Arsenic Levels in Soil and Rice and Health Risk Assessment via Rice Consumption in Industrial Areas of East Java, Indonesia

Nurul Laela[1], Satriani Aga Pasma[1*], and Muhayatun Santoso[2]

[1]*Medical Intelligence Postgraduate Program, State Intelligence College, Bogor, Indonesia*
[2]*Research Center for Radiation Detection and Nuclear Analysis, Nuclear Energy Research Organization, National Research and Innovation Agency, Bandung, Indonesia*

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*** Corresponding author:**
E-mail:
satriani.aga.pasma@stin.ac.id

ABSTRACT

Industrial use of arsenic can potentially cause environmental problems in water, soil, and air. Arsenic is one of heavy metals that is highly toxic and carcinogenic. Arsenic contamination in the environment is harmful to human health because it can enter the body through the food chain. This study determined the concentration of arsenic in soil and rice and its impact on human health risks. Sampling was carried out in several East Java industrial cities or districts, for instance, Gresik, Mojokerto, Sidoarjo, Nganjuk, Ponorogo, and Surabaya. The measurement of arsenic in soil was done using Energy Dispersive X-Ray fluorescence (EDXRF), while the measurement of arsenic in rice was done by Total X-Ray Fluorescence (TXRF). The results showed that arsenic concentration in several areas of East Java has varying levels. The concentration of arsenic in soil was highest in Gresik (13,786 mg/kg). The highest arsenic concentration in rice was found in Mojokerto (0.154 mg/kg). The results of risk assessment in this study showed that the Hazard Quotient (HQ) value was >1 and the Excess Cancer Risk (ECR) was $>10^{-4}$ in all areas at the age of children <2 years. Health risk assessment of adults showed HQ>1 and ECR$>10^{-4}$ in several areas of East Java. This indicates that consumption of rice contaminated with arsenic has the potential to pose non-carcinogenic and carcinogenic health risks.

1. INTRODUCTION

Indonesia, as one of the developing countries, has experienced massive growth in the industrial sector. The Ministry of Industry (2017) stated that Indonesia is included in the top five countries with a fairly high industrial contribution. Nevertheless, industry also has negative impacts on the escalation of environmental pollution, whether in the water, soil, or air. This is due to the fact that high industrial activity has the potential to produce hazardous waste that can damage the environment and ecosystems. One of the hazardous industrial wastes that currently concerns the world is arsenic. Arsenic is a metal that is widely used in industrial activities; for instance, in the paint industry, ore processing and mining (Andhani and Husaini, 2017).

The contamination of the environment with arsenic becomes an important issue for global health because arsenic can be harmful to human health, especially for children. Arsenic is a non-essential heavy metal that is highly toxic and carcinogenic (Anetor et al., 2007). Arsenic exposure in children is more vulnerable than in adults, this is because children's weight is lower than adults, so their intake level is higher. In addition, children have organs that are still developing, so metabolism for elimination is still lacking compared to adults (Ferguson et al., 2018). Gardner et al. (2013) showed that arsenic exposure caused poor growth in children. In a study in Bangladesh, Wasserman et al. (2004) showed that there is a strong relationship of high concentrations of arsenic in the urine of children with low intellectual function.

In groundwater, arsenic exists in two forms, namely aerobic and anaerobic. Arsenic in the anaerobic form is reduced arsenic, also called arsenite (ASIII). Arsenite is fat-soluble and can be absorbed, by the body, through the digestive tract, respiratory tract, or skin. In contrast, arsenic, in its aerobic form,

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is oxidized arsenic, also called arsenate (ASV) (Majmuder et al., 2019). Arsenic contamination in the soil is the primary source of contamination of water or food. The results of Hamzah and Hapsari (2017) research proved that arsenic content in Batu City paddy soil exceeded the threshold value, with a concentration of 0.89 ppm. The studies of Komarawidjaja (2017) showed that there was arsenic contamination that exceeded the threshold in the soil in the paddy fields of Jelegong Village, Rancaekek, Bandung (4.0 mg/L). Hazardous arsenic contamination in the soil can be harmful to human health because it can enter the body through the food chain. The nature of heavy metals makes them difficult to decompose, and deposits on the soil surface can be absorbed by organisms. This process is known as biomagnification, which is an increase in heavy metal contamination in the tissues of organisms through the food chain (Hidayah et al., 2014).

Arsenic can be easily accumulated in all types of cereals, especially rice, because of its high bioavailability in the soil (Huang et al., 2013). Rice is a major food in Asian countries and it's the main staple food source for Indonesian people. Several studies have shown high levels of arsenic in rice. It was reported that the levels of arsenic in several countries are 0.257 mg/kg (American rice), 0.188 mg/kg (Australian rice), 0.183 mg/kg (France rice) 0.147 mg/kg (Paksitani rice), and 0.103 mg/kg (Indian rice) (Shraim, 2017). Meanwhile, arsenic levels in several districts of Indonesia were reported to be 0.33 mg/kg in Medan (Ginting et al., 2018) and 1.76 mg/kg in Yogyakarta (Alfrianti, 2019). High concentrations of arsenic in rice can potentially be a major source of arsenic exposure, especially in countries that have rice diets.

