

## Risks Assessment Of Skin Lesion Posed By Arsenic Contaminated Well Water And Rice Consumption From Buyat, North Sulawesi Province, Indonesia

Anwar Daud

Department of Environmental Health, Faculty of Public Health Hasanuddin University, Indonesia

### ABSTRACT

*High arsenic contaminated well water and rice in most of the area in Buyat have been risen a fierce debate among community, local government and mine industry. The occurrence of high arsenic (As) in groundwater and surface water mostly due to a combination of geological processes and/or anthropogenic activities. In Buyat areas which is located in Raratotok regency covered about 200 km<sup>2</sup> and about 130,000 people are living within that ranges. About 90 percent of them drink high-arsenic well water and consumed rice with Arsenic (As.). This sampling survey was conducted to investigate the arsenic level concentration in drinking water from well and rice grain and to characterize arsenic-induced skin lesion. Total water arsenic was measured by flow injection analysis using atomic fluorescence detection with inline photooxidation and continuous hydride generation. Rice arsenic was measured by different parts of rice plant and rice samples were oven-dried, digested with HNO<sub>3</sub>, HCl and perchloric acid, filtered and tested with GFAAS. Then we assess the potential risks posed by community who are living in the area in term of target hazard quotient (THQ). We selected cases (persons with arsenic-induced skin lesions) and control (persons without arsenic-induced skin lesions) from the area concern and at the background sites area for control. Results revealed that, the high level arsenic concentration in drinking water, fish and rice were 0.098 mg liter<sup>-1</sup>,*

*3.867 mg kg<sup>-1</sup>ww and 1.556 mg kg<sup>-1</sup>ww. The average latency for skin lesions was 6 years from first exposure. We found strong dose-response gradients with both peak and average arsenic water concentrations. Conclusions, The lowest peak arsenic ingested by a confirmed case for water, fish and rice were 0.012 mg liter<sup>-1</sup>, 0.011 mg kg<sup>-1</sup>ww and 0.022 mg kg<sup>-1</sup>ww, respectively. Confirmation of case diagnosis and exposure assessment provide the basis for a detailed dose-response evaluation of arsenic-caused skin lesions. We found the highest THQ values in water, fish and rice were 0.010, 0.365 and 0.259, respectively.*

*Key Words: Arsenic contamination, well water, rice, skin lesion, target hazard quotient.*

### I. INTRODUCTION

Trace metal concentration determination in natural water system has received increasing attention for monitoring environmental pollution. This due to the fact that some metals are not biodegradable and their way in food chain through a number of path ways and may accumulate in different organs of human beings or animals.[1] High arsenic levels can also come from certain fertilizers and animal feeding operations. Industrial activities such as copper smelting, mining and coal burning also contribute to arsenic

in our environment.[2] Arsenic is recognized as toxic pollutants. It is used in paints, dyes, metals, drugs, soaps, semiconductors and also is used as wood preservative.

The presence of arsenic in drinking water is difficult to detect without complex analytical techniques. Millions Hundreds of people, mostly in developing countries including Indonesia, daily use drinking water with arsenic concentrations several times higher than the World Health Organization (WHO) recommended limit of 10 millionths of a gram per liter of water (10  $\mu\text{g/L}$ ).[3] Recently several surveys of arsenic in rice grain (788 samples total) have been undertaken, [4],[5],[6] since mining activities increased significantly and may initiate the rise of the arsenic at the superficial.

Arsenic enters aquifers through the dissolution of minerals and ores, resulting in high concentrations in groundwater in some areas. Drinking-water from surface sources does not normally contain high concentrations of arsenic, unless those supplies come from arsenic-contaminated irrigation groundwater. Exposure to inorganic arsenic through the food chain is limited, although absorption by crops irrigated with water highly contaminated with arsenic warrants further research. [3, 7]

Absorption of arsenic through the skin may through washing hands or clothes and bathing in water containing arsenic or working in paddy fields with arsenic-contaminated waters do not pose risks to human health. The guideline for arsenic in drinking water specified by the World Health Organization (WHO) is 10  $\mu\text{g/L}$ , and the permissible level by the Government of Republic of Indonesia is 50  $\mu\text{g/L}$ .

