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Research Article

Mercury contamination in soil, tailing and plants on agricultural fields near closed gold mine in Buru Island, Maluku

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Abstract: Agricultural productivity in Buru Island, Maluku is threatened by tailings which are generated from formerly gold mine in Botak Mountain in Wamsait Village. Gold that extracted by using mercury was carried out in mining area as well agricultural field. High content of mercury in tailings and agricultural field pose a serious problem of food production and quality; and further endangers human health. The purpose of this research was to determine the contaminant level of mercury in tailing, soil and its accumulation in edible part of some food crops. Soil, tailing and plant samples for Hg testing were taken by purposive method based on mining activities in Waelata, Waeapo and Namlea sub district. Six soil samples had been analyzed for their chemical properties. Total mercury levels in tailings and plants were measured by Atomic Adsorption Spectrophotometer. This study showed that agricultural field where tailings were deposited contained Hg above the threshold but agricultural area which is far from hot spot did not. Most edible parts of food crops accumulated mercury more than Indonesian threshold for mercury content in food. This evidence explained that tailings deposited on the surface of agricultural field had an impact on soil quality and crop quality. Tailing accumulated on soil will decreased soil quality since naturally soil fertility in agricultural field in Buru is low.

Keywords: *gold mine, mercury, paddy field, soil quality.*

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Introduction

Buru Regency in Maluku Province which cover an area of 5,577.48 km² is the center of food crop production especially rice in Eastern part of Indonesia. During 2011-2015, massive illegal gold mining occurred in Botak Mountain around Wamsait Village where Wamsait River and Anhoni River flow. The gold deposit at the site was the primary ore-type vein ore, which was mined by underground technique of gophering method.

The remaining tailings must be managed properly to reduce the amount of moisture and avoid heavy metal discharge to stream and atmosphere before they are placed back on the underground or used for another purpose. Unfortunately local miner communities had neither post mining management plan nor tailings management facility. In Botak Mountain, tailing from the upper zone of mountain flowed into the Anhoni River to reach the lower zone of area; significant amount of tailing in lower zone was stored on nearby area and agricultural filed without any treatment. Tailing accumulated on the surface of sago plantation; former important staple food in Maluku. Mining material was also sold to local farmers or bigger miner and was extracted in agricultural area. In certain area, agricultural area in Waelata and Waeapo sub-districts are covered with tailings.

Efforts to increase the productivity of food crops now are threaten by gold-mine tailings. Illegal miner has extracted gold with amalgamation method by using mercury (Hg); one of the non-essential elements that has no biological and physiological function in normal cells (Gadd, 1992). Naturally, Hg is low abundance in the Earth's crust, The most important Hg mineral is cinnabar (HgS), which appeared on earth firstly more than three billion years ago (Hazen et al., 2012). Natural Hg content in soil is primarily determined by the composition of the parent material and the soil genesis (Bradl, 2005). Average Hg in world soils is only 0.15 mg/kg - 5 mg/kg. However, our preliminary study at 2015 showed that Hg content of tailings in the Anhoni river basin was only 0.64 mg/kg but those in tailings pile was 10.77 mg/kg. The soil covered by tailings contains Hg of 0.24 mg/kg to 20.36 mg/kg. Mercury-containing tailing which is left in the open environment will be washed; eroded and volatilized (Moreno et al., 2004), and biochemical transformations namely metallic and biological reduction (Morel et al., 1998) will take place. This poses ecological, food quality and health problem of toxic contamination.

Tailings of gold mines have poor physical and chemical properties. They are usually dominated by sand materials to silty materials with low organic matter and low cation exchange capacity (Taberima et al., 2010). The acidity of tailing is extreme; very acid or too alkaline. These properties are less supportive of plant growth and high Hg levels are potentially accumulating in edible part of food crop.

After mining activity is officially closed by Government of Maluku, assessment concerning Hg contaminant levels in the upper watersheds of Wamsiat and Anhoni river as well as surrounding agricultural areas has never been done. The possibility of Hg contamination in edible part of food crops grown in contaminated area has not been ascertained. This research was conducted to study the fertility of native soil, level of Hg in tailing and soil; and Hg accumulation in the edible part of food crops grown in tailing pile and tailing-contaminated soil in agricultural field of Buru District.

