# Impact of mercury level and health assessment of artisanal gold miners of Gorontalo Utararegency, Gorontalo Province, Indonesia

Yayu I. ARIFIN[1, 2], Masayuki. SAKAKIBARA[1], Koichiro SERA[3] zuhriana k. YUSUF [2], Nanang PARAMATA[2],

## Abstract

**Background** : Mercury concentrations of the hair sample from Sumalata, Anggrek, Tolinggula dan Kwandang of North Gorontalo regency have been documented, a home of active and oldest artisanal and gold mining (ASGM) sites. However, neither status of contamination of human nor its sources, routes and medical health condition have been elucidated.

**Methods:** Concentration of Mercury in hair of inhabitants who are living in villages around ASGM sites in Gorontalo Utara regency of Gorontalo Province, Northern Sulawesi, Indonesia,were investigated using proton induced x-ray emission (PIXE). Hair samples (n=95) were collected between 2012 and 2013 from inhabitants in Anggrek, Sumalata and other of Gorontalo Utara regency.

**Results** : Mean mercury's hairs concentration of group of inhabitants of Sumalata and Anggrek are higher than Tolinggula and Kwandang. Mean of hair mercury of female's inhabitants is higher than male's, in each group. This is showing that female inhabitants are morepermeable to mercury contamination compared to male. Neurological examinations wereperformed on 25 participants. From 12 symptoms objectives were searched, there are three common symptoms which already observed, namely Bluish of gums, Babinski reflex and tremor. Group of people of Sumalata with hair mercury level of 6.2 – 10.4 were showing at least three dominant symptoms, while group of people of Anggrek with mercury level of 3.4 – 14.9 were dominantly showing only Babinski reflex.

**Conclussion**s : Such difference maybe caused by higher metylmercury concentration in the body Sumalata people due to longtime mercury exposure from ASGM in Sumalata which are older than ASGM activities in Anggrek.

## Introduction

The Gorontalo province of Northern Sulawesi, Indonesia, has several Artisanal and Small scale gold mining (ASGM)site in each regency: (1) Pohuwato regency: Gunung Pani and Bulontio; (2) Boalemo regency: Bilato; (3) Bonebolango regency: Tulabolo; and (4)

Gorontalo Utara regency: Hulawa and Ilangata villages. The ASGM activities in Hulawa village of Sumalata subdistrict are started since 1970s, while ASGM in Ilangata and Illangata Barat villages of Anggrek Subdistrict just started 5 years ago. Every year, about 572 kg of mercury contaminates the environment of Gorontalo Utara regency [1].

Mercury contaminations and health assessment of miners and inhabitants from some ASGM sites in Indonesia namely Talawaan, Tatelu, Galangan and Sekotonghave been reported [1-6]. The Human hair is one of the biological sample. Human hair as sample for determination of mercury levels in the human body; more than 40 elements have so far been detected in hair [7-9]. Health status of miners mainly determined using standardized protocol which performed by medical doctors. Relationship between mercury in hairs, habits, health status and localization of ASGM are often discussed [10-13].

Scalp hair analysis was used as first step for risks assessment of heavy metals exposure to human body of people who are working and living in the vicinity of mine area, outsideand in metropolitan city [5, 14-17]. The advantages of using hair sample for monitoring the impact of environmental pollution on human health were reported elsewhere [18-20].

This study is aimed to find out the mercury contamination of people of Gorontalo Utara regency; who are living nearby ASGM sites and others who are living in Gorontalo utara. We used scalp hair as a bioindicator of mercury exposure to human body. Health conditions of miners and inhabitants who living around the mining sites were inverstigated using standard protocol of neurological examianation.

## Methods

### Study Area

Samples were collected from five locations in Gorontalo utara regency:Anggrek (district), Kwandang (district), Monano (district), Sumalata (district) and Tolinggula (district) (see Fig. 1).Geographically Tolinggula, Sumalata, Monano, Anggrek and Kwandang situated on hills and mountains along coastline of Gorontalo Utara regency. Inhabitants of Gorontalo Utara regency mainly works as farmer and fisherman.

