

HEALTH RISK ASSESSMENT DUE TO THE METALS EXPOSURE OF GROUNDWATER IN THE AREA OF SIDOARJO MUD (LUSI), EAST JAVA, INDONESIA

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ABSTRACT

Lumpur Sidoarjo (LUSI) (Sidoarjo mud), which has its center in Porong, Sidoarjo, East Java, has had a major impact on environmental conditions, especially on water quality. Our previous analysis proves that the groundwater in this area has been contaminated, especially by heavy metals. In this case, the presence of contaminants from Fe, Mn, and Hg was detected. Heavy metals in this water will certainly affect human health if this contaminated water is consumed, considering that groundwater is the main water resource utilized by local people in this area. Thus, the objective of this study is assessment of health risk from exposure to Fe, Mn, Zn, As, and Hg in the groundwater near to LUSI. The calculation method used the formula recommended by the United States Environmental Protection Agency (US EPA). Scenario subjects for the calculation included 1) children aged 1-3 years, 2) children aged 4-8, 3) children aged 9-12, 4) career women, and 5) career men. The calculation results show that a Hazard Quotient (HQ) value of more than 1 has been detected in several samples and subjects. The highest HI value is in sample B5, and the smallest is in sample B2. In addition, the calculations show that there is a risk of cancer effects from Hg-contaminated water.

Keywords: *exposure, health, heavy metal, risk, water*

1. INTRODUCTION

Sidoarjo is a district that administratively located in province of East Java, Indonesia, which is known as a place where hot mud appears due to drilling of a geological exploration well for gas that was carried out by PT Lapindo Brantas which give anthropogenic impact on mining and geological conditions (Davies et al., 2007). Furthermore, there was a hypothesis that an earthquake in Yogyakarta, Central Java also become one of the reason of this condition that happened two days before this incident occurred with a magnitude of 6.3. This incident has been going on for more than 16 years and has already submerged 3 sub-districts in Sidoarjo, which includes roughly 16 villages in this area. This condition is exacerbated by the large volume of mudflow. The latest data shows that the volume of this mudflow has reached more than 100.000 m^3/day. The large area submerged due to Sidoarjo mud causes a decrease in environmental quality. Several studies have shown a change of decline in the quality of the environment, including air, soil, as well as water (Auvaria & Munfarida, 2020; Zannah, 2021).

Water is the essential material in human history since water is utilized in every aspect of life, including agriculture as well as industry. The Sidoarjo mudflow (known as LUSI, which is the acronym of Lumpur means mud, and Sidoarjo as the region where the hot mud flows) which has occurred since 2006 has had a significant impact on water quality in the subregion of Porong, as well as the surroundings subregion, including Tanggulangin and Jabon. More importantly, this quality decreasing also occurs in groundwater, where groundwater is considered as the main water resource used by local people for any purposes in daily activity, such as cooking, hygienic purposes, as well as drinking. The water problem in Sidoarjo is very actual, considering that Sidoarjo is one of the most densely populated districts in East Java, with a population density in this region reaching 2.869 people/km^2 (Trisna, 2018). Thus, it can be ascertained that the need for clean water in this region is quite large, and is projected to increase every year by increasing of population.

The increasing of population also increases the demand of clean and fresh water for any purposes, including for drinking. The need for clean water will always be linear with the increase in human population (Boretti & Rosa, 2019). But unfortunately, in Sidoarjo this condition is not supported by an increase in a centralized water system, thus there is no control over water resources in this area. In general, every home should have its own well to have access of clean water. This inadequate system gives more disadvantage because there is no certainty about the quality of the groundwater used in every home. Certainly, consuming water without understanding its quality will bring a high risk to the health of people who consumes it if consumed in large quantities and over a relatively long period of time. Moreover, Sidoarjo has a large population of children, who are more vulnerable to health.

Our previous analysis shows that the quality of groundwater in the area near to Sidoarjo mud (LUSI), especially in the Porong area, has decreased. Based on the analysis results and previous calculations, some samples are classified as 'strongly polluted', and some are classified as 'maximum polluted'. From the samples analyzed, the groundwater in this area was polluted by several heavy metals such as Mn, Hg, and several other heavy metals found in several samples (Adiyaksa et al., 2023). In general, therefore, the results of our previous objective calculations of determining groundwater quality show that the groundwater around the Sidoarjo mudflow is polluted.

