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Analysis of Heavy Metals (Cadmium, Chromium, Lead, Manganese, and Zinc) in Well Water in East Java Province, Indonesia

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ABSTRACT

Introduction: Indonesia is one of the countries whose inhabitants use well water for drinking and cooking purposes. In East Java, 48.90% of the population uses well water for their daily needs. Well water contained heavy metals had bad effects on health such as cancer, damage of liver, kidneys, and others. The objective of this study was to evaluate the concentration of heavy metals in well water and relate them to a potential health outcomes. **Methods:** The method used in this study was analytical descriptive. Data used was secondary from East Java Environmental Office. A total of 101 samples were collected from 33 locations. There were 5 heavy metals analyzed, namely cadmium, chromium, lead, manganese, and zinc. Equipment using ICPMS and AAS. Data analyzed with descriptive statistics by SPSS. Data obtained were compared to the WHO Standard for Drinking Water Quality. **Results:** Concentration for cadmium was 0.002 mg/l, followed by manganese at 1.80 mg/l and zinc at 0.020 mg/l. Besides, all water samples had levels of chromium and lead below the detection limit. **Conclusion:** All heavy metals had concentrations below the maximum allowable standard, except for five water samples from three locations with levels of manganese which was above the maximum standard. Long term effects of manganese include neurological problems, intelligence, and low birth weight. Further studies need to be done to determine the source of manganese contamination. It is recommended that bottled water is used for drinking purposes in an area where heavy metal concentration is above the allowable limit.

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Commented [S2]: Revised: remove "5" and "3"

Keywords: *heavy metal, well water, manganese, drinking water, health risk*

INTRODUCTION

Water is important to human life that is always needed every day. Water consumed by humans is needed by all organs of the body so that health maintained. The availability of water on earth is tremendous, but only a small portion can be used for drinking. Experts mentioned that the surface of the earth is covered by water by 71% but only 2.5% is freshwater that can be consumed by humans (1). Freshwater can be obtained from underground water such as wells. Many countries use well water as their main source of drinking water, in study Indonesia. As many as 27.04% of households in Indonesia use well water for drinking and cooking purposes. That number is the second-highest after bottled drinking water sources, at 31.30% of households (2). One province that uses well water as a source of drinking water is East Java. As many as 48.90% of households in East Java use well water, which is the highest compared to other sources such as bottled water 29% and 8.70% pipe water (3).

Well water quality consumed by the community for their daily needs is the most important. Currently, the quality of well water is threatened by a large number of water pollutants and it mainly occurs in urban areas and areas that have become centers of intensive agriculture (1). Contamination that occurs in well water may be caused by natural processes and the consequences of human activities. Some contaminants that have become important to comply with guidelines of the United States Environmental Protection Agency (USEPA) are microorganisms (bacteria, viruses, and parasites), nitrates and nitrites through chemical fertilizers, human sewage, and animal waste; heavy metals like copper, chromium, arsenic, lead, antimony, cadmium, selenium, organic chemical; radionuclides; and fluoride (4,5).

Heavy metals are a group of metal elements that have a density of more than 5 gr/cm^3 and a high relative density. Heavy metals have toxic properties down to ppb levels. Examples are Cd, Zn, Fe, Pb, Mg, As, Pt, and Cu. The sources of these metals from natural and human or anthropogenic activities such as mining, transportation, and industrial activities. Heavy metals cannot be lost naturally because they continue to accumulate so that they can cause harm to human health if exposed continuously for a long time. Some research has be conducted to examine the quality of well water in several regions in Indonesia. The results show that the concentration of manganese (Mn) in some areas has exceeded the maximum drinking water standard. For example well water in Jakarta indicated that Mn levels in Jakarta residents have exceeded the maximum standard of 0.970 - 1.022 mg/l (6). Also, the research in Sukoharjo shows water is at 1.43 mg/l, exceeded drinking water standards (7). Iron (Fe) concentration, also showed exceeded standards such as in areas around the Kaliyasa river, Cilacap City where the level was 2.3 mg/l (8). Similarly, well water in Sukoharjo has a high Fe level of 1.45 mg/l (7). In addition to these two elements, lead (Pb) level also recorded high reading from the maximum standard (0.01 mg/l), such as in Palembang, where it was known that Pb in well water around landfills as 0.03 – 0.05 mg/l (9). And research in Pekalongan also showed that Pb levels in well water was high 0.04 mg/l (10). Cadmium (Cd) levels were also reported to be above the maximum standard (0.003 mg/l). In a Yogyakarta study, cadmium levels in well water reached 0.0178 mg/l (11). Similarly, a research report in Jember, East Java, showed the level of Cd in well water around landfills were 80% higher than the maximum standard (12). This shows a little picture of the condition of well water pollution in Indonesia. Besides, other studies discussed the quality of well water in East Java, especially heavy metals contamination.