It is crucial for East Java Indonesia to identify arsenic levels in soil and rice. Based on data from SI (2020), East Java is the province that has the largest contribution to rice production in Indonesia, producing around 9,944,538.26 tons of GKG or the equivalent of 5,712,597.01 tons of rice. On the other hand, East Java is also one of the industrial center provinces with 6,746 large and medium industries, and 92,031 micro industries (SI, 2019). The large rice production and high industrial activity in East Java can cause a potential hazard of arsenic contamination from industrial activities, which will spread into the environment and accumulate in the rice consumed by society. In addition, the existence of potential health hazards to children cannot be ignored, so it is necessary to analyze the potential risks of arsenic exposure to children's health.

The analysis of the arsenic content was carried out using the X-Ray Fluorescence (XRF) method. The method used for analysis of arsenic in soil was Energy Dispersive X-ray Fluorescence (EDXRF). This method is an analytical method that can measure elemental content from low atomic number to high atomic number, from the range of % to ppm, the method is fast, sensitive, and the equipment is easy to operate (Kurniawati et al., 2014). The method used for the analysis of arsenic in rice is Total X-Ray Fluorescence (TXRF). This method is a simple procedure, has good capability in measuring samples in very small concentrations in nanograms or micrograms, high sensitivity, and low detection limits in the order of ppb (Gruber et al., 2020).

2. METHODOLOGY

2.1 Materials

In this study, measurement of the arsenic in soil used the Energy Dispersive X-ray Fluorescence (EDXRF) spectrometer MiniPal 4 (PANalytical). Measurement of arsenic in rice used Total X-ray Fluorescence (TXRF) spectrometer S4 T-STAR (Bruker). Other equipment used in this study includes an analytical balance type 2842 (Sartorius), titan-eyed blender, hot plate (SI Analitycal), desiccator, Memmert oven, mortar, American standards testing stainless steel and material (ASTM) sieve with sizes of 100 mesh and 200 mesh, ultrasonic Elma 37 KHz, and other supporting equipment. The materials needed in this study included soil samples, rice samples, Standard Reference Material National Institute of Standards and Technology (SRM NIST) 2711a Montana Soil, SRM NIST 1568a Wheat Flour, demineralized water, standard Ga solution, triton X-100 solution, quartz glass, and other common materials.

2.2 Sampling of soil and rice

In this study, the sampling method was conducted by purposive sampling, in which the samples were obtained from agricultural areas around industrial activity. Soil and rice samples were collected from six cities/district in East Java, Indonesia. They were from several industrial area of Gresik (S07.174498°; E112.537473°), Mojokerto (S07.460749°; E112.469620°), Sidoarjo (S07.383-4359°; E112.6375614°), Nganjuk (S07.5857786; E112.5933989°), Ponorogo (S07.56177°; E112.26-463°), and Surabaya (S07.247968°; E112.651635°), shown in Figure 1. Soil samples were obtained from paddy field by randomly taking soils of depths 0-10 cm (surface) and 10-30 cm (subsurface) from three spots and mixed to give representative samples. Soil samples taken at each sampling point were approximately 1 kg. Meanwhile, rice samples were taken by collecting rice yields in the soil sampling area. Rice samples were taken from direct agricultural products at each location point and about 1 kg samples were collected. The type of the tested rice was white rice (*Oryza sativa* L).