In this case, dangerous pollutants come from heavy metals. The heavy metals contain in water for consumption will potentially disrupt human health, as consumers, especially when it has high concentration (Fadhila & Purwanti, 2022; Putra & Setiani, 2020). Apart from having an impact on the environment, onset of some pathologies affecting the skin and internal organs, connected to the consumption of water contaminated with heavy metals, have been observed (Behrooz & Burger, 2021; Suryani & Sriwahyuni, 2021). Our previous findings show that children are highly vulnerable group to exposure to chemicals from well water in Tanggulangin, Sidoarjo, East Java, where is located only around 3 km from the center of LUSI (Adiyaksa et al., 2022). Therefore, monitoring water quality is highly required to improve the quality of human life.

Based on the explanation above, the aim of this study is to objectively assess by calculating the effect of exposure to heavy metals (Fe, Mn, Zn, As, and Hg) on human health using the formula by the United States Environmental Protection Agency (US EPA). In current case, the calculation has been carried out by including the Hazard Quotient (HQ) score, the sum of HQs as Hazard Index (HI) value, and Cancer Risk Index (CRI) from As- and Hg-contaminated water.

2. METHODOLOGY

2.1 Area of Study

Sidoarjo is located in East Java province, which enters the Kendeng basin. This basin is known as a zone of the central depression of the island of Java due to the collision of the Eurasian plate with the Indo-Australian plate. Sidoarjo also known as a delta surrounded by two major rivers. These 2 big rivers are located in Surabaya, which is Surabaya River, and in Sidoarjo, which is Porong river. Geographically, Sidoarjo has an area of 6.256 hectares, where is located with an altitude of 23-32 m above sea level. Geographical position of Sidoarjo with longitude of 112.741987 and latitude of -7.459679. The hydrogeological condition of Sidoarjo consists in the presence of areas of groundwater, brackish, and saline waters. The area of this hydrogeology of Sidoarjo reaches up to 16.312,69 hectares. The depth of groundwater is on the average of 5-10 meters from the surface of the land.

The average temperature in Sidoarjo did not change significantly during the year, due to the geographical location of Indonesia in the equator, with only two seasons. In 2021, based on the data of Meteorology Climatology and Geophysics Council (BMKG) Juanda, Sidoarjo, the lowest average monthly temperature was in January, which was 27,03 C, and the highest average monthly temperature was in October, which was 29,5 C.

2.2 Sampling and Chemical Analysis

The sample used was groundwater taken from 5 points near to the area of Sidoarjo mud (LUSI), with a distance of about 2 km from the center of the mudflow to every point of sample (Figure 1, Table 1). Groundwater was taken from wells with a depth of 6-10 m from the surface, and transferred in the 600 ml sample-bottles. Samples were taken on October 2nd, 2022. Chemical analysis was carried out at the Hydrogeochemical Laboratory, School of Earth

Sciences and Engineering, National Research Tomsk Polytechnic University, Tomsk, Russia. Heavy metals of groundwater were analyzed by using inductively coupled plasma mass spectrometry (NexION 300D mass spectrometer). The complete geochemical analysis method and calculation of groundwater quality is shown in (Adiyaksa et al., 2023).