Seawater fishes are common diet alongside with rice, corn and vegetables which is also produced near by hills alongside the coastline. Annual mercury emission from ASGM activities in Sumalata and Anggrek subdistricts are estimated contaminates the environments along coastline of Gorontalo Utara regency. Accumulation and biomagnanification of

mercury which may occur in living organisms like paddy rice, corn and sea water fishes become agents of spreading of mercury contamination through food web of inhabitants of Gorontalo Utara regency.

Sumalata and Anggrek subdistricts are locations of ASGM activities, while Kwandang, Monano and Tolinggula are subdistricts without mining activities. The residents of Anggrek and Sumalata are considered as ASGM miners groups, while residents of Kwandang, Monano and Tolinggula are considered as the control group.

Mercury concentration in hair sample will be used to characterize the risk by compared with reference values published by the German Human Biomonitoring Commission in 1999 (Commission Human –Biomointoring of the Federal Environmental Agency Berlin, 1999) (see Table 1).

Table 1 Toxicology established threshold limits for mercury in hair; HBM = Human Bio Monitoring (Commission Human –Biomonitoring of the Federal Environmental Agency Berlin, 1999)

| | Hg hair (ppm) | Note |
|---|---|---|
| Below HBM I | 0 – 1 | Normal |
| HBM I to HBM II | 1-5 | Alert level |
| Over HMB II | >5 | High level |

