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MERCURY AND CHROMIUM DISTRIBUTION IN SOIL NEAR MAROS KARST ECOSYSTEM

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Abstract: The legal karst protection in Indonesia seems unclear where karst is still seen as a resource that can be utilized, not as an ecosystem that must be preserved. In this study, a total of 20 soil samples were collected around industries and abandoned mine of Maros Karst Ecosystems. Quantitative assessment is done by measuring the factor of single element enrichment and potential biological effects on living things. Calculated Hg accumulation indicates that soil conditions in the area around the Karst Maros Ecosystem have been polluted. In contrast, Cr accumulation means the soil condition remains unpolluted and not harmful.

Keywords: Mercury, chromium, soil pollution, karst ecosystem.

1. INTRODUCTION

Maros is one of the regions with the largest karst resources in the world. Karst is a hill region consisting of arid carbonate rocks. Karst rocks dissolve easily and store abundant water underneath (Arsyad, et al., 2016). It causes karst to be easily damaged and eroded. The cement industry uses karst rock with an abundance of limestone as a raw material for making cement (Ikhsan, et al., 2019; Zhang et al., 2018). Unfortunately, in many developing countries, cement-making fuels still rely on coal (Biksey, et al., 2012). The use of coal with a very high-temperature combination will release some heavy metal particulates (Darmono, 2010; Nordberg, et al., 2015), silica and dust which are harmful to the health of the population around the industrial area (Mallongi et al., 2019). Soil contamination are coming from anthropogenic activity, such as mining industry, vehicle emission, land use and farming (Kurt, 2018).

Mercury is a dangerous heavy metal. The atomic number of this element is 80, and able to spread in various media ranging from rocks, water, soil and air. The presence of mercury is usually found in the vicinity of the battery industry, gold mining and coal mining activities (Hu & Cheng, 2016). Mercury, even in low concentrations, is very dangerous for the environment and human health. Mercury contamination problems can occur naturally and originally from anthropogenic activities. One-third of the total release of mercury arises from human activity (ATSDR, 1999; Marrugo-Negrete, et al., & Díez, 2017). The amount of mercury released from various soil throughout the region depends on the pattern of vegetation, moist, weathering and also the amount of sunlight reached to the ground (Department of the Interior U.S Geological Survey, 2016). Weathering is also influenced by the movement of water and wind (ATSDR, 1999). The presence of mercury in the soil was affected by meteorological factors, proximity to sources of emission and the use of fertilizers in agricultural areas (Huang et al., 2018; Mallongi et al., 2020). Three common forms of mercury are elemental mercury (Hg^0), inorganic mercury (Hg^{+2}) and organic mercury (MeHg) (Kim & Zoh, 2012; UNEP, 2008). Mercury poisoning can lead to health problems such as nerve disorders, skin disorders and cancers (ATSDR, 1999; Sundseth, et al., 2016).

Cr in human causes more deaths in women due to upper gastrointestinal cancer, breast cancer and NHL, but this relationship was not found in men (Núñez et al., 2016). Cr (VI) is a part of the carcinogenic group (group 1), while Cr (III) is determined as non-carcinogenic in humans (group 3) (IARC MONOGRAPH, 2020). Cr in natural environment accumulates in the soil and its concentrations is between 10 and 50 mg/kg (De Vivo, et al., 2004). Cr would reach higher quantity depending on substrate characteristics. For example,

the total of Cr reserve in agricultural land around several cities in Italy was able to be traced back by the natural degradation process of the geological substrate. Exposure to Cr through dust and soil can cause dermatitis, eczema, while high levels of Cr in the blood may cause respiratory problem (Afridi et al., 2011; Hedberg, et al., 2016; Kridin, et al., 2016).

Safe concentrations of Hg and Cr induced in human skin are 0.10 mg/m^3 and 0.50 mg/m^3, respectively. The oral limit is 0.10 mg/m^3 for methyl mercury, 0.10 mg/m^3 for inorganic mercury (ATSDR, 1999; USEPA, 1989) and 0.10 mg/L for Cr (VI). Heavy metal identification sources in agricultural land is a basis for taking appropriate actions to protect soil quality and to develop sustainable management strategies (Lu et al., 2012). Determining the number of heavy metals on agricultural land is crucial because its effect influenced food quality and plantation (Ali, et al., 2019; Song et al., 2018). After 30 years, the concentration of metals in the form of Hg and Cr at the ground surface will slowly increase. It is necessary to monitor the ecological potential that increases the deposits of metals (Liang et al., 2017). The accumulation of heavy metal in the soil can be identified by Contamination Factor (CF), Geoaccumulation Index (Igeo) and Pollution Load Index (PLI) (Hakanson, 1980; Muller, 1981). By determining CF and Igeo, the effect of anthropogenic activities before and after industrialization will be shown.