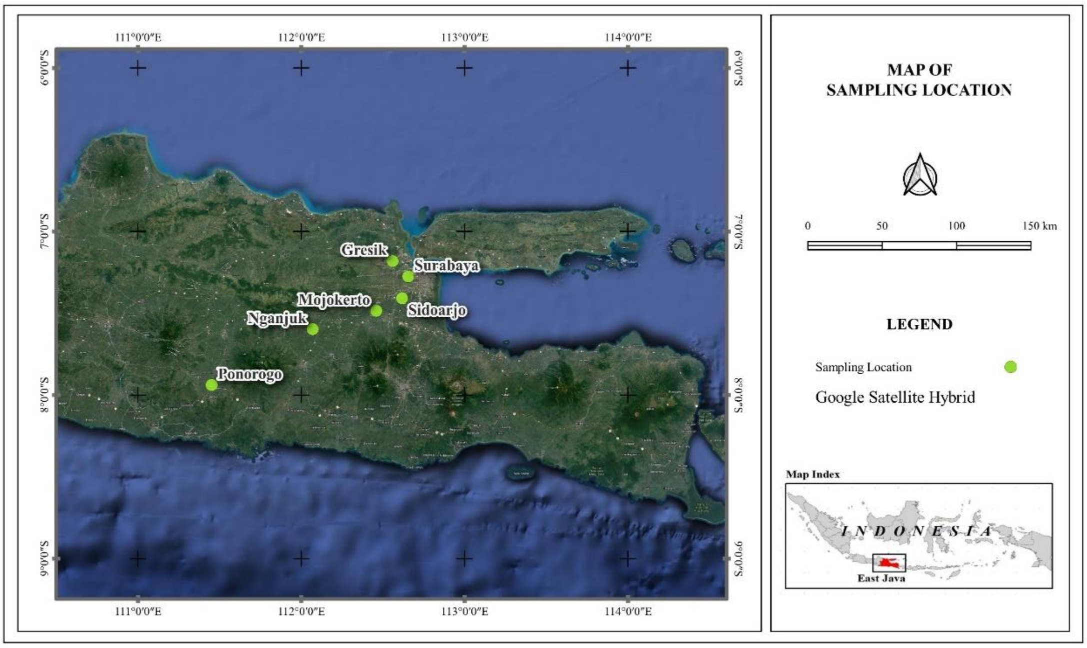

Figure 1. Sampling location map

2.3 Sample preparation and analysis

2.3.1 Preparation

Sample and standard preparation were carried out according to Adventini et al. (2016) and Syahfitri (2021) with several modifications. Paddy soil samples that had been obtained were sun-dried for 5-7 days. Then, the soil sample was homogenized with a mortar and filtered through a 200 mesh sieve. The soil sample was put into plastic and labeled according to the location point. The standard used for validating the EDXRF MiniPal 4 method for soil measurement was the SRM NIST 2711a Montana Soil.

Soil samples and standards were weighed at ±1 g, then placed into the sample holder cup, which was covered with mylar plastic. Soil samples and standards were measured using EDXRF spectrometer MiniPal 4. Meanwhile, the rice samples were mashed using a titan-eyed blender and dried in an oven at 105°C, and then the water content was calculated. Rice samples with a moisture content below 14% were homogenized using a 100-mesh sieve. About 200 mg of the rice sample was put into a corning tube, then 5 mL of triton X-100 solution and 10 µL of 1,000 ppm Ga standard were added. Then the sample was vortexed and incubated in an ultrasonic incubator for five minutes; then, 10 µL of the sample was pipetted onto quartz. Rice samples were analyzed using the TXRF spectrometer.

2.3.1 Measurement

Soil samples were placed in a sample holder cup and loaded into the EDXRF spectrometer MiniPal 4. Then, soil samples were irradiated with an X-Ray generated from the Rhodium (Rh) tube in the tool using soil sediment application software with optimum conditions: voltage 30 kV, current 150 uA, measurement time of 300 sec, Al filter, and air media. The rice samples were irradiated using a TXRF spectrometer device with TPPA Mo-K and TPPA W-Brem applications. The optimum measurement conditions of arsenic were as follows: voltage 50 kV, current 1,000 uA, and measurement time 1,000 sec. The measurement results were in the form of intensity, while the concentration elements were in the sample obtained by comparing the intensity of the sample with the standard based on formula (1) as follows:

$$C_{spl} = \frac{I_{spl}}{I_{std}} \times C_{std} \quad (1)$$

Where; C_{spl} is the concentration of the element in the sample, I_{spl} is the intensity of the sample, I_{std} is the intensity of the standard, and C_{std} is the concentration of the element in the standard.

2.4 Health risk assessment

In this study, the health risk assessment of arsenic focused only on the rice exposure assessment and risk characterization. This risk assessment aims to determine the average daily intake (ADI), hazard quotient (HQ), and excess cancer risk (ECR). The ADI value is used to determine the exposure dose received by the body through food so that arsenic intake from contaminated rice can be determined with the following equation:

$$ADI = \frac{C \times IR}{BM} \quad (2)$$

Where; ADI is the Average Daily Intake (mg/kg/day), C is the Concentration of heavy metals in rice, IR is the Ingestion Rate, and BM is Body Mass. The total rice consumption in East Java is 88 kg/capita/year or equal to 0.241 kg/capita/day (MCITI, 2016). The risk assessment in this study was conducted on children aged 6-8 months, 9-12 months, and 13-24 months. The average body weight is 7.6 kg for children aged 6-8 months, 8.4 kg for children aged 9-12 months, 11.9 kg for children aged 13-24 months, and 60 kg for adults (Suyanto et al., 2021; Adventini et al., 2016).

The Hazard Quotient (HQ) value is needed to determine the potential health risk of non-carcinogenic (non-cancerous) contaminants. If the value of HQ<1, then the potential health risk is low, and it can be said that the pollution which occurs is still within safe limits. If HQ>1, then the potential health risk is high. It needs to be controlled. The HQ value is obtained by the following equation:

$$HQ = \frac{ADI}{RfD} \quad (3)$$

Where; ADI is the daily intake of heavy metals, and the RfD is the estimated maximum daily dose intake allowed.

The level of risk of carcinogenic effects is expressed in Excess Cancer Risk (ECR). The ECR value determines an individual's lifetime estimate of cancer risk. If the ECR value is >10-4, it is at risk of causing cancer. The ECR value is obtained by payment as follows:

$$ECR = ADI \times SF \quad (4)$$

Where; ADI is the average daily intake of heavy metals and SF is the cancer slope factor.

3. RESULTS AND DISCUSSION

3.1 Validation result

In this study, method validation tests were used to confirm the test results quality by using Standard Reference Materials (SRM). Furthermore, soil sample testing was validated with EDXRF spectrometer MiniPal 4 using SRM NIST 2711a Montana Soil. Meanwhile, the rice samples testing was validated with TXRF spectrometer using SRM NIST 1568b Rice Flour. The results of the SRM NIST 2711a Montana Soil validation compared to the certificate value are shown in Table 1. The results of the SRM NIST 1568 b Rice Flour validation are shown in Table 2.