Contaminated well water may cause a variety of public health problems. Many studies have reported that pollutants entering the human body through drinking water will cause negative health impacts on the consumers. Some health problems that may arise include microorganism infections, reduction of the blood's ability to carry oxygen, heavy metal risks such as acute and chronic toxicity on some organs including the liver, kidneys, and intestines, and medical conditions like anemia, cancer, and hormonal disorders (4,13). A Studied in northeast Iran on the content of heavy metals in drinking water showed that Cr levels exceed the safe level of US EPA risk. This condition is stated from the results of the Hazard Index that will be carcinogenic in children and adults through the consumption of water and skin pathways (14).

As many people rely on well water for their daily needs and the challenges of health problems that may arise due to contamination, a study is needed that will illustrate well water quality in East Java. This province is the third-largest population in Indonesia. Various kinds of population activities such as very dense transportation, many large-scale industries to accommodate workers, home industries in each district area, as well as household activities in densely populated housing carryout negative impacts on the environment including the occurrence of well water pollution. This situation is very appropriate to be the basis for evaluating heavy metals in well water because a study in Port Klang, Malaysia also states that heavy metal pollution in water and sediment will increase along with the intention industrialization and urbanization (15). This is also an effort to improve the 6th sustainable development goals indicator, which is about safe drinking water. A managed safe drinking water services are defined as one definite location, always available for daily needs, and safe from contamination (16). The aims of this study, therefore, were to identify and analyze the quality of well water and the health impacts caused by heavy metal contaminants in East Java Province, Indonesia.

MATERIALS AND METHODS

Study Design, Location, and Time: This was an analytical descriptive study using secondary data. The data was sourced from the Information on Environmental Management Performance Report in 2016 - 2017 by the Environmental Office, East Java Province, Indonesia.

Population dan Sample: Water samples were obtained from 101 groundwater sources at 33 district locations and were collected from February 2016 - August 2017. The sample selection in these areas was based on community reports to the environmental office in each region. The community provided information on the differences in water quality based on their observations by the physical quality of the water. Therefore it needs proper monitoring with the appropriate equipment to ensure the quality of this water. Well water sampling technique refers to the guidelines from the Ministry of Health of The Republic of Indonesia. To ensure that quality of sampling and inspection, was carried out by a research team who had received training and a certificate from the environmental office training center agency.

Variable Data: Variables in this study were 5 heavy metals, namely Lead (Pb), Cadmium (Cd), Manganese (Mn), Chromium (Cr), and Zinc (Zn) in well water.

Processing, Analysis, and Presentation of Data: The level of each heavy metals were determined at the Laboratory of Technical Center of Environmental Health and Disease Control in Surabaya. The equipment used were Inductively Coupled Plasma Mass Spectrometry (ICP-MS) and Atomic Absorption Spectrophotometer (AAS). ICP-MS was an analytical technique that can be used to measure elements at trace levels in biological fluids. The advantages of this technique were multi-element technique, large analytical range, low detection limit, high sample throughput, low sample volume, simple sample preparation, high-resolution and tandem mass spectrometry (triple-quadrupole) instruments offer a very high level of interference control.

Commented [S3]: Remove: The data collected were presented in tabular, graphic and narrative forms

Then data was analyzed with descriptive statistics by SPSS. While water quality assessment was compared to The Drinking Water Quality Standard by the World Health Organization (2017). According to the regulation, there were the limit standards of heavy metals, 0.003 mg/l for cadmium, 0.05 mg//l for chromium, 0.01 mg/l for lead, 0.4 mg/l for manganese, and 3 mg/l for zinc (17). A review of potential health effects was also discussed by literature research. Recommendations with regards to health impacts were presented at the conclusion.