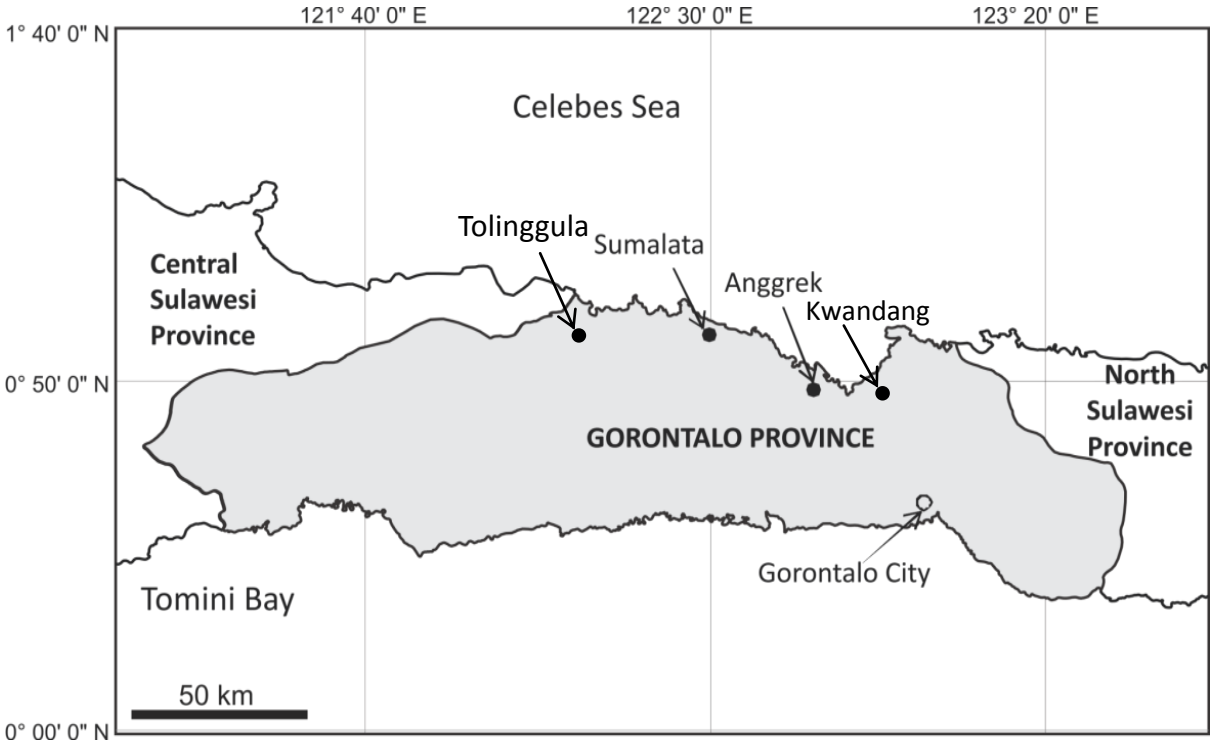


Fig. 1. Map showing sampling locations (●) of human hair from Gorontalo, showing Tolinggula Sumalata, Anggrek, Monano and kwandang districts.

**Hair Sampling**

Human scalp hair samples were taken from of95participating inhabitants who have range of agefrom 8 months to 63 years. The distribution of participants according to sex, locations and occupations are summarized in Table 2. About 10-20 strands of hair were cut close to skin from righ backside (mastoidal region of the temporal bone) and then labeled and stored in sample plastic bag [18].

Table 2.Information on donors of hair samples in Gorontalo Utara

| Group | Anggrek | Kwandang | Monano | Sumalata | Tolinggula |
|---|---|---|---|---|---|
| All | | | | | |
| No. of samples | 25 | 7 | 37 | 23 | 3 |
| Age (years)[a] | (3 - 53) | (8 months - 62) | | (7- 63) | (17-46) |
| Male | | | | | |
| No. of samples | 14 | 1 | 15 | 12 | 0 |
| Age (years)[a] | (3 - 53) | (2) | (7-14) | 36.3 ±17.0 | |
| Female | | | | | |
| No. of samples | 11 | 6 | 22 | 11 | 3 |
| Age (years) | (6 - 45) | (8 months - 62) | (6-37) | (7 - 63) | (17-46) |
| Occupation | | | | | |
| Mine Worker | 8 | 0 | 0 | 11 | 0 |
| House wife | 3 | 3 | 3 | 4 | 2 |
| Teacher | 0 | 1 | 0 | 0 | 0 |
| Univ. Student | 0 | 1 | 0 | 0 | 0 |
| Unemployed | 5 | 0 | 0 | 1 | 0 |
| Children ( 0 -17 years) | 4 | 1 | 34 | 7 | 1 |

**Analytical Procedure**

Elemental analysis for each sample was performed by **Particle Induced X-ray Emission (PIXE)** in cyclotron research center, Iwate Medical University, Japan. Precision and accuracy of this method having been reported elsewhere [21-22]. Hair samples were washed using milli-Q water and shake in ultrasonic bath for 1 minutes. Then dried by wiped it with tissuea. Dried hair samples again washed by stirred in acetone for 5 minutes. Then

washed again using milli-Q water, and wiped well with tissue and left dried in the room temperature.The hair samples (Approximately 7 hairs) were stuck on a target holder. A 2.9 MeV-proton beam hit the target after passing through beam collimator of graphite whose diameter of 6 mm. X-rays of energy higher than that of K-Kα line are detected by a Si(Li) detector (25.4μm-thick Be window; 6μm in active diameter) with a 300μm-thick Mylar absorber. For measurement of X-rays lower than K-Ka, a Si(Li) detector (80 mm Be; 4 mm in active diameter) which has large detection efficiency for low energy X-rays is used. Description of the data acquisition system and the measuring conditions are reported in the reference [22]. Typical beam current and integrated beam charge were 100nA and 40mC, respectively. Procedure of the standard-free method for untreated hairs is almost same as was reported in the previous studies [21].

**Neurologicalexamination**

Neurological examinations were performed by team of medical doctors using standard protocol to limited number of participants. The participants were 25 and 9 persons from Sumalata and Anggrek districts, respectively. The examinations were done on site : mining sites for miners and home for inhabitants.

