

# Evaluation of Heavy Metal Exposure to Soil and Paddy Plant around the Closed Municipal Solid Waste Landfill: Case Study at Gunung Tugel Landfill, Banyumas-Central Java

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**Abstract.** This work was focused on assessing the exposure of heavy metal from closed municipal solid waste (MSW) landfill on soil and paddy plants. This study aimed to determine heavy metal content whether at the soil in the around Gunung Tugel landfill included and accumulated in the paddy plant tissues. The investigated metals include chromium (Cr), copper (Cu), cadmium (Cd), iron (Fe), and zinc (Zn). The samples were acid-digested before the desired elements were measured using Atomic Absorption Spectrophotometry (AAS). The results are presented as distribution map of the landfill area based on the total heavy metals content distribution in the soil and paddy plants. The samples shown that the concentrations of heavy metals around Gunung Tugel landfill are 6.27-34.71 mg/kg, 0.17-0.42 mg/kg, 28.29-48.69 mg/kg, 18,997.26-32,572.29 mg/kg, 342.74-834.49 mg/kg, 136.10-290.14 mg/kg at the top soil and 0.00-1.70 mg/kg, 0.00-0.26 mg/kg, 0.79-10.46 mg/kg, 13.88-61.46 mg/kg, 18.79-50.56 mg/kg, 87.27-273.22 mg/kg at the paddy for Cr, Cd, Cu, Fe, Mn, and Zn respectively. According to the results, The Gunung Tugel landfill is not a direct source of heavy metal pollution at paddy plant in the landfill area, but through surface water and soil media. Rainfall around landfill is quite high ie more 2000 mm/year of rainfall and soil permeability is 1.0 cm/sec.

**Keywords:** MSW, Landfill, Heavy metal, Exposure

## 1. Introduction

In the developing countries of Asia, due to the lack of technical knowledge, economic and infrastructural arrangements, 70-90 % of the solid waste is disposed in open dumps [1]. The MSW in landfill have been generating of leachate which is polluting the environment. In most landfills, leachate is composed of the liquid that has entered the landfill from external sources, such as surface drainage and rainfall and the liquid produced from the decomposition of the wastes, if any [2]. Organic and inorganic pollutants, including heavy metals are the major categories of pollutants [3]. Unlike other pollutants, heavy metals are hazardous, since they cannot be degraded by



biological or chemical means leading to harmful effects on ecosystems, including bioaccumulation [4].

Heavy metals are pollutants that are classified as toxic to the environment, even at minute concentrations. Although different sources of heavy metals exist, metals also occur naturally in the environment as ores and few other complexes; hence, several metals play useful roles in the environment, particularly in plant growth [5-7]. However, when heavy metals exist in excessively high concentrations, pollution occurs and can be catastrophic because of its effect on microbes.

The problem of heavy metal pollution is becoming more serious with increasing industrialization and the disturbance of natural biogeochemical cycles. Unlike organic compounds, heavy metals are essentially non-biodegradable and, therefore, accumulate in the environment. The accumulation of heavy metals in soils and waters poses a risk to the environmental and human health [8]. Landfills accumulate great amounts of waste and are an important source of liquid effluents, named leachates, which can have an adverse impact on the environment when released in an uncontrolled manner [9].

Heavy metals cause toxicities and other harmful effects not only in humans and animals but also in plants and soil microorganisms. Heavy metals disrupt many biochemical and physiological activities in bacteria, including growth, development, enzyme and hormone production. Indole acetic acid (IAA) is one of the most important hormones in plants, which is secreted by both bacteria and plants [10]. Plants are under the continual threat of changing climatic conditions that are associated with various types of abiotic stresses. In particular, heavy metal contamination is a major environmental concern that restricts plant growth. Plants absorb heavy metals along with essential elements from the soil and have evolved different strategies to cope with the accumulation of heavy metals [11]. The metals only contaminate the environment for now since their concentrations are below permissible limits. Solid waste that deposited on the surface adds metallic contaminants to the soil on which economic crops are cultivated. It is necessary to undertake regular environmental impact study to assess waste dumpsites. The higher concentration of copper in vegetation than in soils is attributed to direct contact rather than absorption from the soil [12].

The objective of this study was to determine the total concentration of heavy metals in rice from paddy plant growing in contaminated sites and to investigate the exposure distribution of selected heavy metals from rice from paddy plan and soils.