2. MATERIALS AND METHODS

2.1. Study area

The Maros Karst region is located in northern part of the Bulusaraung National Park (Brumm et al., 2019). This area spends dry season in May-October and rainy season on November-April. This area also has been nominated by World Heritage in 2019. There was pre-historic area called Leang-Leang Cave and the archaeological tourist destination of Karst Rammang-Rammang. The minimum temperature of the study area is 23°C in July and the maximum temperature is 34°C in October. The majority of wind direction are blown to east and southeast in last five years. The sampling location was done around the residential area close to industrial activities that handle karst processing. The area includes Lau District, Bantimurung District and Bontoa District.

2.2. Sampling and procedures

Soil sampling was taken from April 8-10, 2020. In 1998, PT. Semen Bosowa began to produce cement products with an initial capacity of 1.8 million tons/year. In the southeast, there is a former Bosowa Mining company that produces casts, marble and raw materials but is no longer active. The type of soil and rocks around the Karst Maros area is alluvial rocks. A total of 20 soil samples were taken in the east, west and south of the cement plant. Sample was not taken from northern region because it was an empty area where rock is quite high, arid and broad, called Tonasa Formation. There is no residential area in that location. The concerned area was the closest to human settlements and industrial activity areas. Soil sampling distance is adjusted to the closest spot to main emission source which is the cement plant (500-4,000 m). The sampling location is shown in Figure 1.

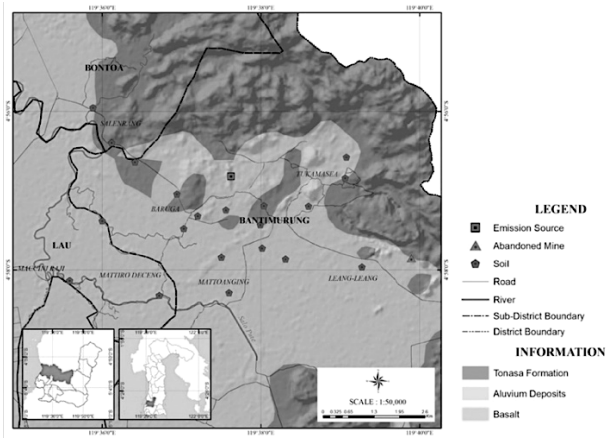

Figure 1. The area of sampling station in Maros Karst Area

Samples were collected by a small hoe. The soil was taken in a rainy season between 0-10 cm from the ground and put into polyethylene bag, labelled and stored in a cooler box with ice immediately after collection. Before determining the concentration of the sample, all samples were dried and separated from leaves, grass and unnecessary rocks. Samples were analyzed and determined at Makassar Center for Plantation Industries (BBIHP). Laboratory quality control was determined by Standard Reference Material (SRM 1646a estuarine sediment) from the Department of Commerce, National Institute of Standards and Technology (NIST) Gaithersburg, MD 20899 with 3 replications. Heavy metal concentrations were determined by using the PinAAcle 900H Atomic Absorbance Spectroscopy (AAS), PerkinElmer. Each sample was weighed in approximately 2 grams and five drops of HNO_3 and 10 mL HCl 1:1 was placed. Samples were heated until dry. Then, it was diluted with hot distilled water and filtered. The concentrations were read in AAS according to the metal wavelength.

2.2.1. Ecological risk assessment

Anthropogenic metal contamination in the soil

was determined quantitatively by EF, CF and Igeo. The contamination factor is the value of pollution status for a single heavy metal. Hakanson (1980) suggested a model to assess a contamination and the degree of contamination of soil. In Table 1, Hakanson (1980) revealed the pre-industrial value of each heavy metal for assessment of heavy metal accumulation. The value of CF was calculated as follows:

$$C_f^i = \frac{C_0^i}{C_n^i}$$

*w*here C_0^i mean result of heavy metals from at least five sampling sites. The C_n^i is background site of each heavy metal (preindustrial). The reference value can be used from the pre-industrial values of several lakes in Europe and America, as in the following table:

Table 1. Pre-industrial reference value (µg/g) and toxic response factors according to Hakanson.