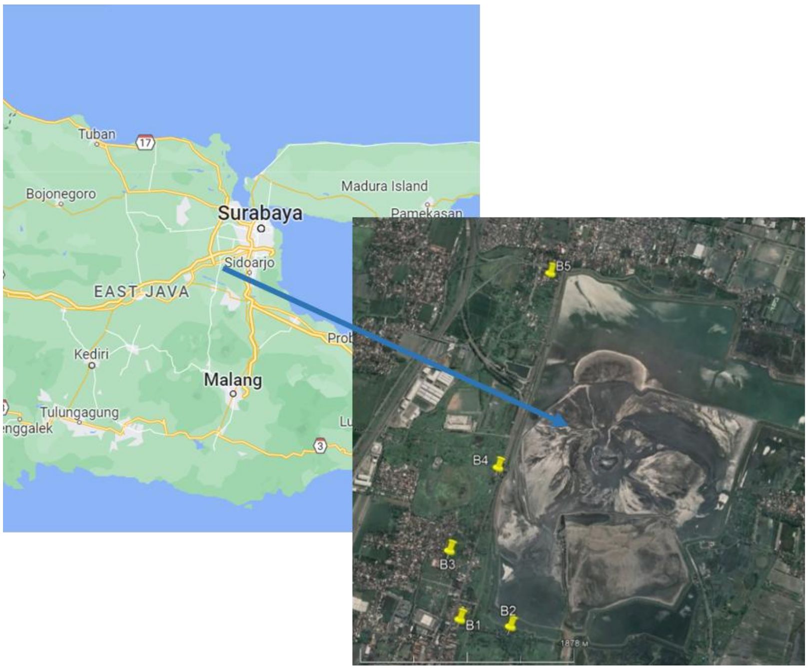

Fig 1. Sampling points of groundwater in the area of Sidoarjo mud (LUSI), East Java, Indonesia

Table 1. Coordinate of sampling location the area of LUSI, East Java, Indonesia

Sample	Latitude, φ	Longitude, λ
B1	-7,54	112,7
B2	-7,541	112,704
B3	-7,535	112,699
B4	-7,528	112,703
B5	-7,512	112,706

2.3 Method of Health Risk Assessment Due to The Metals Exposure

Health risk assessment for non-carcinogenic was calculated by considering the daily intake of the amount concentration of metals in groundwater and its value of Hazard Quotient (HQ) for various cases, namely:

1. Children aged 1-3 years old (as C1)
2. Children aged 4-8 years old (as C2)
3. Children aged 9-12 years old (as C3)
4. Career women (as D1)
5. Careen men (as D2)

Some calculation parameters based on the above 5 subjects/cases follow the parameters shown in Table 2 (Kończak & Janson, 2021).

A method recommended by the United States Environmental Protection Agency (US EPA) was used to assess the health risk from heavy metal exposure. The steps of this calculation following the work done by *(Adiyaksa et al., 2022)*. Firstly, calculation of the daily input of contaminants is carried out using equation (1). Several calculation parameters are following Table 2.

$$I = \frac{C \: x \: IR \: x \: FE \: x \: TE}{WT} \: x \: \frac{1}{TA} \quad (1)$$

Where:
I : daily contaminant intake (mg/kg-day)
C : concentration of each heavy metals (mg/L), in this case concentration of Fe, Mn, Zn, As, and Hg
FE : frequency of exposure (308 days/year)
IR: ingestion rate of water (L/day)
TE : time of exposure (years)
WT : weight of consumer (kg)
TA : time average (days)

$$HQ = \frac{I}{RfD} \quad (2)$$

$$HI = \Sigma \: HQ = HQ_{Fe} + HQ_{Mn} + HQ_{Zn} + HQ_{As} + HQ_{Hg} \quad (3)$$

Where:
I : daily contaminant intake from the previous equations (mg/kg-day)
RfD : reference dose (mg/kg-day), for Fe is 0,7; Mn is 0,14; Zn is 0,3; As is 0,0003; and Hg is 0,0001

When the value of HQ is less than 1 (HQ<1), the consumer of water in the research area is safe from any specific effects, but when HQ is more than 1 (HQ>1), there is possibility for the water consumer from any specific harmful effects. As a cumulative value that considers each HQs of the metal, the Hazard Index (HI) was calculated. There is a possibility of a health risk if HI>1 obtained, while HI<1 describes the absence of a possible health risk.

While to calculate the Cancer Risk (CR) from the As- and Hg-contaminated water, the following equation (4) was implemented.

$$CR = I \times SF \quad (4)$$

Where:
I : daily contaminant intake from the previous equations (mg/kg-day)
SF : oral slope factor (mg/kg-day). SF value of As is 1,5 mg/kg-day, and Hg is 1 mg/kg-day (Djadé et al., 2021)

Table 2. Calculation parameters for several cases

Parameters for calculation	C1	C2	C3	D1	D2
FE – frequency of exposure (days/year)	308	308	308	308	308
IR – ingestion rate of water (L/day)	1,3	1,6	1,9	2,7	3,7
WT – weight of consumer (kg)	15	20	35	65	78

Source: (Kończak & Janson, 2021)

3. RESULT AND DISCUSSION

3.1 Metals Content in The Groundwater of The LUSI Area, East Java, Indonesia

The Ministry of Health of the Republic of Indonesia regulation No. 32 2017 concerning “Environmental Health Quality Standards and Water Health Requirements for Sanitation Hygiene Purposes, Swimming Pools, Per Aqua Solutions, and Public Bathing” has been regulating the maximum standard for the heavy metal concentration in water (Menteri Kesehatan RI, 2017). This standard was formulated to provide safety and health to water users in Indonesia, especially for drinking water. As we know, drinking water has a significant impact on the human health, thus maintaining its quality, in accordance with applicable standards, is highly required. Based on the standard, the threshold concentrations, which should not be exceed are: As 0,05 mg/L, Fe 1 mg/L, Mn 0,5 mg/L, Zn 15 mg/L, and Hg 0,001 mg/L (Table 3).

