipitation of mercury pollutants in the soil mobilized by plant roots by accumulation, and absorption by the root surface and deposited in the root zone. After from the root, mercury is translocated towards certain plant organs such as the stem and leaves and then locate metals in certain tissues to keep them from inhibiting the plant metabolism [11].

3.1.4. Analysis results of mercury concentration on plant roots around settlement area. The analysis results showed that the highest mercury concentration was in the roots of *pandanus* plants with 1.94505 mg / kg (*Pandanus Amarylifolius*). The lowest concentration was in mayyana medicinal plants (*Coleus Atropurpureus benth*) with <0.00014 mg / kg. Several types of garden plants consumed by the

community had been above the quality standard, namely lemongrass (*Cymbopogon citratus*), galangal (*Alpinia galanga*), water spinach (*Ipomea Reptana Poir*), Pandan (*Pandanus Amarylifolius*), and curcuma (*Curcuma Zanthorrhiza*). It will endanger humans who consumed it. The analysis results of mercury concentrations in the roots that lived in the resident yard can be seen in Table 5.

Table 5. Analysis Results of Mercury Concentration on Plant Roots in the resident yard

No	Names	Analysis Result mg/kg	Quality Standard Mg/kg
1	Pandan (<i>Pandanus Amarylifolius</i>)	1.94505	0.5
2	Lemongrass ((<i>Cymbopogon citratus</i>)	1.21653	0.5
3	Galangal (<i>Alpinia galanga</i>)	1.09105	0.5
4	Water spinach (<i>Ipomea Reptana Poir</i>)	1.18	0.5
5	Kumis Kucing (<i>Orthosiphon Aristatus</i>)	0.44447	0.5
6	Maiyana (<i>Coleus Atropurpureus benth</i>)	0.04787	0.5
7	Pandan (<i>Pandanus Amarylifolius</i>)	0.73919	0.5
8	Kencur (<i>Kaemferia galaga L</i>)	0.08194	0.5
9	Lemongrass (<i>Cymbopogon citratus</i>)	0.03934	0.5
10	Galangal (<i>Alpinia galangal</i>)	0.064	0.5
11	Water spinach (<i>Ipomea Reptana Poir</i>)	0.26518	0.5
12	Curcuma (<i>Curcuma Zanthorrhiza</i>)	1.44368	0.5
13	Rice (<i>Oryza sativa</i>)	0.01073	0.5
14	Water spinach (<i>Ipomea Reptana Poir</i>)	0.05493	0.5
15	Mayyana (<i>Coleus Atropurpureus benth</i>)	<0.00014	0.5
16	Rice (<i>Oryza sativa</i>)	0.00950	0.5

Source: 2016 Primary Data

The amount of mercury concentration in plants is directly proportional to the amount of mercury in the growing media; in this case it is the resident area. If the soil is slightly contaminated with mercury, there will be little mercury accumulated in the plant. Research conducted by Khairuddin, [12] showed that the concentration and weight of metal mercury absorbed in the roots of spinach plants is directly proportional to the medium of planting. The results of a study conducted by Santoso et al [13] in Poboya gold mining showed that the concentration of mercury in crops such as peanuts ranged from 0.098-0.45 ppm, corn 0.07-0.43 ppm, rice plants 0.03-0.21 ppm and shallots ranged 0.01 - 0.32 ppm.

This result is lower than the concentration of mercury in *pandanus* plants (*Pandanus Amarylifolius*) with 0.73919 - 1.94505 mg / kg, lemongrass (*Cymbopogon citratus*) 0.03934-1.21653 mg / kg, galangal (*Alpinia galangal*) 0.064-1.09105, water spinach (*Ipomea Reptana Poir*) 0.05493 - 1.18, rice (*Oryza sativa*) with 0.00950 - 0.01073 mg / kg, curcuma (*Curcuma Zanthorrhiza*) 1.44368 mg / kg, mayyana 0.00014 - 0.04787 mg / kg, kumis kucing (*Orthosiphon Aristatus*) 0.44447 mg / kg and kencur (*Kaemferia galaga L*) 0.08194 mg / kg living in the resident area around Buladu mining location. These plants were planted by residents to be used daily. The high accumulation of mercury concentrations in plant roots consumed by people around Buladu mining location was very dangerous because it will endanger human health.

