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Research Article
Assessing the Mercury Hazard Risks among Communities and Gold Miners in Artisanal Buladu Gold Mine, Indonesia

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Abstract

Background and Objective: Mercury (Hg) direct emission of amalgam open burning from Artisanal Buladu gold mine is the main Hg pollutant point source of air ambient, atmospheric deposition, surface water, soil and plants. This process has been occurred for more than 25 years in the Buladu gold mine but no informative data from this area is available. This study aimed to investigate the Hg emission in air, atmospheric dry deposition and Hg inhalation rate among the children school, communities and miners in the mining area. **Materials and Methods:** Dry deposition sampling was conducted using a set of glass dish sampler. Both Total dry deposition and THg atmosphere were estimated in the summer which corresponded to the climatologically dry seasons in Gorontalo Province, like wise personal inhalation rate were measured among the school children, communities and gold miners in nine respondents each. Statistical analyses of the survey data were performed using Stata 13.0 (Stata, College Station, TX, USA) and SPSS (Version 16). In addition to descriptive analyses, descriptive table, Student t-tests were used to compare means of Likert scales. **Results:** The average of total atmospheric of Hg showed a small concentration (0.0034 μg m^{-2}) in the background site whereas highly elevated (1.19-1.54 μg m^{-2}) in the study area. In addition, dry deposition Hg show a similar level (0.06 μg m^{-2}) in background and (1.11-1.54 μg m^{-2}) in the study area, respectively. Since St. 2 is the closest distance from the point source, the peak level THg generated by the direct fall of THg in the site. Result of target hazard quotient of the Hg are in the decreasing order of mine workers >community >students and their highest risk values are 1.54, 0.98 and 0.93, respectively. Finally, the THQ values obtained from this study due to this primary exposure route for the Hg investigated were higher than for all in the working mine site. **Conclusion:** The results showed that the working years have reasonable correlation with the sum of the positive findings in the 10 neurological symptoms among miners. Then, the closest Hg source distance the higher Hg concentration accumulation found. The recorded mercury in Buladu gold mine area showed that the magnitude concentrations of THg (p) in amalgam burning centre was higher than those of school and in community area.

Key words: Air Hg emission, atmospheric dry deposition, total mercury, artisanal mining, personal hg inhalation, gold miners, local community

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Competing Interest: The authors have declared that no competing interest exists.

Data Availability: All relevant data are within the paper and its supporting information files.

INTRODUCTION

Mercury (Hg) air pollution generate disable and premature deaths annually worldwide, especially in the countries that have mercury mine site. Those artisanal mine making poor air quality one of the most serious environmental health risks in the world. It is associated with the input of Hg into air, water, vegetation and the surface soil, the resulting toxicity of which can harm public health assessment of potential hazard mercury (Hg) is necessarily required in order to secure the environmental sustainable and the human health[1]. Contamination of the environment by mercury (Hg) is a major concern because of the impact to human activities on its cycling, toxicity and its bioconcentration potential[2]. The Hg emission from gold mining during the amalgam open burning is one of the most important pollutant anthropogenic Hg sources. Global emissions to the atmosphere from Hg mining is estimated about 10-30 t annually and probably exceeded 10,000 t historically[3]. Large quantity of Hg has been released into the atmosphere from gold mining activity in Buladu gold mine. One of the major Hg contamination sources in the air, water, soil and sediment is the burning of Au-Hg amalgam. As it is a dangerous practice delivered by the gold miners is the gold recovery process from the amalgams that routinely conducted daily. In this process, gold is recovered simply by evaporating the mercury from amalgam over an open burning. The metallic Hg discharge into the river and sea during the amalgamation process represents the second large source of contamination.

The magnitude emission of Hg from gold mining is one of the most important anthropogenic sources. Global emissions to the atmosphere from Hg mining are estimated as 10-30 t year^{-1} currently and probably exceeded 10,000 t historically[3]. It has been recognized that continual atmospheric deposition as a necessary source of mercury contaminants to aquatic and terrestrial environments[4-7]. Atmospheric mercury deposition occurs through both wet and dry processes. Wet deposition of Hg is defined as the air-to-surface flux in precipitation (occurring as rain, snow, fog or ice) which scavenges mainly GOIM and Hg(p) from the atmosphere, whereas dry deposition is Hg deposition in the absence of precipitation[8].