There are 12 symptoms related to merury poisoning included in the neurological examination : 1. Signs of bluish discolaloration of gums, 2. Rigidity and ataxia ( walkin or standing), 3. Alternating movements or dydiadochokinesia 3. Irregular eye movements or so called nytagumes, 4. 5. Field of vision, 6. Knee jerk reflex, 7. Biceps reflex, 8. Babinski Reflex, 9. Labial Reflex, 10. Salivation and dyarthria, 11. Sensory examination, and 12. Tremor:tongues eyelids, finger to nose, pouring, posture holding and the romberg test). We used 1 and 0 for postively and negatively observed symptom, respectively.

**Statitical Analysis**

Data of Mercury hair sample and neurological examination were analysed statistically with Origin, Originlab (2007) version 8.0. Kolmogorov-Smirnov tests were used to study the normality of the distribution of mercury hair sample of inhabitants. Since the data are log-normally distributed, Kruskall-Wallis ANOVA test was used to see different between subgroups.

**Results**

**Mercury in hairs**

Distributions, range of mercury level of 95 hair samples collected from 4 districts are summarized in Fig. 2. Hair merury concentration of all participants are more than 1 ppm, which means toxicity level is in alert level already. Number of subjects with high mercury level over 10 ppm was 10 (40 %), 7 (30.4), 4 (8,5%) in Anggrek, Sumalata, control group (Kwandang, Monano and Tolinggula), respectively.According to Klomogorov-Smirnov test that distribution of data of mercury hair of Gorontalo Utara regency was not normal, instead of log normally distributed. The geometrical mean is more suitable for log normal data distriburion.

Table 3. Mean and range of hair mercury content of inhabitants of Gorontalo Utara regency.

| **Residence** | **Sex** | **n** | **Hair mercury content (ppm)** | | |
|---|---|---|---|---|---|
| | | | Mean ± SD | Min | Max |
| Anggrek | F | 33 | 14.2 ± 2.9 | 4.7 | 144.8 |
| | M | 14 | 7.0 ± 1.9 | 2.1 | 17.9 |
| | Total | 25 | 9.6± 2.5 | 2.1 | 144.8 |
| Kwandang | F | 6 | 6.7 ± 1.6 | 4.0 | 14.6 |
| | M | 1 | 3.5 ± | 3.5 | 3.5 |
| | Total | 7 | 6.1 ± 1.7 | 3.5 | 14.6 |
| Monano | F | 22 | 6.2 ± 1.6 | 2.8 | 28.1 |
| | M | 15 | 5.0 ± 1.3 | 3.5 | 6.9 |
| | Total | 37 | 5.7 ± 1.5 | 3.8 | 28.1 |
| Sumalata | F | 11 | 10.0 ± 2.1 | 3.8 | 69.8 |
| | M | 12 | 6.6 ± 1.7 | 2.5 | 13.7 |
| | Total | 23 | 8.0 ± 2.0 | 2.5 | 69.8 |
| Tolinggula | F | 3 | 5.0 ± 1.2 | 4.4 | 5.9 |
| | M | 0 | | | |
| | Total | 3 | 5.0 ± 1.2 | 4.4 | 5.9 |
| Total | F | 53 | 8.1 ± 2.1 | 2.8 | 144.8 |
| | M | 42 | 6.0 ± 1.7 | 2.1 | 17.9 |
| | **Total** | **95** | **7.1 ± 2.0** | **2.1** | **144.8** |

Average hair mercury level of all, male and female inhabitants in Monano district are 5.7, 5.0 and 6.2 ppm, respectively. Showing the lognormal distribution between male and female subjects. While average of all mercury hair content for female is 8.1 ppm, which is more than 50 percent higher than male (6.9 ppm), such conditions also found for subgroup of Kwandang and Sumalata. The condition in Anggrek is even higher, which is more than 3

times. Large discrepancy of mercury level between female and male inhabitants suggests that female inhabitant are more vulnerable to mercury contamination than male.