Based on these standards, the B1 sample was contaminated with Mn. The Mn content in sample B1 was 1,4759 mg/L while the maximum applicable standards were only 0,5 mg/L. The same contaminants were also identified in sample B2, where the Mn content was 0,7135 mg/L. In the B3 sample, all heavy metals content did not exceed the standard. Sample B4 is contaminated with Fe, Mn, and Hg. The content of Fe is 1,4823 mg/L, Mn is 4,1122 mg/L, and Hg is 0,0013 mg/L. In the sample B5, the water is contaminated with Mn 0,6187 mg/L, and Hg 0,0264 mg/L. In general, almost in all water samples the threshold value for Mn is exceeded. Meanwhile, As and Zn were not detected above the standard in all samples.

Sidoarjo mud (LUSI) contains various kinds of heavy metals, with the highest concentration of Fe, followed by Si, and several other chemical compounds such as Al, Mn, Zn, and others (Ciptawati et al., 2022). This then becomes one of the reasons for the heavy metals’ presence in the groundwater around the LUSI area. These heavy metals are an indication that the groundwater in the LUSI area is contaminated and cannot be classified as drinkable.

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Table 3. Chemical parameters and metals content in groundwater the area of LUSI, East Java, Indonesia (October 2022)

Parameters	Unit	Standard	B1	B2	B3	B4	B5
pH	-	6,5-8,5	6,52	6,63	6,59	6,47	6,39
Electrical conductivity, EC	μΣ/χμ	-	591	584	455	1229	3160
Sum of main ions, S_{mi}	mg/L	-	576,2	535,3	441,1	863	2579
Water classification	-	-	Calcium bicarbonate	Calcium bicarbonate	Calcium bicarbonate	Sodium chlordide	Sodium chlordide
As	mg/L	0,05	0,0002	0,0006	0,0064	0,0162	0,0031
Fe	mg/L	1	0,0739	0,4173	0,0874	**1,4823**	0,2003
Mn	mg/L	0,5	**1,4759**	**0,7135**	0,2909	**4,1122**	**0,6187**
Zn	mg/L	15	0,008	0,0647	0,0009	0,0015	0,0075
Hg	mg/L	0,001	0,0007	**0,001**	0,0005	**0,0013**	**0,0264**

*Standard used based on the regulation of Ministry of Health of Republic of Indonesia No. 32 2017

3.2 Hazard Quotient (HQ) of Groundwater in The Area of LUSI, Sidoarjo, Indonesia

Hazard Quotient (HQ) value has big role on determining the impact of water chemical exposure to human health (Qasemi et al., 2018). The result of the HQ calculation of the As content is shown in Fig 2 (a). Samples B1, B2, and B5 generally have HQ below 1 for all subjects, namely children aged 1-3 years (C1), children 4-8 years (C2), children aged 9-12 years (C3), career women (D1), and men career (D2). The lowest HQ is found in sample B1. In sample B3, HQ greater than 1 was observed in the following groups; children aged 1-3 years, children aged 4-8 years, children aged 9-12 years, and career men. Meanwhile, in sample B4, all subjects were estimated to be affected by exposure to the As content in the water. Sample B4 is the place with the highest HQ due to the high content of As found in this area. In general, children, especially those aged 1-3 years, are more susceptible to exposure and their health is affected. Children always have a higher HQ trend.