In recovering amalgam which is simply burned to isolate the gold before being sold to gold shops or gold collectors[9], Hg vapour cools it forms airborne liquid droplets or aerosols. This component rapidly deposits on nearby walls and in soil, adding to the high proximate contamination around and inside gold shops. The Hg compounds in the atmosphere exist in vapor and particulate forms, preferentially partitioning into the vapor phase. Mercury species fall within two main categories: Inorganic Hg compounds and organic Hg compounds. The most common form of inorganic Hg is elemental Hg vapor[10]. Quantifying the Hg dry and wet deposition is obviously necessary in order to minimize the big gaps that exist in estimating the global Hg release in the atmosphere[11]. In addition, Hg deposition rate assessment and source investigation the deposited Hg are also required for the development Hg emissions control policy[8].

The Hg vapors inhalation from open burning may generate vomiting, gastroenteritis, complaints of the kidney and urinary trace, ulcerations in the gums and extreme light sensitivity known as 'photophobia'. If inhaled over a long period, it can cause chronic mercurial poisoning resulting in kidney ulcerations, Hg deposition in the body, speech disturbances and lack of concentration[12]. The situation is also occur in Buladu gold mine as the activities have been continuing conducted for their daily necessary income. This study is necessary needed to investigate the Hg atmospheric deposition and the inhalation rate for all communities in the gold mine site.

MATERIALS AND METHODS

Study area and sampling sites: This study was conducted during the period of 5 months from May-September, 2014 in Buladu artisanal gold mine, Gorontalo province, Indonesia. It has a tropical climate influenced by summer season that usually runs from June through October and rainy season from November through April. The North wind blows in from January-March (summer season), followed by West wind are from April-May. In addition, the East Wind (November-December), which the sea state is quite strong with high sea waves ranging from 1-2 m. The gold mine site was in a small town with a population of around 5800 inhabitants which is located in the North of Gorontalo Province, Indonesia (Statistic of Gorontalo Province, 2010). The Hg emission to the atmosphere due to amalgam open burning and directly discharged of wastewater into the Buladu River which will be end up in the Sulawesi Sea, without any previous treatment become an emergency case. This activity is continually practiced by miners. Samples were collected in Buladu study area and in the vicinity site as the background sample. Artisanal Buladu gold mine is located in Sumalata District, North Gorontalo Province, Indonesia. It is an active and important gold mine which has been managed both in traditional way by local communities and in technological system by mine factories.

Samples were collected both dry deposit and atmospheric Hg in three different sites (in tromol amalgamation centre, in the schools and in community in Buladu) with 9 samples each sites during the period of 3 days. Likewise, Hg inhalation rates were measured to the miners, school children and communities for nine respondents each, respectively.

Sample collection and analysis for particulate mercury in air: Sampling and analysis of particulate Hg samples were collected and analyzed in a manner previously described by Lindberg *et al.*[8]. Briefly, total Hg(p) samples were collected using 47 mm open-faced Teflon filter packs and fine fraction (02.5 mm) Hg(p) samples were collected in a Teflon filter pack attached to a Teflon-coated aluminum cyclone (URG Corporation, Chapel Hill, NC). Total and fine Hg(p) was collected at 30 and 16.7 l min^{-1} nominal flow rates, respectively. Hg(p) filters were microwave digested in HNO_3 and subsequently analyzed using a dual amalgamation CVAFS technique Gambrell[13].

The performance characteristics of this technique were previously reported by UNEP [14], where the System Detection Limit (SDL) for Hg(p) was determined to be 1 μg m^{-3}, calculated using 3 sec of the field blanks. The precision of collocated samples w as 91% and the analytical precision was 97%.

Sample collection and analysis for Hg dry deposition: To measure the contaminant level of dry deposition, the atmospheric fall out particulates were collected. In consideration of possible re-suspense of ground contaminant, dry dish sampling set points were located at level of 2.5 m high of above the ground. A set of glass container (27×17×10 cm) was used to collect THg particulate in the atmosphere. The containers were filled with deionized water about 5 cm at depth of container to avoid of over evaporation, dry dishes container was refilled every day for 5 days of collection with deionized water. In the event of rainfall, deposition dish glass were manually covered with fitting plastic covers to prevent potential contamination from rainfall. These covers were removed immediately following the completion of the rain threat. Samples were analyzed for total mercury.