Table 1. The validation results on SRM NIST 2711a Montana Soil

Element	Certificate value (mg/kg)	Analysis value (average)	Recovery (%)
Arsenic	107±5	107	100

Table 2. The validation results on SRM NIST 1568b Rice Flour

Element	Certificate value (mg/kg)	Analysis value (average)	Recovery (%)
Arsenic	0.29±0.03	0.27	95

The validation results in Table 1 and Table 2 show that the percentage accuracy value (%) on SRM NIST 2711a Montana Soil is 100%, and the percentage accuracy value (%) on SRM NIST 1568b Rice Flour is 95%. This result follows the acceptability limit of the AOAC (2002), in the range of 85-110% for SRM NIST 2711a Montana Soil and 75-120% for SRM NIST 1568b Rice Flour. The validation results also show a good relationship between the measurement results and the certificate value. Thus, the test method is valid and reliable for testing soil and rice samples.

3.2 Arsenic concentration in soil

Arsenic concentration in the soil in several cities/districts of East Java, Indonesia, is shown in Figure 2. Arsenic concentration was analyzed in the soil at two depths, i.e., 0-10 cm and 10-30 cm depth.

The analysis showed no significant difference between arsenic levels in 0-10 cm and 10-30 cm depth, the concentration ratio between the two depths is 1. Arsenic in rice fields in six regencies of East Java was found at high levels, and variation ranged from 11,940-13,786 mg/kg.

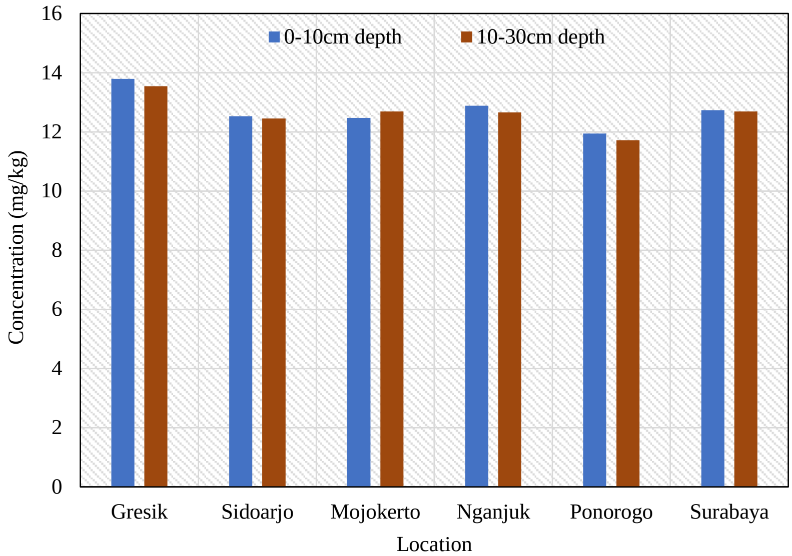

Figure 2. Arsenic concentration in soil

The variability of arsenic concentration in soil differs from region to region, as shown in Figure 3. Arsenic level was higher in industrial area of Gresik were 13,786 mg/kg, followed by Nganjuk (12.884 mg/kg), Surabaya (12.729 mg/kg), Sidoarjo (12.522 mg/kg), Mojokerto (12.474 mg/kg), and Ponorogo (11.940 mg/kg). The ranking order of arsenic level from soil was Gresik > Nganjuk > Surabaya > Sidoarjo > Mojokerto > Ponorogo. Furthermore, Figure 4 shows the comparison results between arsenic levels in this study and the threshold values allowed by World Health Organization (WHO), Food and Agriculture Organization (FAO), and European Union (EU). The results showed that the concentration of arsenic in the soil was above the maximum threshold value of 5 mg/kg (Toth et al., 2016).