Skin lesions pose an important public health problem because advanced forms of keratoses are painful and debilitating, the subsequent disfigurement can lead to social isolation in the villages.

This case was also occur in Buyat area where some local people suffer from skin lesion.

Arsenic is classified as a Class I human carcinogen by the WHO. The arsenic-induced skin lesions may be associated with increased risks of skin, bladder and lung cancers.[8, 9] Arsenic associated with skin cancer, lung, bladder, kidney, and liver cancer. The skin is the main organ of accumulation of arsenic and chronic exposure produces characteristic changes in pigmentation (hyperpigmentation and hypopigmentation), hyperkeratosis, and after a long exposure causes skin cancer.[10-12]

This research study is first in Indonesia to examine the relationship between arsenic levels in drinking water, fish and rice, with skin disease, hyperkeratosis. We also assess the target Hazard risks posed by people living in the area. The estimated prevalence is likely to increase because people who experience skin lesions have the family members who drink water and consume rice come from the same source.

## 2. MATERIALS AND METHODS

### 2.1 Study Area

This research was conducted in Buyat Area, North Sulawesi 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 from January to March (summer season), followed by West wind are from April to May. Administratively, Buyat area consists of four sub districts, this area is surrounded by the Totok Gulf in the North, Sulawesi Sea in the West, PT. Newmont Mine Area in the uphill in the East and Buyat Village in

the West. Its topography consists of plains at the coastal area and becomes hilly several hundred meters ahead from the shorelines.

PT. Newmont gold mining area itself is located at about 3100 m from the sea and on hill side.