Heavy Metal	Hg	Cd	As	Cr	Zn
Preindustrial reference	0.25	1.0	15	90	175
Toxic response value (Tr)	40	30	10	2.0	1.0

Values obtained from the CF result are grouped in some categories: low (CF <1), moderate ($1 \leq CF < 3$), considerable ($3 \leq CF < 6$), dan very high ($CF \geq 6$). Muller (1969) determines soil contamination in industrial zones with geo-accumulation index (I_{geo}). The enrichment of metal concentrations in the soil is based on sample concentrations and background values. Classification of the level of metal pollution can be divided into 7 levels: (1) $I_{geo} \leq 0$: uncontaminated, (2) $0 < I_{geo} < 1$: uncontaminated to moderately uncontaminated, (3) $1 < I_{geo} < 2$: moderately contaminated, (4) $2 < I_{geo} < 3$: moderately to heavily contaminated (5) $3 < I_{geo} < 4$: heavily contaminated (6) $4 < I_{geo} < 5$: heavily to extremely contaminated, dan (7) $5 \leq I_{geo}$: extremely contaminated. Geo-accumulation index can be determined by the following equation:

$$I_{geo} = \log_2 (C_n / 1.5\ B_n)$$

Where Cn is the concentration of the element to be measured, Bn is the background geo-accumulation value of the earth's crust and 1.5 is the variation value in certain substances in the environment and affects the local anthropogenic sources (Muller, 1981). The baseline values of several heavy metals are Cr 56.97 mg / kg, Cd 0.26 mg / kg, Pb 24 mg / kg, Mn 75.6 mg / kg, As 82.1 mg / kg and Hg 4.89 mg / kg from soil in karst area (CNEMC (China National Environmental Monitoring Centre), 1990; Xiao et al., 2019).

2.2.2. Hazard quotient (hq)

Hazard quotient is an occurred level of the potential toxicity of certain chemicals in the study area. This value is an estimated ratio of exposure to concentrations that is considered to represent concentrations/doses that are safe for the environment. If the HQ value <1, there are no health effects derived from chemical exposure. But if HQ> 1, then these chemicals may be possibly harmful to health. Based on EPA (1997), the value of HQ is determined by the following equation:

$$HQ = \frac{SCC}{TD}$$

where SCC is a sediment concentration in mg/kg, toxicity benchmarks is a reference dose where human is expected to have lower concentrations for fewer health effects (ATSDR, 1999; Feng et al., 2011; Syahirah et al., 2019). If HQ <0.1; no hazard exists, HQ 0.1-1; hazard is low, HQ 1.1-10; hazard is moderate and HQ > 10; hazard is high. Heavy metal was assumed taken up from soil. Receptors, a plant and human, will be affected by the presence of metals. If it is consumed, it will be harmful to health. Toxicity dose for Cr in soil residential area is 220 mg/kg (Cr VI) (Canadian Environmental Quality Guidelines, 1999) and Hg is 30,1 mg/kg for consumption produce and attached soil (Environmental Agency, 2004).

3. RESULTS AND DISCUSSION

3.1. Quantity of heavy metal in soil near Maros Karst Ecosystem

Various concentration of Hg and Cr is shown in Table 2. The values obtained at all locations cannot be read by the instrument, especially for Cd and Pb metals. Therefore, the concentration value of Cd and Pb metals is eliminated. The average concentration of Hg and Cr were 60 mg/kg and 48 mg/kg respectively. The highest concentration of Hg was on station 9 (163 mg/kg) and highest concentration of Cr was in station 4 (95 mg/kg).

3.2. Geo-accumulation index

Muller (1981) classified the quality of sediment to explain heavy metal pollution and environmental risk condition. In Figure 2, both metals concentration is higher in agriculture land than non-agriculture land. The accumulation of Hg and Cr are predicted to come from a huge cement industry around the study sites. Near Leang-Leang, a pre historic cave, an abandon mine building was not so far from the residential area. The loading activity,

transportation, fertilization and mining stone have a potential as a source of polluted heavy metal in soil.