Nine stations of dry deposition sampling in school area and three stations in community surround the open burn from daily tromol activity were set to measure the THg atmospheric particulates. After sufficient days, samples were collected, the containers were rinsed using the deionized water then, be analyzed by atomic absorption spectrophotometer, the reduction limit was 0.001 μg L^{-1}.

Samples collection for mercury inhalation rate: Inhalation rate was measured to the Hg miners. Local residents in Buladu gold mine site as well as the children in 3 schools in Buladu and surround were selected. The sampling campaign was conducted in February, 2012. The respondents selected for 9 miners who work in the open burning centre, 9 participants (local residents) and 9 school children in 3 different schools, respectively for the investigation. We collected the Hg inhalation sample for 3 days. The only criterion to select the participants was that they were the local residents and lived in their homes at least for 3 years before the sampling process. The recruitment strategy was to include the worker in the amalgam open burning centre by visiting them at the site. The number of the respondent were poorly selected due to the limited personal inhalation rate devices available, since we measured at miners, local people and school children at the same time. Therefore, these numbers were the maximum numbers we could get from each site during the period of 3 days measurement.

A questionnaire was utilized to collect information on the respondent history, occupational history, dietary habit, life style (smoking), including exposure related questions and asking for typical symptoms of mercury intoxication and other related health history. The present study obtained the ethics approval. All participants joined the investigation on a voluntary basis without any forces and signed the inform consent initially.

Quality control-Quality assurance: Total mercury (THg) concentration was determined using acid digestion followed by Cold Vapor Atomic Absorption Spectrometry (CVAAS). The detection limit (3r of the blank measurement, n = 10) was 0.03 μg. Quality assurance and quality control of the analysis of the THg concentration was determined through the use of 3 duplicates, method blanks and a certified reference material (NMIJ7302-a). The average total Hg concentration of the environmental NMIJ7302-a CRM was 0.53±0.04 mg kg^{-1}, which is comparable to the certified value of 0.52±0.03 mg kg^{-1}.

Analysis procedures: All particulate measurements were performed using separate day time in order to determine potential differences in the chemical characteristics of the day time and night time atmospheric surface layers. After the

acid digestion and BrCl treatment, an aliquot of the sample is analyzed according to the procedure described for precipitation samples.

Ethical consideration: The miners, school child guardians and the people living in the area who were requested as the respondent signed an informed consent letter prior to inclusion in the research. School children were guarded by their parent either father or mother. Confidentiality of initial information and freedom to withdraw from the study anytime was stipulated and without any force from the third parties. Those who found to have health concerns will be provided with the appropriate management and informed secretly as necessary.

Statistical analysis: The statistical analysis of total mercury concentrations among the gold miners, school children and communities samples was conducted using SPSS software (version 16.0, SPSS Inc., Chicago, USA). An unpaired (two-tailed) non-parametric t-test with a 95% confidence level was used to determine the difference between the total mercury concentrations in those three places as well. Correlations between the total mercury concentration and working years of the gold workers were determined using Spearman correlation analysis. Less than $p<0.05$ was considered significant.

RESULTS AND DISCUSSION

Atmospheric total mercury ambient air: Air sampling results showed that the sampling sites were higher than the recommended EPA guidelines for total mercury concentration in ambient air. The highest air concentration of mercury was recorded at open burning centre with 1.19-1.54 μg m^{-3}, Table 1.

Mercury levels in the ambient air were considerably higher in this Buladu site than results obtained in the active mining site in Toi gold mine, Luwuk with 0.455μg m^{-3} and in Slovenia which ranged from 0.01-0.49 μg m^{-3}. This THg level obtained could significantly contribute to the mercury burden of residents in the area especially among the school children whose school is in the breathing zone. Likewise, because children are small, they are close to the ground where they also crawl and play and are possibly exposed to dust and particulates that accumulate on the floors and soil[14-16].