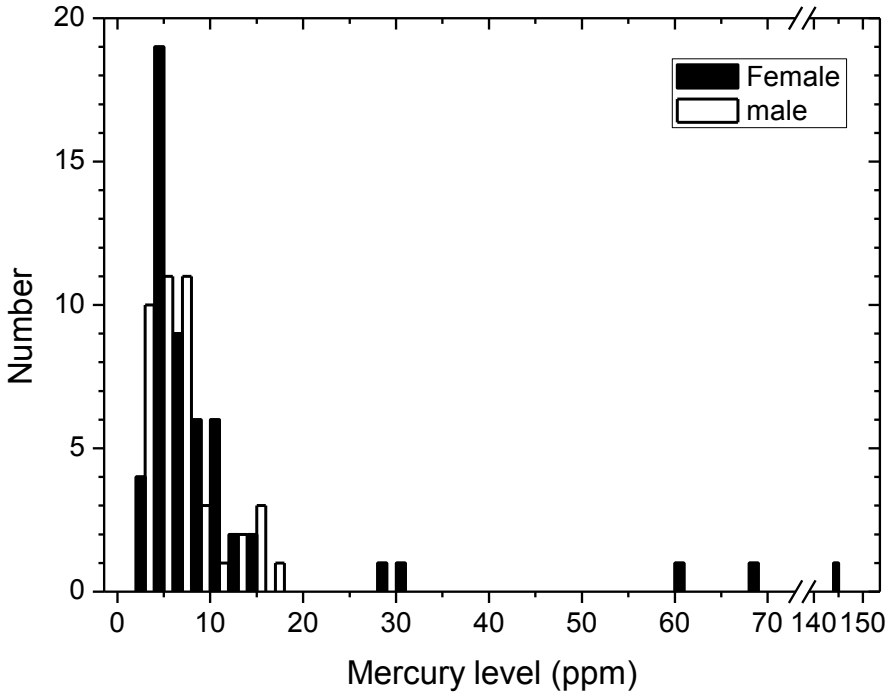


Fig. 2. Distribution of the hair mercury among the total population. Open bar and solid bar indicate male and female, respectively.

### 3.2. Relation between mercury concentration and age

Mercury concentration in human hairs are may depends on several factors, age is one of them. We have plotted in Fig. 3 mercury concentration vs age of miners and nonminers. Relationship between mecury cocentration and age of participant in both groups are determined using Spearman correlation coefficient which is summrized in Table 4. There is positive, strong and significant relationship ($r=0.31$; $p=0.01$) betwen age and mercury content of group of inhabitant, while for group of miners there was no significant correlation ($r=-0.16$;$p=0.44$). Such conditions imply that hair mercury concentration of inhabitants are age dependent while for miners, the correlation is unknown yet. There are some factors related to the hair mercury concentration which are not considered here for example : habit and food consumption and drugs.