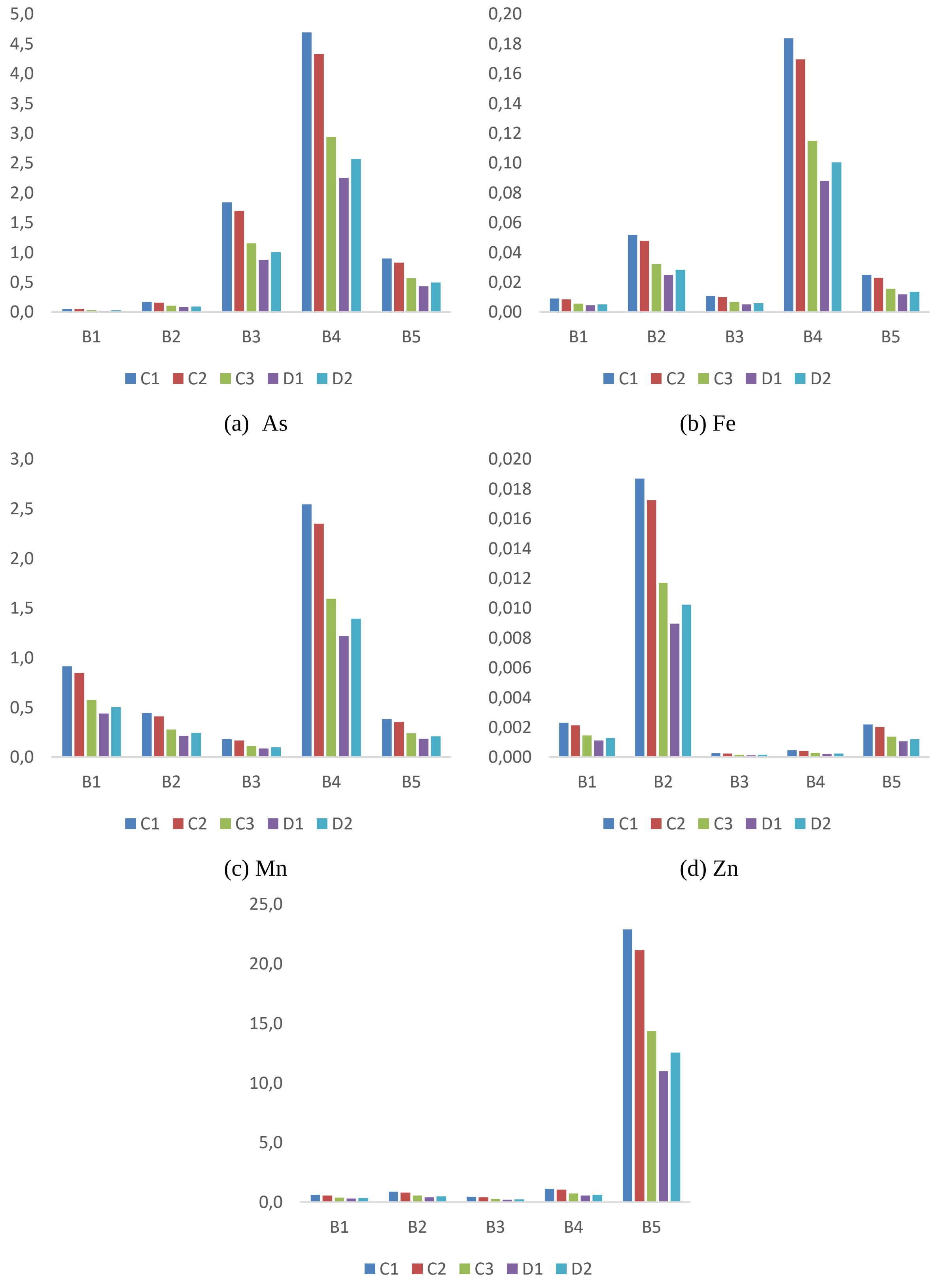
Figure 2 (b) shows the HQ value of the Fe content in water. In general, the HQ values obtained for all samples did not overcome 1. The highest HQ values were in sample 4, while the lowest were in sample 1. Children aged 1-3 years had the highest HQ for each sample, and career women has the smallest HQ for each sample.

In the case of Mn exposure, Figure 2 (c) shows the results of HQ calculations. Sample B4 has an HQ value of more than 1 for all subjects, namely C1, C2, C3, D1, and D2. While the rest have HQ less than 1, with the smallest HQ being in the sample B3.

With a very low Zn content compared to the maximum standard, the HQ calculation results obtained for exposure to Zn in Figure 2 (d) are less than 1. The highest HQ value is in the sample B2, while the smallest HQ is in the sample B3.