THg for dry deposition: The highest elevation of THg concentration of dry deposit fall out was in St. 2 in amalgam open burn centre, then followed St. 3 in community area and St. 1the school, respectively. Since St. 2 is the closest distance from the point source, the peak level THg generated by the direct fall of THg in the site. St. 3 and St. 1 stations however are located quite far to the site of open burning central, with about 300-500 m where the results of amalgamation could be blown by wind to these both sites. In addition, open burn from gold buyer shops are also near to St. 3 which may release Hg into the atmosphere and drop to deposit on the site. The magnitude of Hg release to atmosphere is governed primarily by the scale of the open burn of amalgam and the Hg concentration evaporated in the waste. Table 2 summarized dry deposition rates in artisanal Buladu gold mine in Sumalata.

The deposition rates at open burning centre of about (0.11-1.52 μg m^{-2} $month^{-1}$) was higher than that of Hg (p) < 2.5 (0.04-0.32 μg m^{-2} $month^{-1}$) in community area and those at school. The peak deposition rates for Hg(p) <1.52 were recorded in the 3rd day where only 1.11 in February, 2012 at the same station, where as the lowest levels of Hg (p) were at the school site ranged from < 0.43-0.87 respectively. The lowest recorded values were in the school where as the

Table 1: Concentration of atmospheric total mercury ambient air in school, amalgam open burning site and in community

		THg of atmospheric ambient air			
		Days			
Stations	Stations description	1	2	3	Standards
1	In school	0.93	0.77	0.68	USEPA RFC
		0.79	0.82	0.49	0.4(μg m^{-3})
		0.87	0.57	0.48	
2	In amalgam open burning centre	1.29	1.19	1.54	
		1.34	1.37	1.54	
		1.22	1.28	1.46	
3	In community area	0.81	0.95	0.88	
		0.91	0.86	0.91	
		0.56	0.90	0.70	

Table 2: Deposition rates μg m^{-2} $month^{-1}$ at schools, amalgam centre and community area

		THg(p) of dry deposition			
		Days			
Stations	Stations description	1	2	3	Standards
1	In school	0.87	0.75	0.54	USEPA RFC
		0.77	0.80	0.44	0.4 (μg m^{-3})
		0.75	0.52	0.43	
2	In amalgam open burning centre	1.22	1.12	1.51	
		1.25	1.11	1.52	
		1.20	1.15	1.44	
3	In community area	0.76	0.94	0.76	
		0.87	0.75	0.86	
		0.55	0.87	0.69	

highest one in the amalgam burning centre. Study of Hg (p) Measurements in Toronto in the recent years revealed the high concentrations of atmospheric mercury in summer season which was consistently with the study in Buladu Gold mine. In addition, Base on the data analysis, it has found that buildings, sewage treatment, metal as well as chemical production potential to be the sources of Hg emission to the atmosphere[2,17-18].

The average of mercury dry deposition rates were found to be 0.71 μg m^{-2} $month^{-1}$ in school <1,36 μg m^{-2} $month^{-1}$ amalgam open burning centre >0.83 μg m^{-2} $month^{-1}$ in community area, respectively that represent the dry season. Based on the values, we can depict that the total atmospheric mercury dry deposition, forecasted as the sum THg(p) is 34.8 μg m^{-2} $month^{-1}$ or 1044 μg m^{-2} $year^{-1}$. Similarly, the lower mean of mercury dry deposition rates were found to be 0.65 μg m^{-2} $month^{-1}$ in school <1.28 μg m^{-2} $month^{-1}$ amalgam open burning centre >0.78 μg m^{-2} $Month^{-1}$ in community area, respectively in February, 2012 that represent the rainy season. Although it has some uncertainty in calculating the Hg dry deposition, It should be noted that the use of monthly values used to estimate the annual deposition rate of THg(p) in this case.

The Hg species brought in through long range transport in the warm season might also have contributed to the high deposition rate in the summer[19]. Comparison of particulate mercury reveals that particles having a size >2.5 mm carried up to 83% of the THg(P) to the surface through dry deposition. Other study implied a high THg(p) deposition rate which could be compare was in Beijing with the average THg(p) deposition rate of 407 μg m^{-2} a^{-1}[20], then study did in Sha-Lu, Taiwan revealed that average THg(p) deposition rate was 500 μg m^{-2} a^{-1} [21].

THg of Inhalation rate: The estimates of total mercury ambient air concentration were compared with inhalation reference values for elemental mercury to forecast the potential health impact of exposure from amalgam burning at the site. Most of the inhabitants of Buladu area was estimated to have been exposed to maximum 1 h concentrations of mercury. Table 3 indicated the inhalation rate for 1-3 days exposure with the control in 3 different research sites.