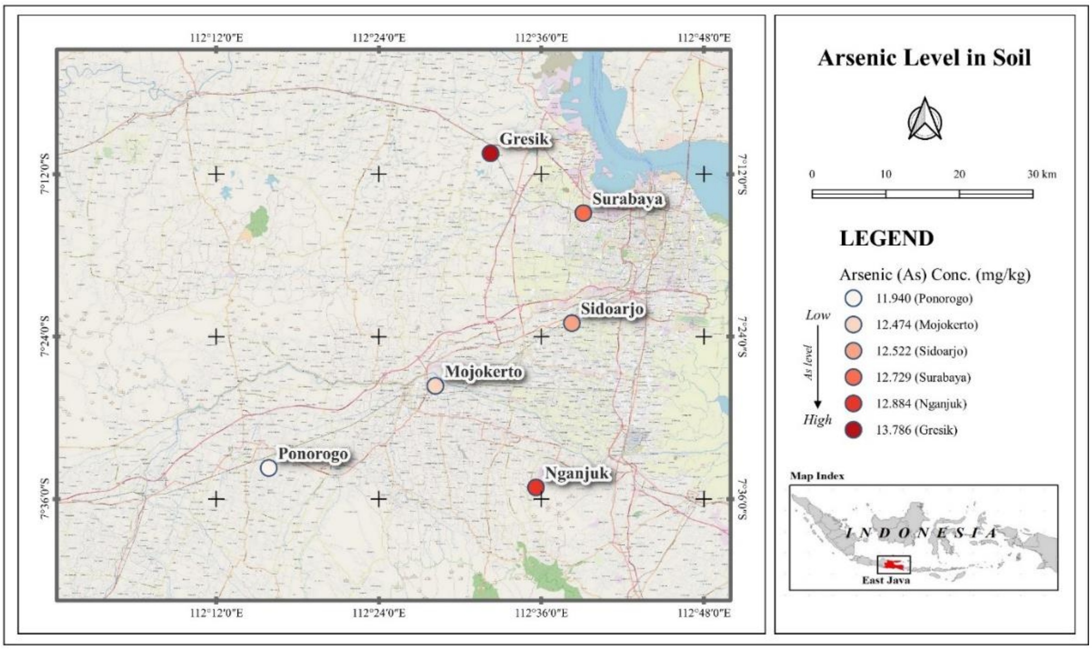

Figure 3. The spatial representation of arsenic levels in soil

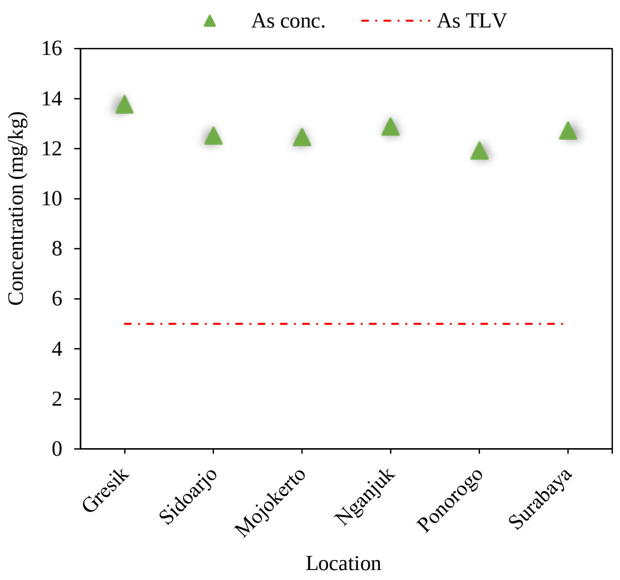

Figure 4. Arsenic concentration in soil with threshold value

Arsenic concentrations in the soil in some areas of East Java have varying levels from one region to another region. The variability of arsenic is not only diverse in the locations of this study but also gives different results from other studies, as shown in Table 3. Arsenic levels in this study were higher than arsenic levels in studies in Mainland China at 10.7 mg/kg (Huang et al., 2019), Thailand at 7.5 mg/kg (Zarcinas et al., 2004b), and Southern Europe at 10 mg/kg (Reimann and de Caritat, 2012). However, there are also research results showing that arsenic levels in this study are lower than those in Malaysia, which were 16.8 mg/kg (Zarcinas et al., 2004a), England and Wales, which were 20 mg/kg (Rawlins et al., 2012), and China's Xunyang, which were 72 mg/kg (Wang et al., 2019). The variability of arsenic levels can be influenced by various factors, such as geological factors and human activities, which are sources of anthropogenic contamination (Zeng et al., 2015).

3.3 Arsenic concentration in rice

Arsenic in the soil can be accumulated into the rice through a process called biomagnification. The biomagnification process is the occurrence of increased heavy metal contamination in organism tissues through the food chain (Hidayah et al., 2014). The result of this study showed that arsenic concentration in rice also varied from one region to another region. Figure 5 shows the analysis of arsenic concentrations in rice in six regions of East Java, ranged from 0.023-0.154 mg/kg.

Table 3. Variability of arsenic levels in soil

Location	Depth (cm)	As concentration (mg/kg)	Reference
Gresik	0-10	13.79	In this study
	10-30	13.55	In this study
Sidoarjo	0-10	12.52	In this study
	10-30	12.45	In this study
Mojokerto	0-10	12.47	In this study
	10-30	12.69	In this study
Jombang	0-10	11.58	In this study
	10-30	11.98	In this study
Nganjuk	0-10	12.88	In this study
	10-30	12.65	In this study
Ponorogo	0-10	11.94	In this study
	10-30	11.72	In this study
Surabaya	0-10	12.73	In this study
	10-30	12.69	In this study
Shandong, China	0-20	13.38	Jia et al. (2010)
Mainland, China	0-20	10.7	Huang et al. (2019)
Peninsular Malaysia	0-15	16.8	Zarcinas et al. (2004a)
Thailand	0-15	7.5	Zarcinas et al. (2004b)
England dan Wales	0-15	20	Rawlins et al. (2012)
Xunyang, China	0-20	72	Wang et al. (2019)
Southern Europe	0-20	10	Reimann and de Caritat (2012)