Materials and Methods

The research was conducted in Buru District of Maluku Province. Soil and plant analyzes were conducted at the Agricultural Environmental Research Institute in Pati which belong to Department of Agriculture. Samples were taken from several villages in three sub-districts (Figure 1) close to Botak Mountain:

1. Wamsait and Parbulu Village in Waelata sub-district.
2. Waikerta, Wanareja, Waitele Village, and Savana Jaya Village in Waeapo sub-district.
3. Siahoni Village in Namlea sub-district.

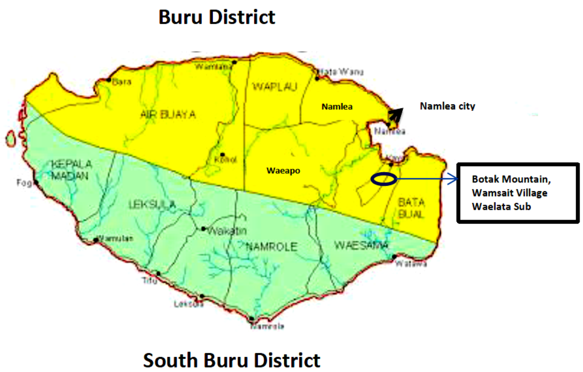

Figure 1. Waelata, Waeapo and Namlea sub-districts in Buru District map

Soil, tailing and plant sampling

Soil, tailing and plant samples for Hg testing were taken by purposive method based on mining activities in Waelata, Waeapo and Namlea sub districts. Soil samples for chemical analysis were taken by purposive method from six uncontaminated paddy field with wet land rice system in Waelata and Waeapo sub district. Six soil samples had been analyzed for their chemical properties (Table 1). Three soil samples were taken from each sampling point to depth of tillage of 30 cm using auger kit. The three soil samples ware mixed and placed in a labeled sample bag.

The soils were air dried for one week and passed through a 2-mm sieve prior to laboratory analyses to obtain soil fertility profile. The soil analyses were determined as follows: soil acidity, total C-organic, N-total, P and K total, available P, Cation exchange capacity, exchangeable cations, base saturation, and soil texture determination. The methods of soil testing for all soil parameters are presented in Table 1. Another soil and tailing samples were taken by using similar sampling method for mercury testing. Some soil samples were taken from uncontaminated site as a reference to compare the results. Tailing sample was collected from tailing pile near agricultural field. Edible part of food crops grown naturally on contaminated and uncontaminated soils was also collected. Soil and tailing were air dried and plant samples were dried at 70^{o}C for two consecutive days before mercury testing. Total mercury levels in tailings and plants were measured by Atomic Adsorption Spectrophotometer after sample extraction with 1 mL of perchloric acid p.a. and 5 mL of nitric acid p.a. (Sulaeman et al., 2005).

Table 1. Soil testing methods of certain chemical soil parameters

Soil Parameter	Testing Method	Unit
Soil acidity (pH_{H2O})	Potentiometric	
Soil acidity (pH_{KCl})	Potentiometric	
Organic Carbon	Walkley & Black	%
Total Nitrogen	Kjeldahl	%
C/N	-	-
Potential K	HCl 25%	mg/100g
Potential P_2O_5	HCl 25%	mg/100g
Available P_2O_5	Olsen	mg/kg P
Cation Exchange Capacity	Extract NH_4OAc pH 7	cmol/kg
Base saturation		%
Exchangeable cation: Na^2, Ca^2 , K^+, Mg^{2+}	Extract NH_4OAc pH 7	cmol/kg