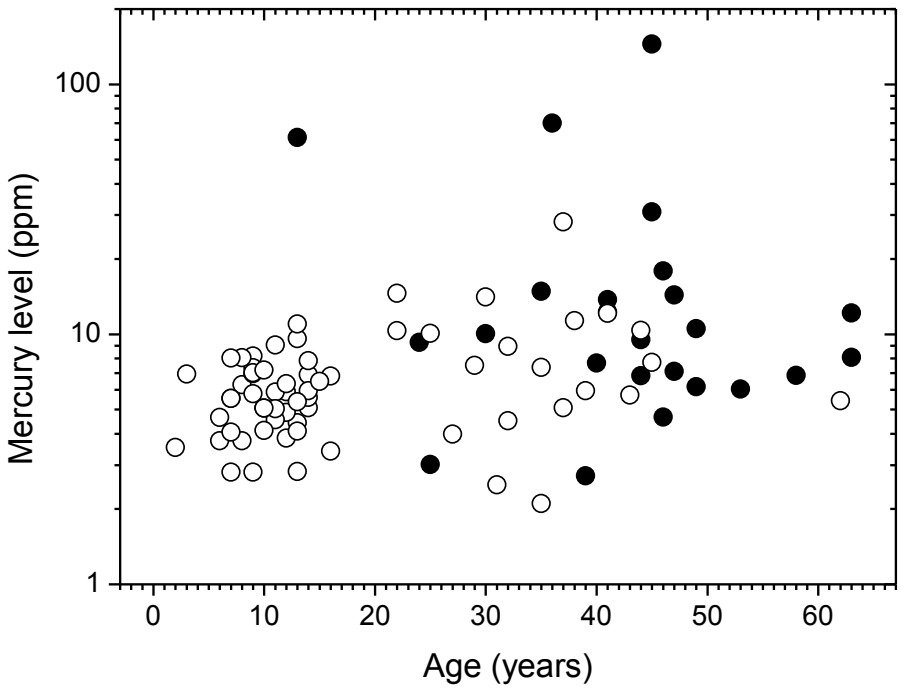


Fig. 3. Distribution of Mercury level of among population of Gorontalo utara regency. Open and closed symbols denoted for non miners and miners groups, respectively.

Table 3. Spearman correlation coefficient between Hg concentrations in hair and age of residents in Gorontalo Utara, Indonesia.

| Group | No | r | p |
|---|---|---|---|
| Miners | 25 | -0.16 | 0.44 |
| Non miners | 70 | 0.31 | 0.01 |

**Relation between mercury concentration and localization**

Comparison of hair mercury distribution among inhabitants of Anggrek and Sumalata districts (ASGM site) and inhabitants of other districts which doesnot have ASGM site is shown in Fig. 4. Mean hair mercury concentration (SD) of ASGM site and non ASGM site groups are 8.8 (2.2) and 5.7 (1.5), respectivley. We used KruskalWallis test to see the difference between both groups, there was no significant difference between two groups in the level of confidence 95%.

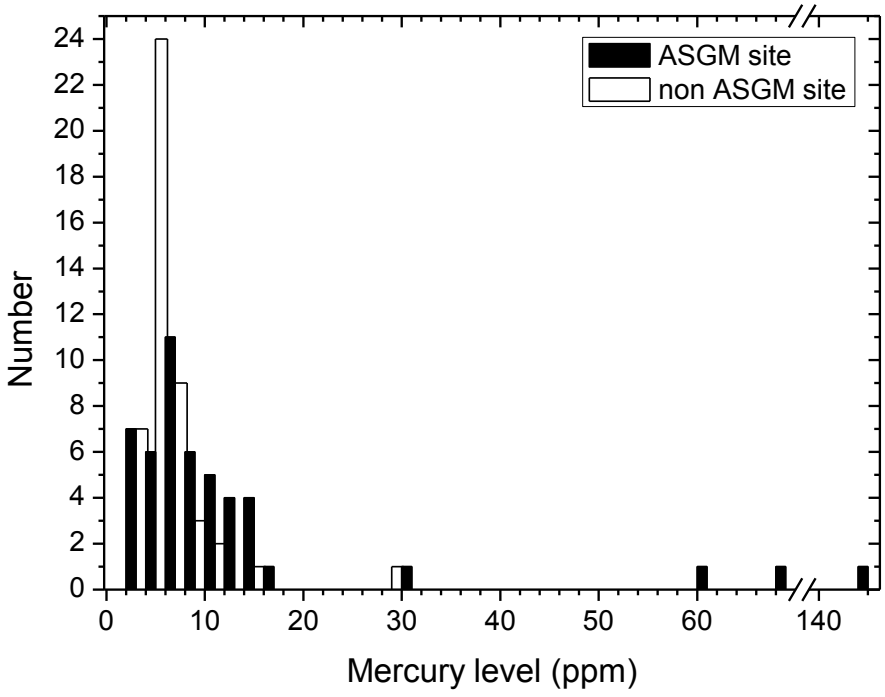


Fig. 4. Distribution of the hair mercury among the total population according to their location from ASGM. Black and white bar indicate ASGM site inhabitants and non ASGM site inhabitants, respectively.