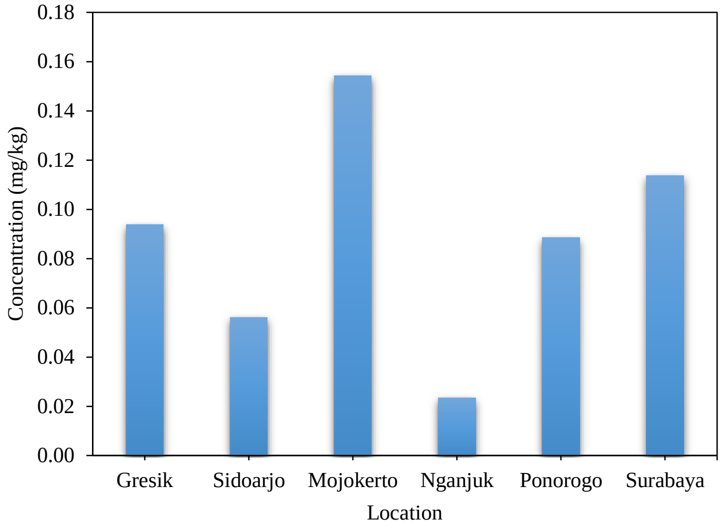

Figure 5. Arsenic concentration in rice

Figure 6 shows a representation of arsenic levels in each region. The highest levels of arsenic are found in the industrial area of Mojokerto, which were 0.154 mg/kg, and the lowest arsenic levels are in Nganjuk, which were 0.023 mg/kg. Arsenic levels in order from highest to lowest were Mojokerto > Gresik > Surabaya > Ponorogo > Sidoarjo > Nganjuk. While Figure 7 shows a comparison of the arsenic levels obtained in this study with the threshold value. The concentration of arsenic in this study was above the maximum value of the National Agency of Drug and Food Control Indonesia (0.1 mg/kg) in Mojokerto and Surabaya.

In comparison, the highest concentration arsenic in soil did not correlate with the highest concentration in rice. The results showed that the concentration of arsenic in several cities/districts was high, but less in rice. This could be due to the absorption or accumulation of arsenic in the rice plant. Abedin et al. (2002) observed a higher accumulation of arsenic in the roots than in any other parts of the plant. This has the effect of lowering the arsenic concentration in grain rice. In this study, the uptake of arsenic by rice plants might be different from one region to another.