Results and Discussion

Soil characteristics and fertility

Waeapo plain where recent paddy fields were constructed was formed about 50 years ago; the natural vegetation grown on this area was sago; traditional staple food. Actual rice yields in Waeapo plain was generally; less than those of paddy yield potential in most irrigated paddy production center in Indonesia, up to 8 t/ha. In Waeapo plain, rice productivity in the planting season of April-September 2014 was about 4.5 t/ha. Only a small amount of paddy field produce rice more around 6 t/ha. Chemical profile of six soil collected from six village was depicted in Table 1. All samples of paddy soil were slightly acid to neutral in acidity. Wetland rice systems have a positive effect on chemical fertility by bringing pH in the neutral range (Sahrawat, 2005). In neutral soil, plant nutrient is more available for roots uptake. A neutral soil acidity inhibit heavy metal movements in the soil so that potential poisoning from heavy metals can be suppressed (Bradl, 2005); heavy metal washing does not occur on neutral soil acidity.

Six sampling spots contained low organic Carbon and total Nitrogen; either their potential P or K of all soil sample was low (Table 2). Soil pudding creates organic matter accumulation due to reduced oxygen supply. In turn limited organic matter decomposition promotes formation of impermeable layer which reduces percolation losses. However, surface water and the first few millimeters of top soil remain relatively oxidized (Sahrawat, 2005). Irrigation system in Buru District is ceased in dry season so that farmers have no opportunity to grow rice twice consecutively. Since impermeable soil layer in Buru's paddy field has not been formed, in dry season submerged paddy soil dried up which in turn aerobic condition induce biological activity to decompose organic matter. Low soil organic matter explained the fact that organic matter application is very limited (Yadav et al., 2000) since the farmer in Buru mixed organic matter only recently.

All soils were low in cation exchange capacity (CEC) whereas their base saturation (BS) varied from very low to average (Table 2). Soil fertility status is determined by CEC, BS, potential P, potential K and organic C. Based on the data in Table 2, the soil fertility status of all soil samples were low (Table 3). Naturally, previous native plant in recent paddy field was sago (*Metroxylon sagu* Rottb; Arecacea) which was grown in wet land.

Table 2. Soil chemistry profile in six village in Buru District

Soil sampling location (village)	pH	Organic Carbon	Total N	Available P	Potential K	Potential P-	CEC	Cation$_{ex}$				BS
								K	Na	Ca	Mg	%
		%		mg/kg				cmol$^{(+)}$/ kg				
Parbulu	6.86	0.32	0.09	4.7	8.0	9.0	6.95	0.26	1.17	1.71	0.27	48.9
Wamsait	6.25	0.64	0.13	9.4	7.4	8.3	10.64	0.26	0.40	2.23	0.52	32.0
Waekerta	7.25	0.25	0.09	7.1	3.7	3.9	6.16	0.14	1.08	0.95	0.28	39.7
Savana Jaya	6.65	0.25	0.07	10.6	2.4	7.0	8.47	0.10	0.40	3.16	0.48	48.7
Waelata	6.31	0.36	0.06	8.2	3.9	8.5	9.35	0.12	0.39	0.73	0.34	16.8
Wanareja	6.31	0.52	0.10	8.5	4.6	4.9	10.16	0.09	0.43	1.06	0.56	21.1

CEC = cation exchange capacity; BS = base saturation

Sago grows in swampy areas, marginal soils or high water content soil (Louhenapessy, 2008). In North Maluku, total N, P and K contents of sago soil are low; and land suitability for irrigated paddy field was only marginally suitable (S) due to N, P and K availability (Ibrahim and Gunawan, 2015). Long-term effect of low organic matter in paddy soil will relate to low potentially nitrogen mineralization and soil neutrality which in turn resulting in low nutrient availability (Sahrawat, 1983).

Table 3. Fertility status of soil in six villages at Buru District based on five soil chemical properties

Village	CEC	BS	Potential P	Potential K	Organic Carbon	Soil Fertility status
Parbulu	Low	Average	Low	Low	Low	Low
Wamsait	Low	Low	Low	Very low	Low	Low
Waekerta	Low	Average	Very Low	Very low	Low	Low
Savana Jaya	Low	Average	Low	Very low	Low	Low
Waelata	Low	Very low	Low	Very low	Low	Low
Wanareja	Low	Low	Low	Very low	Low	Low