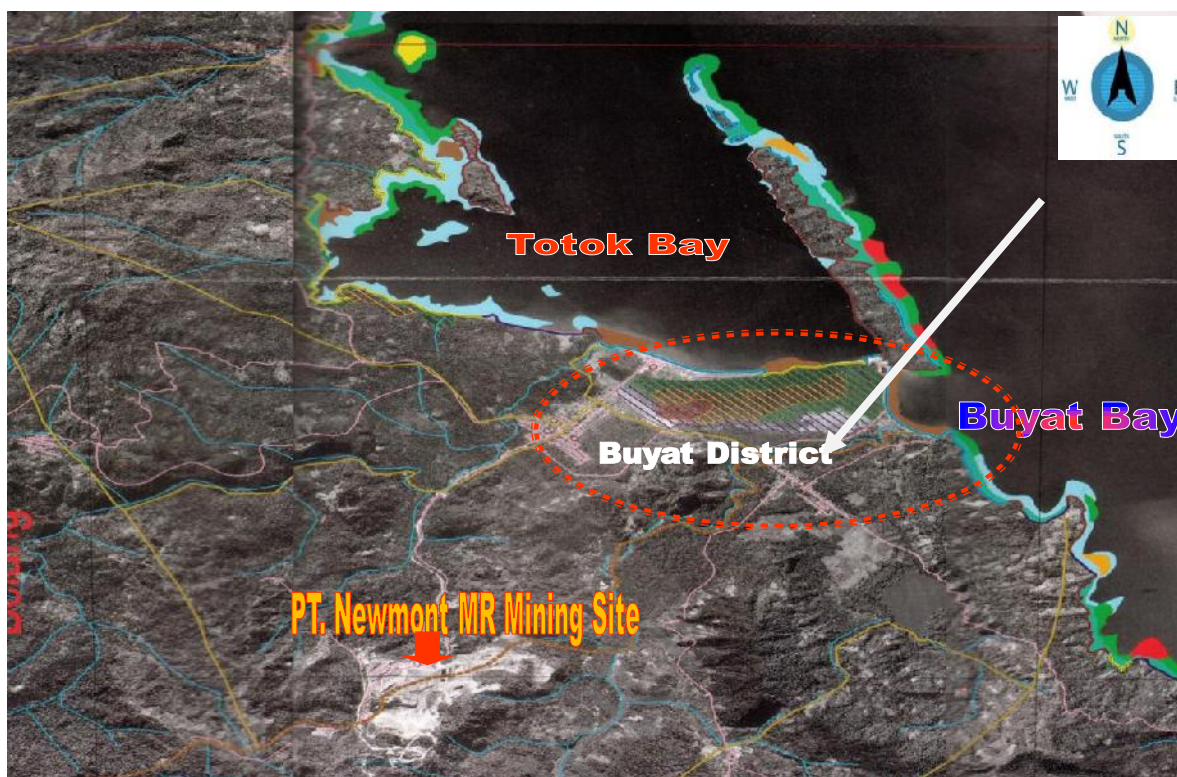


Figure 1. Sampling study in Buyat area

## 2.2 Sampling Methods

This study aimed to investigate arsenic concentration in drinking well water and in rice and to characterize skin lesions at peoples in Buyat Areas, North Sulawesi-Indonesia. Data study was based on survey participants living in and around Buyat areas whose primary drinking- water and rice sources come from local contaminated site. Prior to the respondents' recruitment, shallow well water and rice data were obtained for 15 stations. For blood samples, we took from groups of people who suffering from skin lesion (keratosis or hyperkeratosis) living in Buyat Village (exposure areas) then were frozen at  $-20^{\circ}$  before shipped and analyzed in Laboratory of Institute Environmental Health Technical and Communicable Disease Control (CDC)

in Manado, the procedure was approved by the Ethical Clearance of the Medical Faculty of Hasanuddin University Makassar, Indonesia. Inclusion criteria were age more than 15 years and living at the areas more than 5 years. Arsenic levels were analyzed using AAS-GF (Atomic Absorption Spectrometry-Graphite Furnace) while skin lesion diagnosed by a Dermatologist from Medical Hasanuddin University.

## 2.3 Interviews

We visited the participants in their homes and did face to face interviewed. A physician interviewer who was blind to case or control status administered a structured questionnaire. The questionnaire assessed some following information: lifetime residential history, current and past water

sources at home and work sites, rice source, current and socio economic demographic characteristics as well as body weight scale.

## 2.4 Skin Clinical Exam

All participants underwent a full medical examination conducted according to a written protocol. A careful examination of the skin was conducted in a well-lit area outdoors, under natural light. Visible or palpable dermal lesions were documented noting the location, appearance and whether the patterns were characteristic of arsenic-induced skin toxicity. The interviewer photographed the most highly affected skin areas. Four project physicians later reviewed the slides. After joint review and discussion, the physicians classified the skin lesions (by consensus agreement) as definitely, probably, possibly or not related to arsenic. Dermal changes “definitely” or “probably” induced by arsenic were classified as a current skin lesion.

## 2.6 Water Sample Collection

We collected water samples from all functioning tube or dig wells used by participants in Buyat area. When available, such measurements were incorporated in the data analyses. For 15 closed wells, samples were obtained from the closest tube wells that were of the same depth as the closed well, and located within the same group of houses. We collected samples from approximately 15 functioning tube wells in the Buyat areas. Water samples were stored in a cooler containing an ice block and transported to the laboratory in Makassar, North Sulawesi at the same day. The water samples were then kept frozen at -20°C prior to be analyzed.

## 2.7 Rice Sample Collection

Sampling grains of rice (*Oryza Sativa* L.) were collected once in harvested season in summer, at the close site as collection of drinking well water samples in Buyat District. Non glutinous rice grains were available in all stations.. Rice samples were bagged in polyethylene bags and preserved at 4°C.

## 2.8 Target Hazard Quotient

Consumption of food containing arsenic has been identified as a health risk. The U.S. Environmental Protection Agency (US EPA) and the National Academy of Sciences recommend keeping this corresponds to a reference dose (RfD) of arsenic do not greater than  $5.0 \times 10^{-4} \mu\text{g} \cdot \text{g}^{-1} / \text{day}$ . 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. The method is available in US EPA Region III Risk based Concentration table US EPA, 2000 and it is described by the following equation

$$\text{THQ} = \frac{\text{EF} \times \text{ED} \times \text{FIR} \times \text{C}}{\text{RFD} \times \text{BW} \times \text{AT}} \times 10^{-3}$$

where; THQ is target hazard Quotient; EF is exposure frequency (365 days/year); ED is the exposure duration (70 years); FIR is the rice ingestion rate (100grams /person/day); C is the metal As concentration; RFD is the oral reference dose (As =  $3.0 \times 10^{-4} \mu\text{g} \cdot \text{g}^{-1} / \text{day}$ ); BW is average body weight (70 kg) and AT is averaging exposure time for non-carcinogens (365 days/year x ED)