Soil pollution by mercury (Hg) is often related to the addition of heavy metals from various factors such as fertilizer, lime, mud and manure. The dynamics between the amount of Hg taken from the soil and plants is not linear and depends on several variables such as soil sediment content, carbon exchange capacity, content of oxides and carbonates, redox potential, formulations used and total metal content. In general, mercury uptake in plants is related to pollution levels. If the pollution level is low, the amount of mercury is below the allowable level [14]. Many plants absorb Hg which tends to be accumulated in the roots [15], and some can even accumulate in moderate amounts in the shoots [16]. Research by Suszycynsky and Shann [17] showed that plant exposure of Hg^o can be taken and accumulated by the shoots but not transferred to the roots [18].

3.1.5. Analysis Results of Mercury Concentration on Plant Leaves around Settlement Area. The analysis results of mercury concentrations in the leaves of plants that lived in the residential areas showed that the highest concentration was 1.44368 mg / kg and the lowest was 0.00014 mg / kg. The average rate of mercury concentration was 0.27107 mg / kg. Some plants that had been above the quality standard of the Director General of National Agency of Drug and Food Control No. 03725 / B / SK / VII / 89 are *Pandan (Pandanus Amaryllifolius)*, galangal (*Alpinia galanga*), water spinach (*Ipomea Reptana Poir*) and curcuma (*Cymbopogon citratus*). Research conducted by Ordak, et al [19] showed that herbal plants had higher mercury concentrations in spring than in autumn. Perennial plants have high levels of mercury compared to seasonal plants (monocarpic). Therefore, usually the use of herbal plants had significantly higher levels of mercury compared to others.

Based on these results, cultivated plants consumed by people living around the mining location had been contaminated with mercury. This would endanger humans who consume these plants as food. The toxicity of mercury in plants can affect the antioxidant system, affect photosynthesis, inhibit the growth and production of plants and also affect nutrient uptake [20]. The results of the analysis of mercury concentration in the roots of plants that lived around residential areas are shown in Table 6.

Table 6. Analysis Results of Mercury Concentration on Plant Leaves in Residential Areas

No	Names	Analysis Result mg/kg	Quality Standard Mg/kg
1	<i>Pandan (Pandanus Amaryllifolius)</i>	0.52674	0.5
2	Lemongrass (<i>Cymbopogon citratus</i>)	0.08691	0.5
3	Galangal (<i>Alpinia galanga</i>)	0.95055	0.5
4	Water spinach (<i>Ipomea Reptana Poir</i>)	0.52858	0.5
5	<i>Kumis Kucing (Orthosiphon Aristatus)</i>	0.04159	0.5
6	<i>Maiyana (Coleus Atropurpureus benth)</i>	0.00753	0.5
7	<i>Pandan (Pandanus Amaryllifolius)</i>	<0.00014	0.5
8	<i>Kencur (Kaemferia galaga L)</i>	0.08194	0.5
9	Lemongrass ((<i>Cymbopogon citratus</i>)	0.03934	0.5
10	Galangal (<i>Alpinia galanga</i>)	0.064	0.5
11	Water spinach (<i>Ipomea Reptana Poir</i>)	0.26518	0.5
12	Curcuma/ Lemongrass (<i>Cymbopogon citratus</i>)	1.44368	0.5
13	Rice (<i>Oryza sativa</i>)	0.13247	0.5
14	Water spinach (<i>Ipomea Reptana Poir</i>)	0.09708	0.5
15	<i>Mayyana (Coleus Atropurpureus benth)</i>	0.06938	0.5
16	Rice (<i>Oryza sativa</i>)	<0.0014	0.5

Source: 2016 Primary Data

In absorbing heavy metals, plants form reductase enzyme in the root membrane which functions to reduce metal. Heavy metal cycles from roots show that mercury must be transported through transporting tissues, namely xylem and phloem, to all parts of the plant to improve the efficiency of transporting metals in the chelate molecule (binding molecule). In further, mercury is accumulated in all parts of the plant in the roots, stems and leaves [21,22]. Mercury contaminants are very dangerous for health risks for those who consume foods that contain mercury. The entry of mercury in the population, such as vegetables and grains that have exceeded the permitted limit can cause serious risks [23,24].

4. Conclusion

The results of the study showed that the concentration of mercury in the leaves of plants that lived along the Hulawa River ranged from 0.00142 mg / kg to 0.41617 mg / l and at the root ranged from <0.00014 - 14.890 mg / kg. Mercury concentrations in plants that lived around the tailings in leaves ranged from <0.00014 - 1.30822 and at the roots ranged from 0.01058 - 12.59366. In the residential area, the concentration of mercury in the leaves ranged from <0.00014-1.44368 mg / kg and at the root ranged

from <0.00014 - 1.94505 . Based on these results, the concentration of mercury in plants both along the river, around the tailings and in resident areas tended to be above the quality standards set by the Decree of the Director General of National Agency of Drug and Food Control No: 03725 / B / SK / VII / 89.

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