RESULTS

The results of heavy metals measurement in this study were presented in Figure 1. It showed that only manganese (Mn) had 5 readings which exceeded than standard (0.4 mg/l) detected in samples from location 55 (1.8 mg/l) in Malang District, location 87 (1.78 mg/l), 100 (0.68 mg/l) and 101 (0.43 mg/l) in Surabaya City, and location 89 in Mojokerto District (1.20 mg/l). While the other metals had a low level in all of the locations.

Table I showed a comparison between measurement results with the maximum standard for heavy metals in drinking water. The levels of cadmium (Cd), chromium (Cr), lead (Pb), and zinc (Zn) were below the allowable standard, even the level of chromium and lead were below limit detection in all samples. While the level of manganese (Mn) exceeded the WHO standard for drinking water.

According to figure 3, all water samples did not exceed the drinking water quality standard for Cd (0.003 mg/l) but there are 2 locations (location 55 and 56 in Malang) with the highest level of Cd ie 0.0024 mg/l. While Mn levels (showed in figure 4) in 95% of the samples were under the detection limit of 0.05 mg/l, but 5 samples were above the standard (0.4 mg/l) and the points of locations were mentioned above. Zn level in figure 5 showed that all levels below the standard in drinking water (3 mg/l), but there were 2 locations with the highest level of Zn ie 0.020 mg/l (location 26 in Bangkalan and location 28 in Sampang). Very good results were showed by Cr and Pb which had all samples (100%) in figure 1 had levels under the detection limit. It was below the measurement ability of the equipment.

This figure (Fig. 2) was the result of mapping from the measurement of heavy metals level in samples area (East Java Province). This area was one of the 34 provinces in Indonesia country. This map showed 28 district locations with the blue color had below standard for all heavy metals, 2 district locations with the green color had the highest level of Zn, 1 district location with the yellow color had the highest level of Cd, and 3 district locations with the red color had Mn level above the standard. While the black color was not sampled locations.

DISCUSSION

Cadmium (Cd): Cd sources in natural waters are more commonly found in basic sediments and suspended particles. Besides, that Cd will be affected by the degree of acidity of the water. While, the anthropogenic sources of Cd are from agriculture, landfill, sewage sludge, and a lot of activities in urban areas such as high emissions, industry, and mining. However, identification can be difficult because one pollution route can be from multiple sources simultaneously leading to groundwater sources (18). The results of the data in this study showed similarities with research on assessment of well water quality in the city of Kakamega, Kenya which stated that the levels of cadmium were lower than the WHO standard (19). However, this level is different from research in East Delhi, India. It is stated in the study that the sample of groundwater had a level up to 0.28 mg/l, it was above the safe level standard (20). The research was conducted in the Krishna Vigar industrial area. The presence of cadmium in groundwater or well water is dangerous if it is consumed by humans. As happened in Namo Bintang, Deli Serdang, North Sumatera Province, Indonesia, it was stated that 65 respondents (65.7%) were exposed by drinking water which contains cadmium with a concentration level above the safe standard. Apart from the research also showed the urine test 99% of the sample showed a Cd level exceeded the normal limit, so the recommendation of researchers was residents do not use well water as the main source of drinking water (21). Cd that has levels exceeding the WHO standard will potentially damage human health. In general, organs that are targeted by Cd are kidneys and bones. Canada Health Authority explained that "Cadmium exposure is well known to result in damage to the nephron's proximal tubule, causing impaired reabsorption of low molecular weight proteins and enzymes by the kidneys" (19 p. 21). Besides, "cadmium exposure has long been associated with reduced bone mineral density, osteoporosis, and fractures" (19 p. 23). Another long-term effect is on the onset of cancers based on animal testing, making cadmium a carcinogen to humans (23). Various ways can be taken to reduce the level of Cd in water, one of which is the Bioreduction Adsorbent with the bacteria Bacillus sp. and durian leather. The biosorbent is carried out in several steps, starting with the manufacture of activated charcoal from durian leather and continuously by administering the isolated bacteria with medium Cd. Both of them can be done sequentially to get the optimum results (24).