### 3. RESULTS AND DISCUSSIONS

#### 3.1 Arsenic in Drinking Well Water

Arsenic removal from waters to reach levels in accordance with the regulations for drinking water purposes ( $<10 \mu\text{g L}^{-1}$ , World Health Organization, 2004) is not an easy task. Especially in a rural area like Buyat that economically depends on the natural resources and mining activity. The results of well water showed in Table. 1. The mean arsenic concentration (0.455ppm) in shallow well water was about 10 times higher than the Indonesia maximum permissible limit of 0.05 ppm. Arsenic content in drinking well water samples varied from 0.023 to 0.097 ppm. in Buyat village.

It is reported that shallow aquifer layer is contaminated with arsenic in almost all of the districts of Buyat. In this study, elevated level of arsenic in drinking well

water was found as the primary source of contamination. Arsenic dissolved in water can be easily bio-accumulated in algae, fish and shellfish via different trophic pathways and consequently to the human food chain. The risk of As-contamination in the human food chain has recently received much public attention because of the potential human health hazard people can be exposed to As from various sources (such as food, water, and air), but exposure through ingestion of fish and seafood can be significant.[13, 14]

Study in Mexico revealed As concentration in water, soil, forage and milk in Comarca Lagunera, where As concentrations were determined in 73 well water samples and 50 soil, forage and milk samples. The i-As concentration in well water ranged between 7 and 740  $\mu\text{g/L}$  [ $\sim 90\%$  As (V) species]. The soils contained t-As of up to 30 mg/kg.[15]

**Table 1.** Arsenic concentration and accumulation in water column and Rice grain from Buyat Area North Sulawesi Province, Indonesia.

Station	Location	Drinking well water ( $\text{mg L}^{-1}$ )	Fish ( $\text{mg kg}^{-1}\text{ww}$ )	Rice Grain ( $\text{mg kg}^{-1}\text{ww}$ )
1	Upstream, about 10 km from St.7	0.045	0.011	0.213
2	Upstream, 8 km from St.7	0.068	0.015	0.456
3	Upstream, 6 km from St.7	0.033	0.098	0.241
4	Close to river mouth in the west	0.082	0.125	0.988
5	At river mouth in the West	0.026	0.452	0.543
6	At the community Housing	0.071	2.435	1.022
7	At the river mouth in the North	0.098	2.876	0.067
8	Close to river mouth in the North	0.085	1.089	0.874
9	Downstream, 6 km from St.7	0.012	2.033	1.556
10	Downstream , 8 km from St.7	0.045	3.867	1.018
11	Downstream, 9.km from St.7	0.065	1.563	0.675
12	Downstream , 10 km from St.7	0.078	1.524	0.452
13	Downstream , 11 km from St.7	0.092	2.083	0.865
14	Background site	0.087	1.002	0.145
15	Background site	0.023	1.232	0.653

Another survey by Sompongchaiyakul and Sirinawin, [16] reported that As concentrations in sediment from Thale Noi, Inner and Middle Lake, and Outer Lake were  $8.2 \pm 1.7$  (5.7-10.8),  $5.9 \pm 1.5$  (3.7-10.8) and  $10.7 \pm 5.5$  (5.1-25.7) mg kil, respectively. Pradit et al, [17] took the bottom sediments from 44 stations in Songkhla Lake to study the trace element contaminations in sediments. The trace element concentrations in surface sediment ranged from 0.8- 70.7 llg g-1 dry wt for As, 0.1 - 2.4 ~tg g-1 dry wt for Cd and 8.2-131 llg g-1 dry wt for Pb.