## **2. Materials and methods**

### *2.1. Study Area*

The soil and paddy plant samples were collected from a landfill site in Banyumas Region in Central Java Province, Indonesia named Gunung Tugel Landfill, as shown in Figure 1. The investigated landfill is located in the Kedungrandu Village, Patikraja Sub Region. This landfill was operated in 1983 with capacity around 282 m<sup>3</sup> per day, and is being used to dispose of mixed municipal waste and the disposal method is open dumping method.

The type of soil in Gunung Tugel landfill is ultisol, thus allowing leachate to seep and pollute the groundwater of the surrounding population [13]. Ultisol is a land with a sourly argill horizon with low base saturation. The saturation of the base at a depth of less than 1.8 m from the soil surface is <35%. This soil texture is clay to sandy clay, bulk density between 1.3-1.5, and slow to moderate permeability [14]. Meanwhile, that Ultisol soil reaction is generally acid to very acid (pH 3.1-5.0) [15].

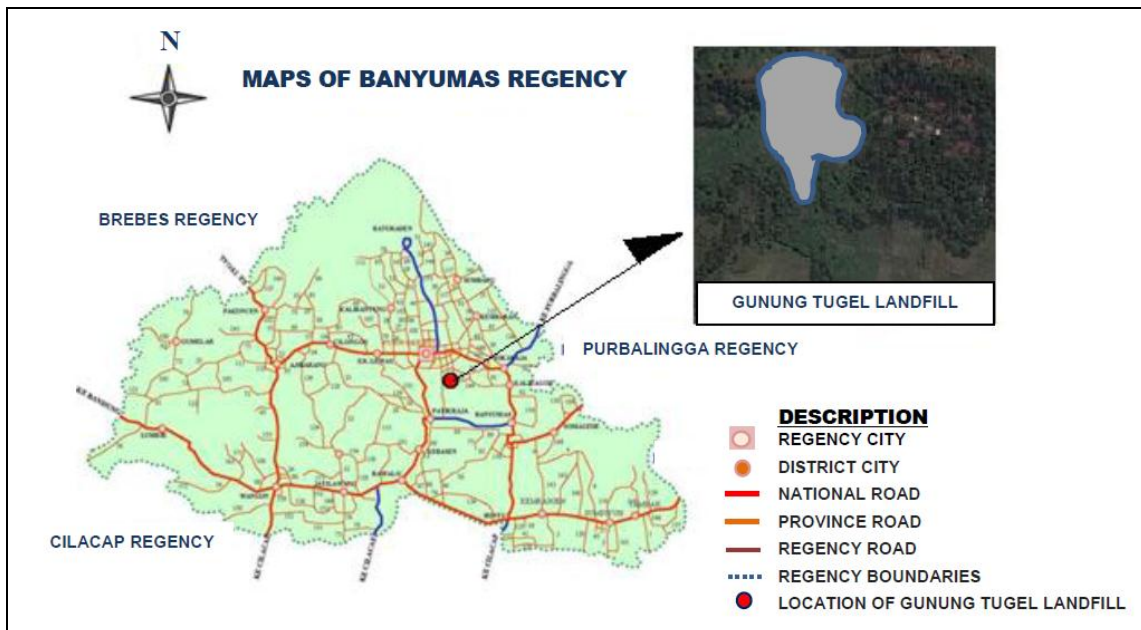


Figure 1. Location of Gunung Tugel Landfill

## 2.2. Sampling and Sample Preparation

The Sample was divided in 2 parts; soil and rice samples. Soil sampling method used in this research is rigid grid method for landfill area and stratified method for rice field area. Rigid grid method is a method that determining sampling point based on coordinate line which form square (grid) with interval distance 60 x 60 m with depth 50 cm. Soil samples were collected using stratified method. This method based on topography condition divided by zone in paddy field area with depth 50 cm. From the observation by using Quantum Geographic Information System (QGIS) software, 12 sampling points cover the area around landfill that has been closed and 6 sampling points covering rice field area. The location of the sampling point to be used can be seen in Fig. 2.