Chromium (Cr): This study is supported by research conducted at Agbor and Owa Community of Nigeria, which stated that chromium levels in all groundwater samples were below the detection limit (25). In contrast to chromium detection in the Unnao district of India, Cr levels were higher than the safe limit of 0.141 mg/l in the water hand pump samples at Gupta Gate, Shivnagar (26). The high level of chromium at this location is probably due to the study location being close to the Ganga River, whereas the river holds nearly 70% of the untreated waste from the 50 leather industries. In addition, research in Bangladesh also has different results from this study. It is stated that the Cr pollutant in groundwater located close to the Meghna Ghat industrial area has a Cr level that exceeds the safe standard because it reaches 0.07 mg/l (27). Reviewing from these studies, the value of Cr is always high in areas that are located close to industrial areas. This indicates that the potential for pollution occurs due to the lack of monitoring of waste management from the factory. Chromium in the environment is usually divided into 2 groups namely Cr^{3+} and Cr^{6+} but for drinking water quality, total chromium. In general, the concentration of chromium in groundwater is low (< 1 µg/liter). Because of that, it takes a long time to find out the effects of Cr exposure on the human body. One way is through an examination of Cr levels in blood and hair. As mentioned in previous studies, personals exposure dose of chromium for drinking water during lifetime showed associations with levels of chromium in the blood and hair, furthermore in other parameters such as hematological and biochemical also (28). Another way is urine examination by the previous study, which showed that the method is effective to determine the chromium level in the body. It's mentioned that 53% of workers sampled detected chromium levels are exceeded the normal standard (29).

Lead (Pb): This research had similar data with the study conducted in Surabaya. It was stated that the Pb contamination in groundwater was below the detection limit (30). The research was conducted in the coastal area of Surabaya City. Compared to research in Algeria where

Pb levels (0.072 up to 0.458 mg/l) had exceeded the WHO standard (0.01 mg/l). It has a potential health risk especially impact for the population who consumed the water (31). It similar to a study in East of Algeria also said that the concentration of the lead is above the maximum permissible standard, the level of sample ranges from 0.017 to 0.292 mg/l (32). In addition, the case study in Surulere also showed that 36.73% of well water samples had a Pb concentration level above the maximum contaminant level (33). Children and infants are two groups who are highly sensitive towards health problems caused by Pb. Estimated from a 5 µg/l Pb concentration in drinking water, the Pb intake for infants is assumed to be around 3.8 µg/day, whereas for adults up to 10 µg/day (34). Health problems caused by Pb are very diverse, including neurological effects in children and adults, and also renal, cardiovascular, hematological, immunological, and reproductive effects (35). But, this study had no worries about health risk potential to the population because the level of Pb was safe.

Manganese (Mn): This study showed similarities with results of a study on heavy metal concentrations in well water in Tamil Nadu, India, that mentioned manganese (1.276 mg/l) in the water exceeded the maximum standard (36). One study which analyzed well water quality in the Sidoarjo mudflow area, stated that Mn level exceeded the WHO standard as the mud is the source of Mn contamination of groundwater (37). The other research in Western Amazonia, Peru also had similarities with this study. Which is the contamination of Mn in groundwater was very high from maximum standard, which was 4 mg/l (38). However, it is different from research on groundwater at Kilvelur Taluk, Nagapattinan District, Tamil Nadu, India which showed that the Mn levels detected in 3 phases (pre-monsoon, monsoon, post-monsoon seasons) from 20 locations had a safe condition of concentration levels (0.02 – 0.26 mg/l) below WHO maximum limit (39).