In Figure 3, all locations have different accumulated values. Hg accumulation at station 9 is the highest in Igeo value (6.69) followed by station 13, which is extremely contaminated. Station 1, 10, 7 (heavily contaminated); Station 11, 2, 19, 6, 17, 3, 18 (moderately to heavily contaminated); Station 8, 5, 15, 4 (moderately contaminated); Station 14, 12,16 dan 20 (uncontaminated). Meanwhile, the highest Cr geo-accumulation index was at station 4 with Igeo value (0.43), which is uncontaminated to moderately uncontaminated and the rests of them is classified as uncontaminated (Igeo<1). Geo-accumulation index of the two metals can be seen from Figure 3.

Table 2. Heavy metal quantity (mg/kg) and CF Around Maros Karst Ecosystem.

Sampling location	Land Use	Concentration (mg/kg dry)		Contamination Factor (CF)	
		Hg	Cr	Hg	Cr
1	NAL	82	63	326	70
2	AL	71	86	285	96
3	NAL	52	48	210	53
4	AL	29	95	114	106
5	NAL	38	84	150	93
6	AL	60	92	242	102
7	AL	81	77	324	86
8	NAL	39	19	154	21
9	AL	163	18	652	20
10	NAL	83	49	333	55
11	AL	72	43	287	48
12	NAL	20	41	80	45
13	AL	145	37	579	41
14	NAL	23	28	94	31
15	AL	36	51	144	56
16	NAL	20	44	80	49
17	AL	56	36	223	40
18	NAL	52	27	209	30
19	AL	60	32	241	36
20	NAL	13	0.60	51	0.70
Average		60	48	238	54
Min		13	0.60	652	106
Max		163	95	51	0.70
Std. Deviation		39	26	156	30
p (*Kolmogorov-Smirnov*)		0.10	0.17	0.11	0.12
Distribution type		N	N	N	N

AL: agricultural land, NAL: non-agricultural land, N : normal

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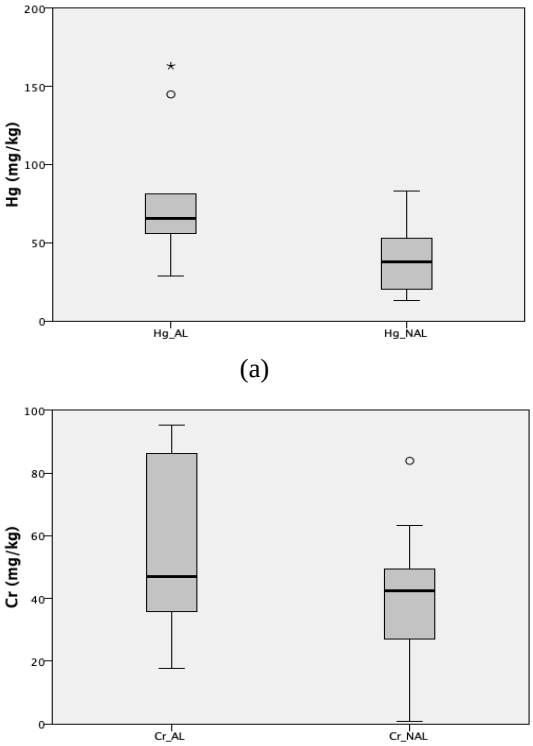

Figure 2. Variation values of Hg (a) and Cr (b) at agriculture and non-agriculture land

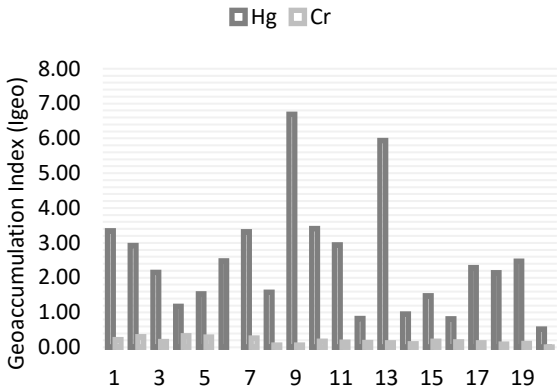

Figure 3. Geo-accumulation of Hg and Cr in soil of each stations

3.3. Hazard Quotient of Hg and Cr in Soil

Dangerous level of contamination to health in certain areas is obtained from the media concentration and toxicity limits which can cause health effects. In Table 3, the highest HQ_{Hg} value resulted from station

9 (5.4) and station 13 (4.8) with moderate status. The lowest HQ_{Hg} came from station 20 (0.43) which is the furthest area from the source of cement emissions and limestone mining activities. HQ_{Cr} in all stations were less than 1, so HQ_{Cr} in all stations were low and has no adverse health effects. Hg possibly had a much higher health risk than Cr in all sampling locations in rainy season in this area.