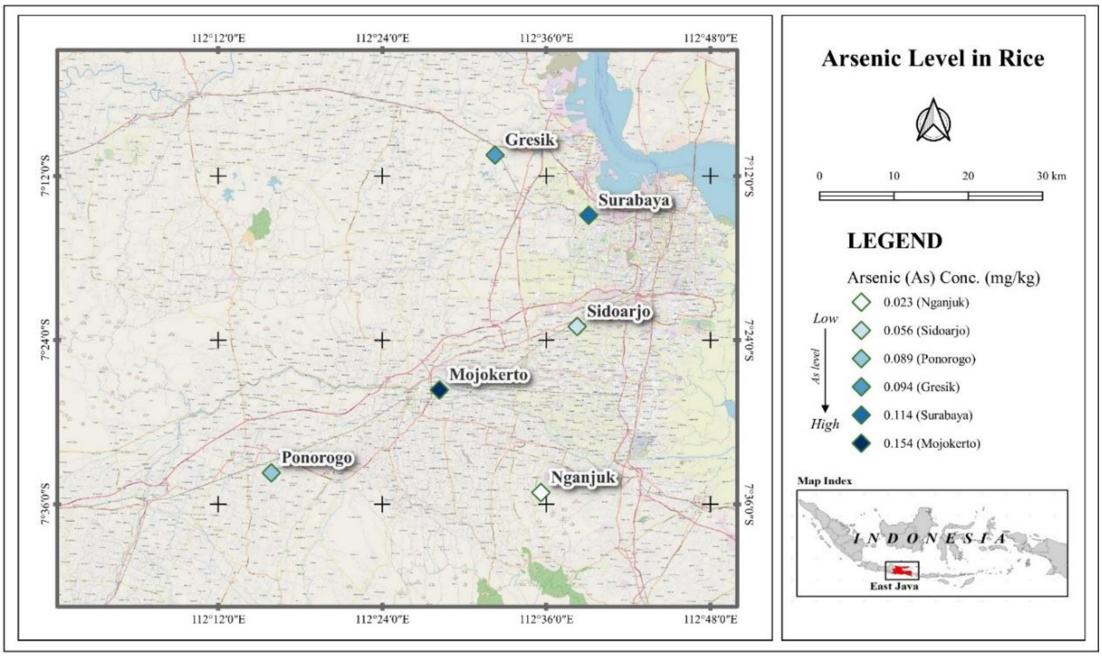

Figure 6. The spatial representation of arsenic levels in rice

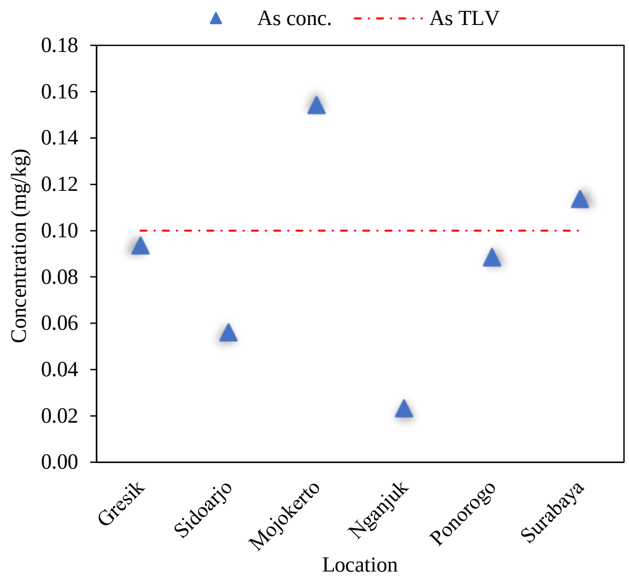

Figure 7. Arsenic concentration in rice with threshold value

The variability of arsenic concentration in rice in this study and other studies is shown in Table 4. Arsenic levels in this study were in the same order as the results of studies in Iran, at 0.12 mg/kg (Rastmanesh et al., 2022), and Savar Bangladesh, at 0.075 mg/kg (Hasan et al., 2022). However, the results of arsenic levels in this study were lower compared to 0.33 mg/kg in Medan (Ginting et al., 2018), 1.79 mg/kg in Yogyakarta (Alfrianti, 2019), 0.224 mg/kg in Matlab Bangladesh (Azmy, 2020), and 0.23 mg/kg in Jiangsu China (Li et al., 2018). Variations in arsenic levels from one region to another region can be caused by various factors, such as the mineral composition of soil, the use of fertilizers, chemical content in the soil, weather conditions during growth, soil pH, rice type, and soil interaction with plant root microbes, that play important roles in regulating movement from soil to plant (Damastuti et al., 2020).

Table 4. Variability of arsenic levels in rice

Location	As concentration (mg/kg)	Reference
Gresik	0.09	In this study
Sidoarjo	0.06	In this study
Mojokerto	0.15	In this study
Nganjuk	0.02	In this study
Ponorogo	0.09	In this study
Surabaya	0.11	In this study
Medan	0.33	Ginting et al. (2018)
Yogyakarta	1.79	Alfrianti (2019)
Iran	0.12	Rastmanesh et al. (2022)
Savar, Bangladesh	0.075	Hasan et al. (2022)
Matlab, Bangladesh	0.224	Azmy (2020)
Jiangsu, China	0.23	Li et al. (2018)

3.4 Human health assessment

The potential health risks caused by arsenic exposure can be determined from the average daily intake of arsenic in the body. The daily intake of arsenic was determined based on the average daily intake of rice per capita/day for the East Java population. The results of study in Figure 8 show that the daily intake of children aged 6-8 months ranged from 0.0007-0.049 mg/kg BW/day, children aged 9-12 months ranged from 0.0007-0.0044 mg/kg BW/day, children aged 13-24 months ranged from 0.0006-0.0049 mg/kg BW/day, and adults ranged from 0.0001-0.0006 mg/kg BW/day. The region with the highest average daily intake of arsenic for all age ranges for children and adults is the Mojokero Region.

Potential non-carcinogenic health risk can be identified from average daily intake value using the Hazard Quotient (HQ) and potential carcinogenic health risks using the Excess Cancer Risk (ECR). HQ results can be seen in Figure 9. These results show that the potential risk of non-carcinogenic exposure for children with HQ>1 ranged from 2.477-16.315 (age 6-8 months), 2.241-14.761 (age 9-12 months), and 1.901-12.525 (age 13-24 months). It shows that at the age of under two years, consuming rice contaminated with arsenic potentially causes non-carcinogenic health effects. While the HQ value in adulthood was found in four areas (Gresik, Mojokerto, Ponorogo, and Surabaya) with HQ>1 ranging from 1.214-2.115, and two areas (Sidoarjo and Nganjuk) with HQ value <1, the HQ value for children is much higher than for adults.