The high concentration of Mn in the three areas mentioned can be caused by several sources, the first of which is the natural content of groundwater. This statement is supported by the theory that Mn in water can come from natural sources in bedrock, specifically, water from deep wells (40) and the United States National Water Quality Assessment Program showed that the levels of manganese in groundwater is about 99% is commonly higher than in surface waters. The natural presence of Mn that enters groundwater and well water can be influenced by several factors such as TDS, fluctuation of groundwater level, and time of residence of water in the ground (41). Other sources of Mn that can pollute well water are agricultural practices and leachate disposal. This may be the reason behind the high Mn level in sample 55 where the well is located near a landfill area. This is evidenced by research that discussed the content of Mn in well water and leachate in Banyuurip landfill, Magelang, Indonesia. It stated that Mn is the main substance in leachate that originates from metal wastes that have accumulated at the landfill. This researcher also explained that "there is a very strong and significant correlation between landfill-to-well distance and total Mn concentration" (32 p. 4). Besides, contamination may occur by industrial activities as evident through samples obtained in Surabaya City and Mojokerto District which are centers of industries. WHO, in Concise International Chemical Assessment Document 63 about manganese, mentioned that Mn pollution which originates from industry has been happening for a long time. Mn can enter water bodies through industrial waste disposal facilities. Since 1983, it has been estimated that Mn that enter the waters from anthropogenic sources worldwide amounted to 109,000 to 414,000 tons. Domestic wastewater and sewage sludge are the most important sources. Reports from the USA stated that Mn does not only pollute surface water, but is also able to penetrate groundwater up to 0.114 tons (43). Although smaller than surface water pollution (up to 17.2 tons), this fact is a real threat to health in the future. Therefore, it can be said that these 3 sources are eligible to be potential causes of Mn pollution.

One health disorder that is often linked to Mn contamination is neurologically-based, such as abnormal gait, ataxia, muscle hypotonicity, and a face without permanent emotions. Besides, liver dysfunction has also been reported (44). Initially, research conducted in Canada involving 259 children between 2012 - 2014 stated that there was nothing to show about the relationship between manganese and the cognitive development of school children, thus allowing for gender-based associations (45). Later, in 2018, another study on Mn in drinking water was conducted in Canada involving 630 children (aged 5.9 - 13.7 years) which discussed drinking water with manganese content and were associated with decreased performance IQ scores up to 5 %. It showed that Mn level of 78 µg/L (0.078 mg/l) contributed to a decrease of 1% Performance IQ, followed by a decrease of 2% for levels of 156 µg/L (0.156 mg/l), and a decrease of 5% for levels of 406 µg/L (0.406 mg/l). Therefore, it is very important to re-checking the maximum concentration of Mn in drinking water, with the aims are to protect and prevent problems in children's health (46). Potential health problem is also shown for adults, such as risks to pregnant women who consume water with Mn. Cohort studies were conducted in three cities in South Korea, with 331 mother-infant respondents from July 2007 to December 2009. The results of the study mentioned that any relationship of Mn levels in the blood (high and low) with low birth weight of infants (less than 3000 g). It is, therefore, clear that high Mn (more than 36 µg/l) in the blood has an association with the presence of low birth weight of infants (47). Some of these studies showed that health problems caused by Mn actually occur. Because of that, it is not recommended for people especially children and pregnant women to consume water with high level of Mn during their lifetime.

Zinc (Zn): This concurs with results from research in Nigeria which stated that Zn concentration of 0.02 mg/l did not exceed the maximum standard by WHO (25). In contrast with a study in Ghana showed that Zn levels in shallow groundwater were exceeded the recomended value of WHO regulation in 2008 (48). Zn is naturally present in groundwater and it can be influenced by the acidity of the water. According to the acidity theory, the higher the acidity of the water, the higher Zn concentration will become. Some types of Zn found in the soil can seep into groundwater so that it is possible if waste pollution occurs at a location, it will affect Zn concentration in the water (49). In general, it is stated that the natural presence of Zn is indeed smaller than other heavy metals. One study that discussed the assessment of heavy metals mobility states that the mobility and bioavailability of Zn were the lowest when compared to Mn and Cu (Mn > Cu > Zn) (50). Even though, Zn is one of the minerals needed by the body for cell and tissue growth. While, recommendation of WHO showed if consuming too much, zinc will cause damages to organs such as prostate, bone, muscle, liver, and gastrointestinal system. The recommended zinc consumption limits based on Recommended Dietary Allowances (RADs) is 11 mg/day for male and 8 mg/l for female (49).