Figure 2 (e) shows the HQ value of the Hg content in water. Hg was identified as very high in the B5 water sample, causing a considerable HQ value, reaching more than 20 in the case of children. In addition, the Hg in the B4 water sample also exceeded the standard, even if slightly. This also causes an increase in HQ values above 1 for cases of children aged 1-8 years.

Consumption of groundwater in this area shows unfavorable results because of the potential for health problems. In line with other studies (Adiyaksa et al., 2022; Fomina et al., 2018; Nikiforova & Vidishcheva, 2018), in general, children has greater risk of exposure to their health from these heavy metal contents than adults. With the same concentration of heavy metal content, children have lower in weight, consume less water daily, and in shorter intervals. Apart from being technically seen from the formula used, children are more vulnerable because it relates to their habits, nutrition, and their mobility.



(e) Hg

Fig 2. HQ value of each heavy metals content exposure for each groundwater samples in the area of LUSI, East Java, Indonesia

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Other research shows that there is a correlation between the ingestion of As-contaminated water and skin disease, including the presence of pigmentation, the formation of lesions and patches, and in same case may act as a precursor in skin cancer (Khosravi-Darani et al., 2022). With exposure to Mn, the human nervous system can be disturbed. Consumption of water with high Fe concentration can cause diabetes to liver disorders. Consumption of water containing Zn compounds can encourage the emergence of several neurodegenerative diseases (Alissa et al., 2007; Ashar, 2007; Kelly, 2002). For children, the desease such as edema, hypertension, as well as redness can be led by the toxicity of Hg content in water. In addition, exposure to high Hg in consumed water can cause kidney dysfunction and high blood pressure. It is reported as well, that gastrointestinal disorders also can be caused by Hg-contaminated water for consumption (Djadé et al., 2021).

3.3 Hazard Index (HI) of Groundwater in The Area of LUSI, East Java, Indonesia

The non-carcinogenic effects potential risk from observed heavy metals exposure through consumption was calculated by summarizing the HQs value as the hazard index (HI) (Castresana et al., 2019). In this study, the HI value is calculated as the sum of the HQ values of As, Fe, Mn, Zn, and Hg exposures. The result of summing HQs to the HI value shows that the water in sample B5 has the biggest potential effect on human health, with an HI value of more than 20 (HI>1) (Table 4). Children, in general, are at greater risk than adults, especially those between 1-3 years old. Furthermore, the second highest HI value was in the sample B4, where the HI values of more than 1 were observed in all scenario. The HI value of B4 is quite high, considering the contaminant of Fe, Mn, and Hg. The third highest HI value is in the sample B3 with HI of more than 1 also in all scenario. in the sample B1 the HI value obtained more than 1 was identified only on children aged 1-8 years old.

In Table 4, it is shown that only samples B1 and B2 have, in general, HI values below 1 (HI<1). This shows that the water in samples B1 and B2 has the lowest risk. However, sample B1 contains Mn which is quite large, exceeding the quality standard set by the regulation of the Ministry of Health, and so does sample B2.

Table 4. The sum of HQ value calculated as HI value for every chemical content

Sum of HQ	Consumer	B1	B2	B3	B4	B5
HI = Σ HQ	**C1**	1,581	1,548	2,46	8,544	24,189
	C2	1,46	1,429	2,271	7,887	22,329
	C3	0,99	0,97	1,541	5,352	15,152
	D1	0,758	0,742	1,179	4,095	11,594
	D2	0,866	0,847	1,347	4,677	13,24

3.4 Cancer Effect Risk of As- and Hg-contaminated Groundwater in The Area of LUSI, East Java, Indonesia

The Cancer Risk Index (CRI) is the likelihood of developing cancer as a result of potential exposure to carcinogens in water (Rahman et al., 2021). In this case, the CRI calculation is performed only for As- and Hg-contaminated water. For the determination of CRI values, the United States Environmental Protection Agency (US EPA) generally suggests that in a million there is one case of cancer (i.e., $1,0 \times 10^{-6}$). This is used for government as a tool to make a management-level decisions. The value of Cancer Risk Index (CRI) surpassing $1,0 \times 10^{-4}$ (in 10.000, there is one case of cancer) is not accepted (US EPA, 2005).