Arsenic levels in ground water (water supply) in Vietnam about 0.9 to 321 ppb.[18] There are 27% of wells exceeded the WHO standard, which is 10 ppb.[19] Rice with arsenic levels of 1.8 mg/kg has been recorded in the arsenic-affected tube well areas of Bangladesh.[6]

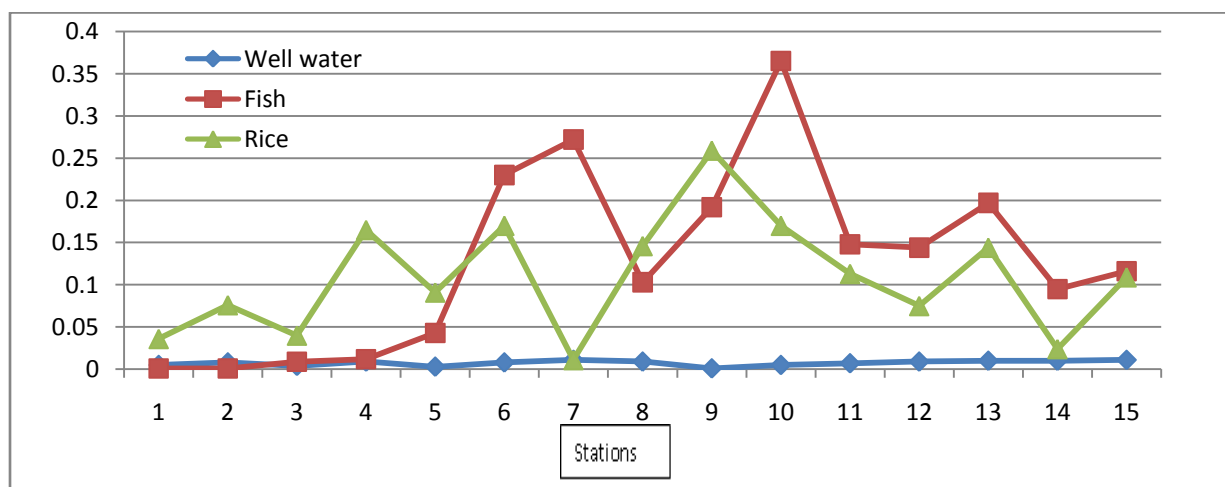
### 3.2 Arsenic in Rice Grain

In order to assess the spread of arsenic from ground water to the biota, some common crops and vegetables grown in the studied areas were analyzed for their arsenic content. Paddy is the most important amongst the crops grown in Indonesia.

Paddy grown on the soils adjacent to contaminated water source is analyzed for total arsenic. The elevated level of As in rice were in three station that were  $>1$  mg kg<sup>-1</sup>ww range from 1.018, 1022 and 1.556 mgkg<sup>-1</sup>ww, respectively.

### 3.3 Target Hazard Quotient

In this study, it is experimentally proved that drinking well water and rice in Buyat area is contaminated with arsenic and acts as the primary source of arsenic poisoning among the community. The results indicate that human population is affected with arsenic locally using the contaminated well water and rice for a long time. From that ground water source, top soils are also locally contaminated with arsenic leading to a significant accumulation in some plants especially rice that are commonly grown. Paddy grown on the soil that was continuously wetted nearby-contaminated irrigation water showed higher levels of arsenic content. The highest levels of THQ arsenic value was found still  $>1$ , mean not risk for water, fish and rice consumption for the whole life time.



**Figure 1.** Target Hazard Quotient (THQ) of Arsenic concentration and accumulation via water, rice and fish consumption from Buyat Area, North Sulawesi, Indonesia.

#### 4. CONCLUSION

Arsenic contamination of drinking well water and rice over substantial areas of North Sulawesi especially in Buyat areas now exists. Water and soils will stay contaminated as a result plants are also contaminated, as it is not cost effective to remediate water and soils, especially in paddy fields. The only option is to breed rice with low levels of arsenic in grain.

To conclude, we suggest that more specific recommendations regarding human consumption such as kind of species, size of species, frequency, sizes of meals are done according to the data concerning levels of environmental pollutants in the most consumed drinking water and paddy.

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