CONCLUSION

Nearly all the heavy metals studied such as Chromium, Cadmium, Zinc, and Lead had levels below the maximum allowable standard by the World Health Organization, except for manganese (Mn) where 5 samples had a concentration which exceeded the standard ie 0.43 mg/l until 1.80 mg/l. The source of Mn in the water can occur by earth's layer content, agriculture practice, leachate disposal, industrial activities, and domestic sewage. Therefore, further studies need to be done to determine the source of Mn contamination at the three locations which have high concentration of Mn. Besides, to determine the long term effects of Mn on the general population. More importantly, to vulnerable populations such as women pregnant and children. Based on previous research Mn that enters the body will cause important problems for the body, including neurological health problems, a decrease in children's intelligence, and affect the low weight of baby birth. Therefore the research team suggests that the government also needs to frequently control the level of Mn in well water. Then examined in-depth the causes and consequences together with researchers from the university. Collaboration between the government and academia is also needed in an effort to improve the quality of well water, more specifically to reduce the level of concentration of heavy metal, manganese. The hope is to realize a technology that is easily applied in the community as a solution to the high levels of manganese in well water. The authors, therefore, would strongly suggest the use of bottled water for drinking purposes.

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FIGURE LEGENDS

Fig. 1. The measurement result of heavy metal samples (cadmium, chromium, lead, manganese, and zinc)

Fig. 2. The map of the result of sample locations and level of heavy metals

Fig. 3. The result of cadmium measurement in 101 samples location

Fig. 4. The result of manganese measurement in 101 samples location

Fig. 5. The result of zinc measurement in 101 samples location

Table 1. The results of descriptive analyze

Desc. Analyze	Cd (mg/l)	Cr (mg/l)	Pb (mg/l)	Mn (mg/l)	Zn (mg/l)
Standard	0,003	0,05	0,01	0,4	3
Limit Detection	0,001	0,003	0,004	0,05	0,008
Minimun	< LD	< LD	< LD	< LD	< LD
Maxium	0,002	< LD	< LD	1,800	0,020
Mean	0,000	0,000	0,000	0,578	0,016
St. Deviation	0,000	0,000	0,000	0,652	0,004

< LD = Less then limit detection

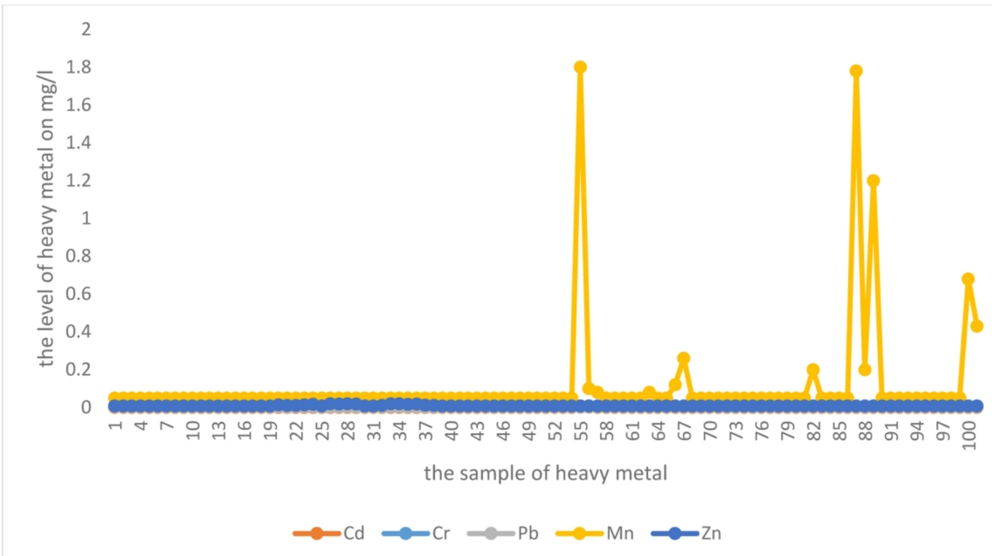

Fig. 1. The measurement result of heavy metal samples (cadmium, chromium, lead, manganese, and zinc)

155x86mm (500 x 500 DPI)