Mercury levels in tailing and soil

Soil testing on total mercury level in soil verified that its level was differed depending on the vicinity of sampling point to tailing deposit (Table 3). Quantification of soil Hg used a strong acid mixed extraction method before Hg content was measured by using an atomic adsorption spectrometry device. This method measures the overall form of Hg; both unstable Hg-inorganic and organic Hg. Table 3 shows that Hg content in tailings and suspected Hg-contaminated area were higher than that of agricultural field that is far from the mine site. Generally the world's Hg levels in the world are 0.3-5 mg/kg (Steinnes, 1995); uncontaminated agricultural land contains Hg between 1-5 mg/kg, still in the normal range. Naturally agricultural fields (rice fields and vegetable areas) in the working area contains high enough Hg even two area contain Hg slightly that exceeded the upper limit of 5 mg/kg.

According to Government Regulation no. 82 year 2002, maximum permitted level of Hg in environment is < 0.001 mg/kg in water, and <0.15 mg/kg in soil. According to Steinnes (1995), Hg levels in soil depend on the process of soil genesis and mineral composition of parent material. However, most top soils and tailing pile contained increased amounts of Hg, especially near mining area (Table 3). This finding agreed with our previous study. Mercury in two soil samples collected from rhizosphere of pioneer plant grown in tailing in Waeapo village was 9.06 and 20.36 mg/kg respectively.

In general, the accumulation of Hg in the soil relates with deposition from anthropogenic activity through biosphere or atmosphere. Tailing pile in Namlea (Table 3) contained up to 166.10 mg/kg of Hg due to double gold extraction by using more Hg. Mercury is also a volatile metal that can be transported over a distance of 20-100 km. Therefore, naturally, soil enrichment by Hg is rarely observable on a wide scale. In this study, it is clear that Hg content in the suspected uncontaminated soil is far below the Hg level on agricultural field affected by gold tailings. Mercury and their compounds present in the soil fractions vary in the degree of mobility (Kabata Pendiaz and Mukherjee, 2007). Similar to other soil heavy metals, mercury bioavailability is regulated by physical, chemical and biological processes and interactions between them (Steinnes, 1995). Elevated level of mercury in gold-mine tailing is supposed to threaten rice production but the fact that soil reaction is neutral; and the availability of mercury in soil to plants might be quite low.

Mercury levels in edible parts of food crops

Maximum permitted mercury in edible parts of plant was 50 μg/kg according to Bradl (2005). Mercury in edible parts of vegetables grown on tailings-free field was lower than 50 μg/kg whereas those grown on tailings as well as on tailings-affected land contained more than 50 μg/kg of Hg (Table 4). Mercury-contaminated rice was an intense issue in Buru but it was not supported by real data. This study showed that grain from rice field irrigated by water from gold mine area only contained 31.16 μg/kg of Hg; slightly higher than those received uncontaminated water; 25.96 μg/kg. Low Hg levels in rice grain is an indication that rice from Buru might not contaminated by Hg.

Table 3. Mercury levels in soil, tailings and agricultural field in Buru District

No	Sub district	Village	Agricultural Field	Hg Level (mg/kg)
		Contaminated Agricultural Field		
1	Waelata	Wamsait (Path A)	Grassland soil	30.66
2		Wamsait (Path B)	Mixed garden soil	13.70
3		Wamsait (Path C)	Mixed garden	15.08
4		Wamsait (Path C)	Former paddy field soil	22.51
5		Parbulu	Tailing on Watercrest area	96.47
6		Parbulu	Tailing on cocoa plantation	22.19
7		Parbulu	Submerged tailing	26.97
8	Waeapo	Wanareja	Paddy field soil	35.68
9		Wanareja	Soybean field soil	9.92
10		Wanareja	Paddy field (1) soil	19.74
11		Wanareja	Paddy field (2) soil	26.98
12		Waikerta	Tailing on fruit orchard (1)	26.06
13		Waikerta	Tailing on Fruit orchard (2)	11.37
14		Waikerta	Tailing on Cocoa plantation	19.50
15		Waikerta	Tailing on vegetable area	11.01
16	Namlea	Siahoni	Tailing in mining installations	166.10
		Uncontaminated Agricultural Field		
1	Waelata	Parbulu	Paddy field soil, Parbulu	5.84
2		Wamsait	Grass field (former paddy field) soil	4.22
3	Waeapo	Waikerta	Paddy field soil	3.78
4		Savana Jaya	Vegetable área soil	1.64
5		Waetele	Paddy field soil	2.81
6		Wanareja	Paddy field soil	5.36