The water contaminated with As in all the samples has no risk of cancer effects in all subjects, considering the CRI value, which is not exceeding 1.0×10^{-4} (Table 5). Meanwhile, for water contaminated with Hg, the CRI value for samples at B5 exceeded the standard value. This is because the Hg content in the B5 sample is very high, causing a risk of cancer. Cases in B4 for children aged 1-8 also had a CRI over the standard, but the difference was quite small. In addition, ingesting groundwater contaminated with As and Hg exposes people, especially children, to cancers, such as liver, kidney, to skin cancer (Djadé et al., 2021).

Table 5. Cancer risk calculation for every scenario of As- and Hg-contaminated groundwater in the area of LUSI, East Java, Indonesia

Contamination	Consumer	B1	B2	B3	B4	B5
As	**C1**	$1{,}26 \times 10^{-7}$	$4{,}31 \times 10^{-7}$	$4{,}68 \times 10^{-6}$	$1{,}20 \times 10^{-5}$	$2{,}29 \times 10^{-6}$
	C2	$1{,}16 \times 10^{-7}$	$3{,}98 \times 10^{-7}$	$4{,}32 \times 10^{-6}$	$1{,}10 \times 10^{-5}$	$2{,}12 \times 10^{-6}$
	C3	$7{,}90 \times 10^{-8}$	$2{,}70 \times 10^{-7}$	$2{,}93 \times 10^{-6}$	$7{,}49 \times 10^{-6}$	$1{,}44 \times 10^{-6}$
	D1	$6{,}05 \times 10^{-8}$	$2{,}07 \times 10^{-7}$	$2{,}24 \times 10^{-6}$	$5{,}73 \times 10^{-6}$	$1{,}10 \times 10^{-6}$
	D2	$6{,}91 \times 10^{-8}$	$2{,}36 \times 10^{-7}$	$2{,}56 \times 10^{-6}$	$6{,}54 \times 10^{-6}$	$1{,}26 \times 10^{-6}$
Hg	**C1**	$6{,}07 \times 10^{-5}$	$8{,}67 \times 10^{-5}$	$4{,}33 \times 10^{-5}$	$\mathbf{1{,}13 \times 10^{-4}}$	$\mathbf{2{,}29 \times 10^{-3}}$
	C2	$5{,}60 \times 10^{-5}$	$8{,}00 \times 10^{-5}$	$4{,}00 \times 10^{-5}$	$\mathbf{1{,}04 \times 10^{-4}}$	$\mathbf{2{,}11 \times 10^{-3}}$
	C3	$3{,}80 \times 10^{-5}$	$5{,}43 \times 10^{-5}$	$2{,}71 \times 10^{-5}$	$7{,}06 \times 10^{-5}$	$\mathbf{1{,}43 \times 10^{-3}}$
	D1	$2{,}91 \times 10^{-5}$	$4{,}15 \times 10^{-5}$	$2{,}08 \times 10^{-5}$	$5{,}40 \times 10^{-5}$	$\mathbf{1{,}10 \times 10^{-3}}$
	D2	$3{,}32 \times 10^{-5}$	$4{,}74 \times 10^{-5}$	$2{,}37 \times 10^{-5}$	$6{,}17 \times 10^{-5}$	$\mathbf{1{,}25 \times 10^{-3}}$

4. CONCLUSION

A preliminary health risk assessment due to the metals exposure of groundwater in the area of the mud volcano in Sidoarjo was obtained based on the results of survey and calculation in 2022. Health risk due to heavy metals exposure was assessed that 1) based on its HI value, groundwater in this area, especially in the sample of B3, B4, and B5 will give impact on human's health, since the HI value is more than 1. The highest HI value was recorded in the B5 sample, since it was noted that Hg concentration in this sample was high; and 2) as mentioned that water in the B5 sample was contaminated by high concentration of Hg, the calculation showed that it will give risk of cancer. In general, water in the Porong area, Sidoarjo, influences health from exposure to these heavy metals, which include As, Fe, Mn, Zn, and Hg. The highest exposure risk was in sample B5, and the lowest in sample B2.

To conclude, it is highly required to do groundwater monitoring in Porong, Sidoarjo. The risk of exposure to As, Fe, Mn, and Zn will raise if there is an increase in concentration. In addition, exposure to other heavy metals cannot be excluded. Thus, for the current conditions, groundwater is not suitable for direct human consumption, but it needs specific treatments to lower the concentration of the contaminants.

5. ACKNOWLEDGEMENT

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