**Neurological examinations**

From 12 objective symptoms were evaluated, only 3 (Blusih of gums, Babinski reflex and tremor) are positively observed in Gorontalo Utara regency miners and inhabitants. Miners from Sumalata and Anggrek already showed positive symptoms of Tremor and Babisnki reflex. The inhabitants of sumalata already showing symptoms of Bluish of Gums, Babinski reflex and tremor while in Anggrek only Babinski reflex is dominant.

Table 4. Objective symptoms of the participants

| | Bluish of Gums | | Babinski Reflex | | Tremor | |
|---|---|---|---|---|---|---|
| | No | Yes | No | Yes | No | Yes |
| Anggrek, Miners | 1 | 0 | 0 | 1 | 0 | 1 |
| Anggrek, Inhabitants | 7 | 1 | 2 | 6 | 6 | 2 |
| Sumalata, Miners | 0 | 3 | 1 | 2 | 0 | 3 |
| Sumalata, Inhabitants | 2 | 20 | 2 | 20 | 5 | 15 |

## Discussion

To the authors knowledge concerning, no study was recently done on mercury contamination using scalp hair as bioindicator and health effects of inhabitants of Gorontalo utara regency related to ASGM activities in Anggrek and Sumalata districts. Using scalp hair as bioindicator of mercury contamination are reflects mainly the uptake of oraganic mercury compound via fish consumtion [23].

Mean mercury levels of inhabitants from Anggrek and Sumalata are 14.2 and 10.0 ppm, respectively. The values are two times higher compared to smallest mean value (5.0 ppm) of Tolinggula district. From 21 subjects have mercury hair concentration more than 10 ppm, 15 (71.4 %) of therm are females and 4 (26.7 %) of those female lives outside ASGM area. Females which have hair mercury concentration of more than 10 ppm are potential to contaminates and risking abnormal brain development of their fetus [24].

Neurological disturbances were observed among ASGM miners and inhabitants of Anggrek and Sumalata are Babisnki reflex and Tremor. Relation between level of methyl mercury in scalp hair and neurological abnormalities found in adults is discussed by [25]. Although total hair mercury level found in miners and inhabitants of Anggrek who participated in neurological examinations are between (3.4 - 14.9) ppm but only Babinski reflex disturbances is dominant. Whereas miners and inhabitants of Sumalata with higher total hair mercury level (6.1 – 10.4 ppm ) already showing at least 3 disturbances (Babinski, Bluish of gums and Tremor). We may assume that probably Methylmercury levels of groups of Sumalata are higher than group of Anggrek, since they were exposed for longer time already. Relation between Methylmercury and total mercury in human hair and fish has been reported [29].

## Conclussions

Contamination status of groups of inhabitants who are living around the ASGM sites (Anggrek and Sumalata) and not (Monano, Kwandang and Tolinggula) are in very high levels according to HBM. Group females have mean hairs concentrationswhich are higher than male, this results showing that female inhabitants are more vulnerable to mercury contamination. Serious health problem indicated by dominant symptoms observed among inhabitants of ASGM are Bluish of gums Babinski reflex, and tremor.

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## Author details

[1]Graduate School of Science & Engineering, Ehime University, 2-5 Bunkyo-cho, Matsuyama, 790-8577 Japan e-mail: yayu_arifin@ung.ac.id

[2]Department of Geology, State University of Gorontalo, Jl. Jend. Sudirman No. 6 Kota Gorontalo, 96128 Indonesia

[3]Cyclotron Research Center, Iwate Medical University, Japan

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