Table 3. Hazard Quotient Values of Hg and Cr

Sampling Location	HQ	
	Hg	Cr
1	2.7	0.28
2	2.3	0.39
3	1.7	0.21
4	0.95	0.43
5	1.2	0.38
6	2.0	0.41
7	2.7	0.35
8	1.2	0.08
9	5.4	0.08
10	2.7	0.22
11	2.4	0.19
12	0.67	0.18
13	4.8	0.16
14	0.78	0.12
15	1.2	0.23
16	0.66	0.20
17	1.8	0.16
18	1.7	0.12
19	2.0	0.14
20	0.43	0.01

3.4. Geospatial Hg and Cr in soil near Maros Karst Ecosystem

Pollutant distribution map has shown metal accumulation in study area. This two-dimensional shape can identify the concentration of various pollutants by color and pattern. In this study, the highest distribution of Hg was collected in the Lau, Tukamasea and Bantimurung regions that is located in the west and east regions. Cr is mostly accumulated around the area of cement factories such as Bantimurung, Baruga and Tukamasea. The concentrations of the two metals have different variations for each station. Location with the highest acquisition and above the levels commonly was in the same place. This indicates that the origin of pollutants in study area comes from the same source.

Industrial activities still active in this region are cement industry and limestone mining. On the southeast, there are an abandoned mine and large rice fields while in the west an area that is crowded with transportation (vehicle, loading activity and main roads). Cement industry in this area is related to the use of coal combustion which is able to produce higher emissions and provide toxic particulates into the atmosphere and then carried by the wind. The dominant wind blows in this region are shown in Figure 5. In addition, agricultural areas have higher heavy metal accumulation rates. It might be influenced by the use of fertilizers and metal intrusion factors in the soil.

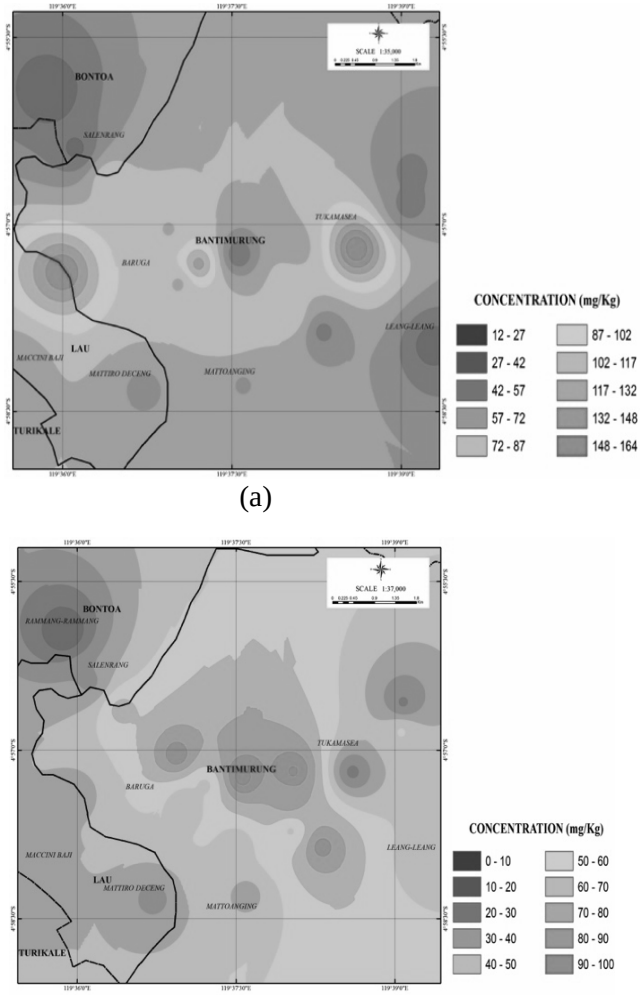

Figure 4. Spatial distribution of Hg (a) and Cr (b) in soil of study area, obtained by GIS Software

Based on the dominant direction of the wind blows in the rainy season for the past five years, it is shown that the region with the most dominant blowing direction is the east and southeast regions. This is consistent with the distribution of pollutants in

Figure 3, where the highest Hg and Cr metal concentrations are in both regions. Except for the Hg concentration in the west where this area is close to the main road. It is assumed that the accumulation of Hg metal in the region comes from vehicle emissions. Hg can spread and stay in the atmosphere for about 7 days (WeissPenzias, et al., 2003). This element is very dangerous and if it exceeds the body's limits, the carcinogenic risks will appear. Hg accumulation in soil media is very prohibited because soil is a medium of rice growth which is the majority crops in the area (Clemens & Ma, 2016).