This value is based on the most sensitive, relevant adverse health effect and is a concentration at or below which there are expected to be no adverse health effects for a 1 h duration of exposure.

Target Hazard Quotient (THQ) of inhalation rate: The U.S. Environmental Protection Agency (US EPA) and the National Academy of Sciences recommend keeping this corresponds to a reference dose (RfD) of total mercury do not greater than 5.0×10^{-4} μg×g^{-1} day^{-1}. Non-cancer risk assessments were typically conducted to estimate the potential health risks of pollutants using the Target Hazard Quotient (THQ), it is a ratio of the estimated dose of a contaminant to the dose level below which there will not be any appreciable risk. If the value of THQ is less than unity, it is assumed to be safe for risk of non-carcinogenic effects. For non carcinogenic effects, risk is expressed as a THQ, the ratio between the exposure and the reference dose.

The health risks resulting from the air contaminated Hg inhalation of the students, workers and communities in the mining sites area have been estimated based on target hazard quotient (THQ). THQ is either $\geq$1 or <1, where THQ >1 indicates a reason for health concern. It must be noted that THQ is not a measure of risk but indicates a level of concern and while the THQ values are additive, they are not multiplicative. In this study, the THQ values were calculated using the measured concentrations of the three different sites for Hg pollutant obtained for the analysis of the inhalation rate. The results of hazard quotient risks of heavy metals exposure route are shown in Table 4.

Table 3: THg of inhalation rate of students, miners and communities around the amalgam centre, Buladu

		THg inhalation rate			
		Days			
Stations	Stations description	1	2	3	Chemical concentration in air (control)
1	Student in school near amalgam	7.76	6.39	5.66	0.0048
		6.56	6.83	4.10	0.0048
		7.21	4.76	3.98	0.0048
2	Worker in amalgam centre	12.09	11.20	14.50	0.0051
		12.64	12.87	14.51	0.0051
		11.84	11.99	13.77	0.0051
3	Community near amalgam centre	7.92	9.33	8.61	0.0049
		8.92	8.38	8.94	0.0049
		5.46	8.81	6.87	0.0049

Table 4: Target Hazard Quotient (THQ) of inhalation rate of students, miners and community in the amalgam centre

		THQ of Hg inhalation rate			
		Days			
Stations	Stations description	1	2	3	Standards
1	Student in school near amalgam	0.93	0.77	0.68	>1 at risk
		0.79	0.82	0.49	
		0.87	0.57	0.48	
2	Worker in amalgam centre	1.29	1.19	1.54	
		1.34	1.37	1.54	
		1.22	1.28	1.46	
3	Community near amalgam centre	0.81	0.95	0.88	
		0.98	0.86	0.91	
		0.56	0.90	0.70	

Results indicated that the target hazard quotient of the Hg are in the decreasing order of mine workers >community >students and their highest risk values are 1.54, 0.98 and 0.93. The THQ values obtained from this study due to this primary exposure route for the Hg investigated were higher than for all in the working mine site. 1 whereas in community and children were less than 1. This indicates a potential health risk hazard to the exposed population at the moment among miners. By contrast, no risks existed both in the community and school children, However, due to bio-accumulative nature of these Hg in the accumulation human body, moderate amount of inhalation of this Hg air emission is advisable by reducing exposure[15].

CONCLUSION

The total mercury accumulation tends to increase with the increase of exposure and working years. Dry deposition was shown to be influenced by temperature changes with higher deposition in summer and lower in rainy season. This suggests the effect of mercury exposure duration play important role in determining the Hg accumulation. Several neurological symptoms were noted in the majority of the gold miners. The gold miners who have a high-level of mercury were diagnosed were subject to a number of neurological symptoms. The awareness of gold worker about adverse health effects of mercury exposure should be raised.

SIGNIFICANCE STATEMENTS

This study discovers the potential effect of Mercury hazard risks among school children, miner and community who are living in the Hg contaminated site. This study will help the researchers, Miners and communities to manage the Hg pollution in reducing it earlier before endangering the health and the environment. Thus, a new method and a good design of closed technology need to be applied in amalgam burning processes.

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