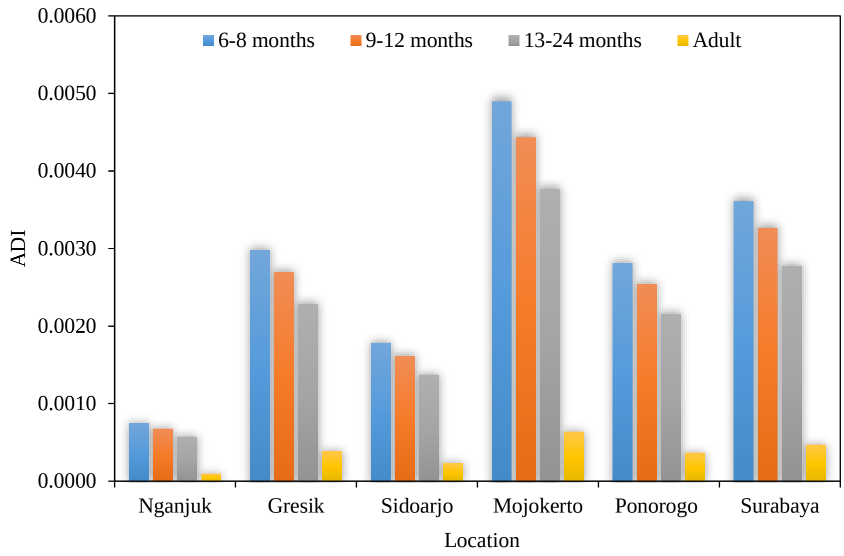

Figure 8. Average daily intake of arsenic

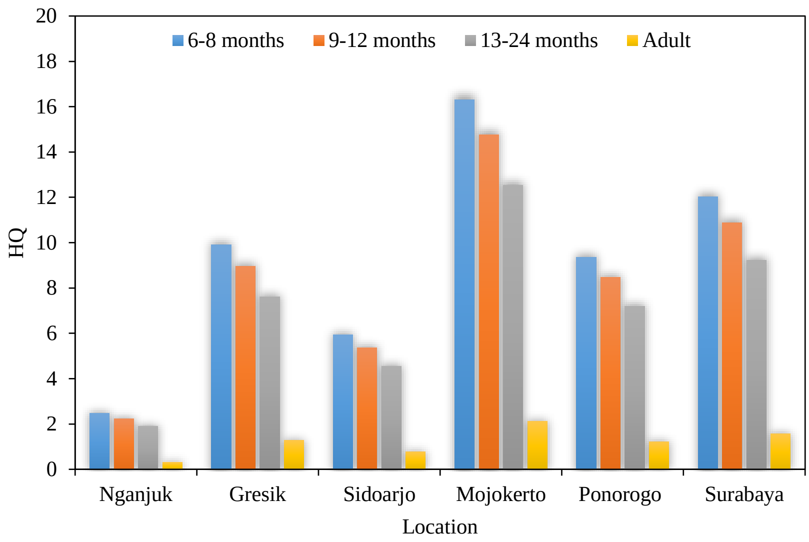

Figure 9. Hazard quotient value in each location

The potential risk of arsenic to carcinogenic health can be seen in Figure 10. The analysis results show that the average daily intake of arsenic from rice consumption in all age ranges of children in all regions has an ECR > 10^{-4}, ranging from 10^{-3}-7×10^{-3} (age 6-8 months), 10^{-3}-6×10^{-3} (age 9-12 months), and 10^{-3}-5×10^{-3} (age 13-24 months). This means that the consumption of rice contaminated with arsenic can pose a cancer risk. The ECR values in adults in some areas of East Java were ECR > 10^{-4}, except in Nganjuk, where they ranged from 2×10^{-4} to 4×10^{-4}. It shows that consuming contaminated rice in the Gresik, Sidoarjo, Mojokerto, Ponorogo, and Surabaya regions poses a cancer risk to adults.

When comparing the HQ and ECR values in adults and children, both values in children are much higher. This shows that the potential risk of arsenic to children's health is much higher than in adults. Children have a higher potential risk because they have smaller bodies with a large amount of rice consumption, while adults have large bodies. As a result, children are exposed to more arsenic than adults. In addition, children are also more sensitive to the harzardous effects of arsenic because their bodies are still developing, so they do not have the mature body system to get rid of harmful chemicals like adults. Arsenic exposure in children continuously and from time to time can cause growth problems, decreased IQ, impaired brain development, an unhealthy immune system, and the development of cancer as an adult (Murray, 2022).

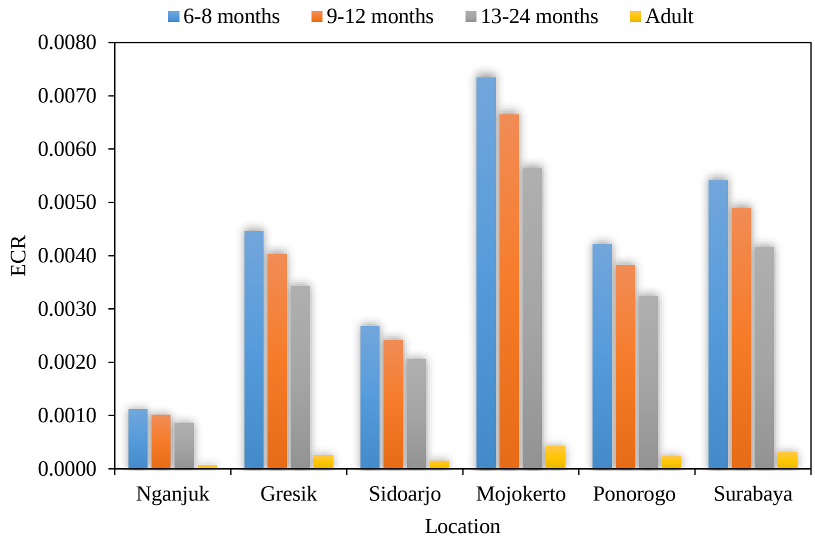

Figure 10. Excess cancer risk value in each location

4. CONCLUSION

In this study, the concentration of arsenic in several industrial areas of East Java had varying levels from one region to another. The highest arsenic level in soil was found in Gresik. The highest level in rice was found in Mojokerto. The concentration of arsenic in the soil exceeded the threshold value in all study areas. Meanwhile, the concentration of arsenic in rice exceeding the threshold value was only found in two regions, namely Mojokerto and Surabaya. Based on the results of this study, arsenic exposure from rice consumption has the potential to pose a health risk to children, both non-carcinogenic and carcinogenic health. Overall, this study provides information about the profile of arsenic concentration in several industrial areas of East Java. These results could be an early warning for local governments to take preventive measures and could also be applied to evaluate the surrounding industrial areas to minimize the hazardous potential of arsenic.

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