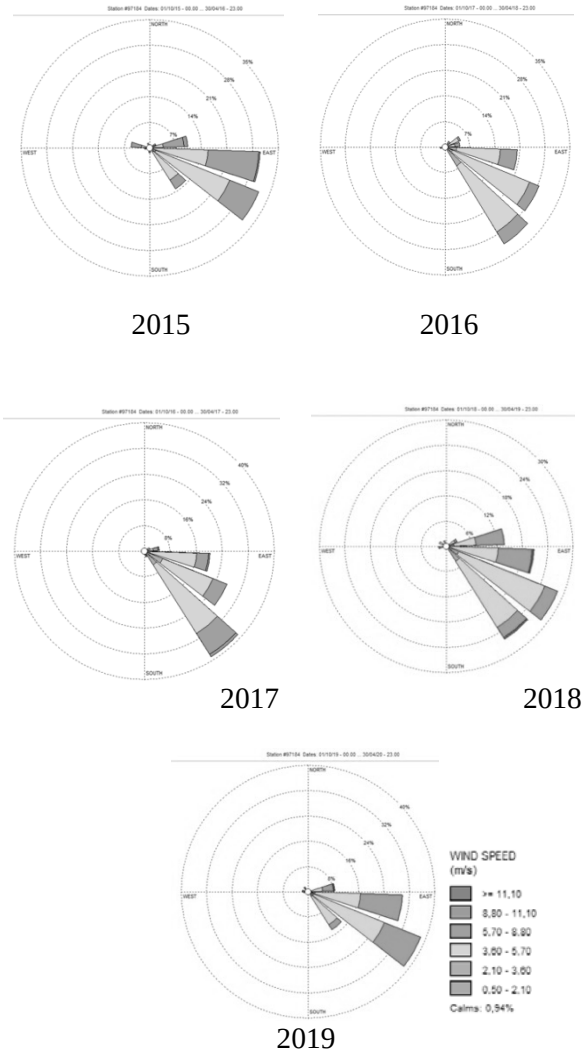

Figure 5. Windrose plot (2015-2019) obtained by *Lakes Environmental Software*

Most of the Hg accumulation in rice is in the root which is the part that is directly in contact with the soil. The use of inorganic fertilizers may be an addition to the quantity of Hg and Cr metals in agricultural areas. This implied that the accumulation of the two metals is higher in the agricultural region than in non-agricultural regions. Further research on the quality of agricultural soils and their effects on rice needs to be done immediately.

Figure 4 shows the location with the highest accumulation of Hg is in Tukamasea. The location is limited by confined wall of karst in the west so that pollutants will be carried by the wind and will likely blocked and fall in the area. High levels of Hg are also found in the Lau region, where this location is the main transportation route bordering to agriculture. Cr mostly accumulated around the cement industry and the highest levels are in Bantimurung area. Cr compound is a typical pollutant in the vicinity of the cement industry. This element will affect the quality of human health, especially for breathing and skin. Frequent exposure of Cr will increase the risk of dermal disorders, cough, eczema, dermatitis and skin cancer. People who do outdoor activities will be very vulnerable to the Cr exposure, especially for farmers and mining employees. In addition, children become vulnerable to Cr exposure due to playing activities on the ground media or even accidentally swallowing the dust or particles.

4. CONCLUSION

The concentration, accumulation and spatial distribution of Hg and Cr in soil were determined near Maros Karst ecosystem. Because the concentration of Hg in soil was high, the area is classified as heavily polluted. For Cr, the major concentrations were found near cement industry area. Except in station 20, all locations contain a high contamination factor (CF) of Hg and Cr in soil and there was a connection between industrial activity with soil pollution status. Igeo values for each station showed the different results in land use classification. In agricultural land, the accumulation of both metals is higher than non-agricultural land. Based on this data, we could assume that fertilizer usage had an important role for metals accumulation. For health effects, it was found that the accumulation of Hg in the soil would have a serious health impact although this must be proven by a health risk assessment. Whereas, HQ values for Cr accumulation in soil is still harmless and can be tolerated. More research is needed to track dispersion and pollution sources of some heavy metals in several media such as water and air, so it can describe the quality of the karst environment precisely.

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