Agricultural lands covered with Hg-contaminated tailing pose two main problem. First, tailings pile on agricultural area will decrease soil quality. Tailing from Anhoni river inhibited nodules formation and groundnut growth in pot culture (Pramya, 2015) although liming and organic matter application was carried. Mine tailings present only a suitable substrate for establishment native plant species such as *Lygeum spartum*, *Zygophyllum fabago* and *Piptatherum miliaceum* (Conesa *et al.*, 2007). A short-term experiment indicated that native plant species *Prosopis velutina* and *Amaranthus watsonii* grow in metal mine tailings are potential for heavy metal phytostabilization (Santos *et al.*, 2017). Native plants have colonized some Buru tailings area, although the distribution is localized and uneven; their slow growth may not sufficient to cover tailing deposit area in short time. Native plants grow on Buru tailing mostly grass of *Cyperus* sp. *Eulesin* sp., and *Cynodon dactylon*. Revegetation of tailing on agricultural soil is the first step to improve soil quality in order to be used as food crop's plant substrate. Using native plant might be a more effective and less cost rehabilitation although take a relatively long time.

The second problem is crushing materials in the process of gold extraction causes heavy metals is released to the environment, not just Hg. Tailing pile in Buru contain Zn and Cd, similar to tailings of gold mines in Thailand which contain Co, Cu, Cd, Cr, Pb, Ni and Zn (Changul et al., 2010). In 2015, the Energy and Mineral Resources Department of Maluku Province has collected tailings from rivers and river basin and that deposited in watersheds. The reduction of toxicity and heavy metal content should be done before tailing release to agricultural field.

Table 4. Levels of mercury in edible part of plant grown in the tailings pile, tailings-impacted area and free-tailing agricultural field.

Sub District	Village	Agricultural field	Plant/ Commodities	Hg (μg/kg)
Waelata	Wamsait	Tailing deposit	Grass	76.95
		Agricultural area, irrigated by waste water from mining installation	Cassava tuber	89.05
			Cassava leaves	80.78
		Tailing affected agricultural area	Tomato	78.94
			Eggplant	66.54
			Watercress	78.64
			Cai sim	84.92
			Cauliflower	90.12
			Chili	89.05
	Parbulu	Paddy field, lower zone of mining installations	Paddy grain irrigated with contaminated water	31.16
			Paddy grain irrigated with uncontaminated water	25.96
			Cucumber	43.42
		Home garden has been irrigated with supposed Hg-contaminated water	Chili	6807
			Eggplant	73.43
			Local bay leaves	56.28
			Local squash	28.56
		Near mining installations	Watercress	70.52
Waeapo	Savana Jaya	Uncontaminated vegetable area	Spring onion	31.62
			Cucumber	38.21
			Long bean	41.73
			Cauliflower	46.78
			Tomato	46.94
	Waekerta	Uncontaminated vegetable area	Tomato	49.08
			chayote	48.77

Conclusion

Gold mining activities in the Botak Mountain of Buru Maluku District produced Hg-contaminated tailings deposited at river and water shed area near mine site, and agricultural land. The mercury content in tailings and tailing-contaminated soil were above the average mercury in the world's soils but their levels in some agricultural area far from mining installation were low and within the normal range. Edible parts of most food crops grown on contaminated soil contained Hg more than permitted level of 50 μg/ kg but Hg level of those in uncontaminated soil were below the threshold. The results of this study explained that mine tailing was contaminated by Hg, but agricultural land far from mining installation still contains Hg within the normal range. To ensure the safety of Agricultural area in the Weapo valley, it is necessary to delineate Hg within the agricultural area located in the vicinity of mining installation in Botak Mountain.

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