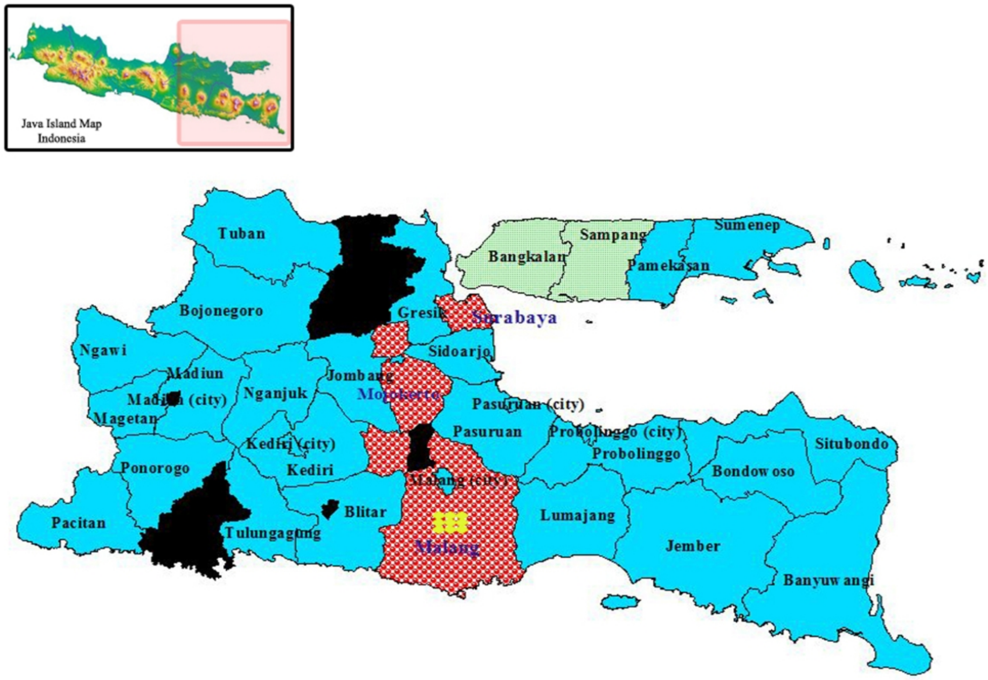

Fig. 2. The map of the result of sample locations and the level of heavy metals

230x159mm (300 x 300 DPI)

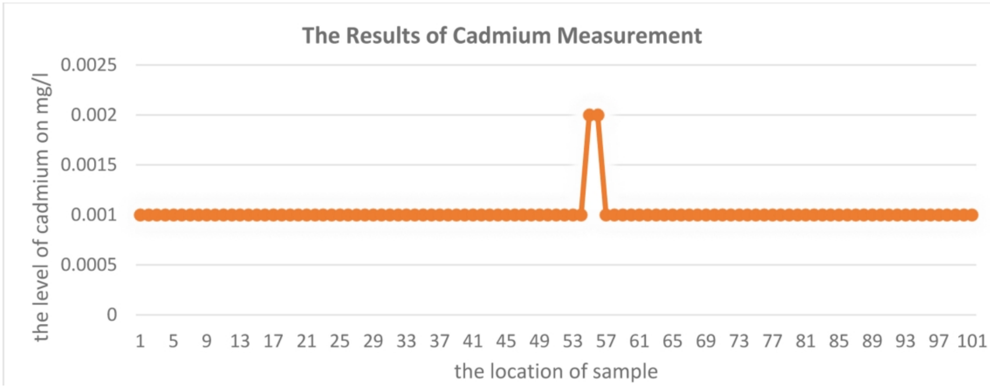

Fig. 3. The result of cadmium measurement in 101 samples location

159x61mm (500 x 500 DPI)

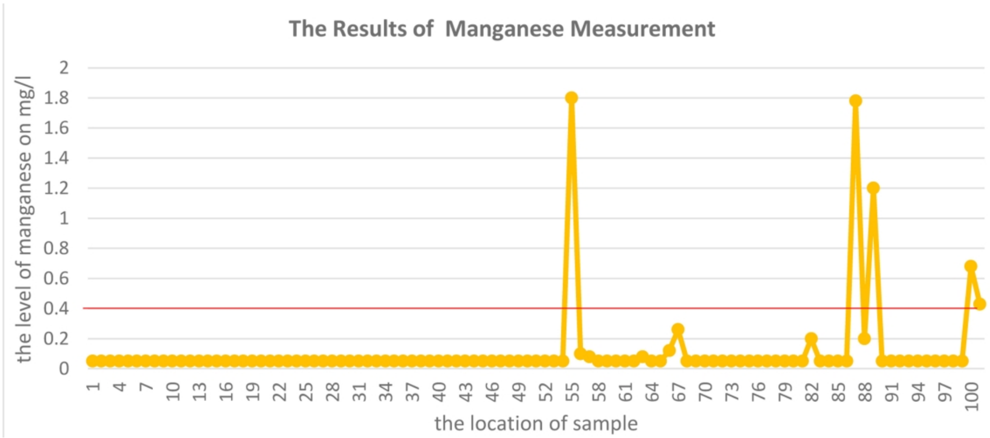

Fig. 4. The result of manganese measurement in 101 samples location

159x69mm (500 x 500 DPI)

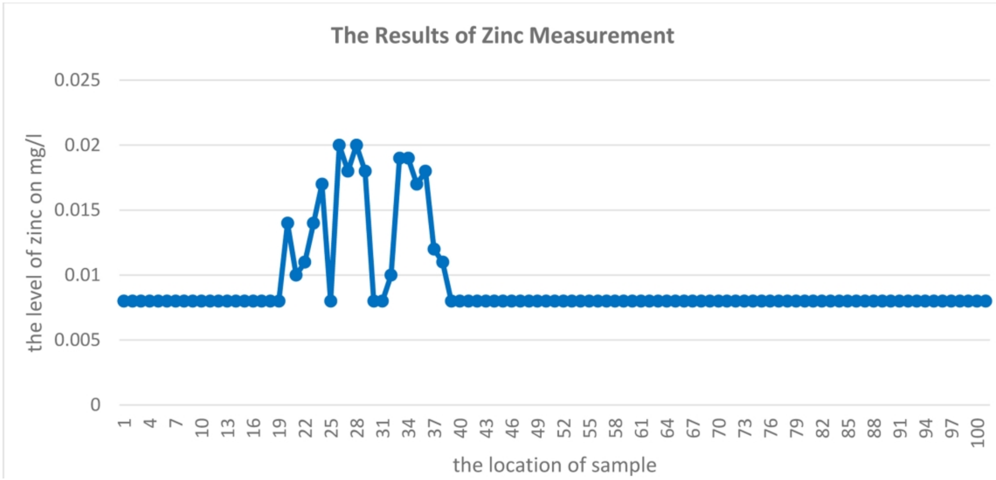

Fig. 5. The result of zinc measurement in 101 samples location

159x75mm (500 x 500 DPI)

Malaysian Journal of Medicine & Health Sciences

Decision Letter (MJMHS-2020-0789.R1)

From: normala_ib@upm.edu.my

To: azizah@fkm.unair.ac.id

CC:

Subject: Malaysian Journal of Medicine & Health Sciences - Decision on Manuscript ID MJMHS-2020-0789.R1

Body: 23-Nov-2020

Dear Dr. Azizah:

It is a pleasure to accept your manuscript entitled "Analysis of Heavy Metals (Cadmium, Chromium, Lead, Manganese, and Zinc) in Well Water in East Java Province, Indonesia" in its current form for publication in the Malaysian Journal of Medicine & Health Sciences. The comments of the reviewer(s) who reviewed your manuscript are included at the foot of this letter.

Thank you for your fine contribution. On behalf of the Editors of the Malaysian Journal of Medicine & Health Sciences, we look forward to your continued contributions to the Journal.

Sincerely,
Dr. Normala Ibrahim
Editor-in-Chief, Malaysian Journal of Medicine & Health Sciences
normala_ib@upm.edu.my

Associate Editor Comments to Author:

Associate Editor
Comments to the Author:
As per reviewer

Reviewer(s)' Comments to Author:

Reviewer: 1

Comments to the Author
Accept

Date Sent: 23-Nov-2020

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