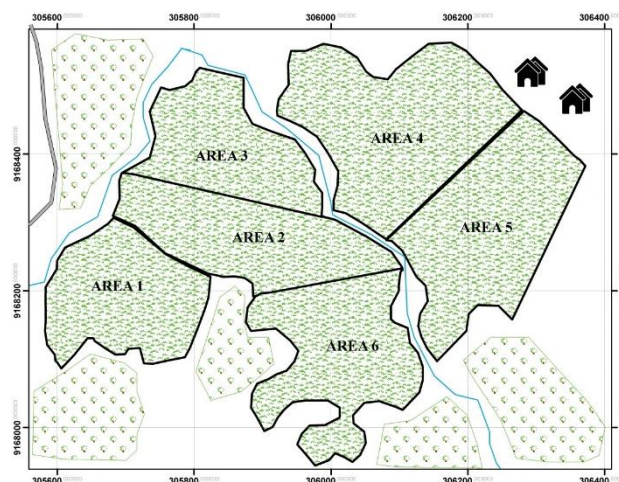


Figure 2. Sampling Point: Soil and Rice Sample in Paddy Field

Soil samples from each sample point were taken about 500g. Meanwhile, for the rice, samples were taken about 290-600g. Soil samples were dried under the sun for one day. Five hundred grams of dried samples were crushed and sieved using 100 mesh. One gram of each fine soil sample was destruct by adding 100 ml aquades and 10 ml of HNO<sub>3</sub> (P) mixture. The destruction was conducting until the samples was clear using using electric stove under the fume hood. The solution was filtered with filter Ø 200 mm, Ø 150 mm and sieve Ø 70 mm, then diluted again with aquades to 100 ml then shaken until homogen and added again with 5 ml of HNO<sub>3</sub> (P), then make destruction again by using electric stove in the cabinet acid until clear, after the solution is clear then the solution filtered with Ø70 mm sieve, added aquades up to 25 ml then the solution shaken until homogeneous and allowed to reach room temperature then stored in sterile bottles before heavy metal analysis. Analysis of heavy metals by AAS were performed at wavelength of 228,80 nm, 228,80 nm, 324,80 nm, 248,30 nm, 279,50 nm, 213,90 nm, 217 nm for Cr, Cd, Cu, Fe, Mn, Zn, Pb respectively.

### 2.3. Analytical Methods

Heavy metal parameters to be analyzed for presence in soil and rice samples are Cu, Fe, Mn, and Zn which are the essential metals, and Cr, Cd, and Pb which are non-essential metals for biological systems of living things. Heavy metals in a sample can be analyzed by AAS flame [16].

### 2.4. Calibration Curve

The first thing to do is preparing 6 (six) 100-ml flasks. Then each flask is filled with 0; 0.5; 1.0; 1.5; 2.0 and 2.5 mL of 1000 ppm Chromium standard solution, each of the flasks was added with aquades until half of the volume of the flask, then the solution was repeatedly repelled, and aquades were added to the boundary marker. The absorbance of each standard solution was measured using an Atomic Absorption Spectrophotometer on a wavelength ( $\lambda$ ) of 228.80 nm. This procedure was repeated for Cadmium, Copper, Iron, Manganese, Zinc and Lead calibration curves using the parent solution of Cadmium, Copper, Iron, Manganese, Zinc and Lead 1000 ppm and 228.80 nm, 324.80 nm, 248.30 nm, 279.50 nm, 213.90 nm, 217.00 nm, respectively.

### 2.5. Data Analysis

The Analysis of the data is obtained by mapping of heavy metal dispersion. The mapping is done by using Quantum Geographic Information System (QGIS) software. After knowing the concentration of heavy metals at each sampling point then determine the plotting based on the quality standard of heavy metal, where each sampling point will represent the area inside, so it can assist in the analysis of the distribution of heavy metals in the soil.

There are three international standards that are used for comparison of heavy metals analysis of Cd, Cu, Cr, Zn, and Pb in soils, namely Environmental Protection Authority of Australia (EPAA) in 2012, Environmental Protection Ministry of China (EPMC) 2015. While the quality standards for heavy metals Fe and Mn use the quality standards of the United States Environmental Protection Agency (US EPA) in 2012. In other haand, for rice sample, standard for Cr and Cd are from National Food Safety Standard of China 2012. Cu, Zn, and Pb from Food Standards Australia New Zealand 2002. The last, for Fe and Mn from USDA Food Compotition Databases 2017.

Each area is recorded in coordinate with UTM (Universal Transverse Mercator) format. The coordinates will be displayed on the map, so that it can be known that the location of high or low hazard according to the class determined according to the respective quality standards of the heavy metals tested by the classification using the scoring method. The classification of concentration levels used in the scoring method can be seen in Table 1.

After the concentration level classification in each parameter of heavy metals, then done overlay with scoring method shown in Table 1. Each area determines the degree of heavy metal content by using the number of scores from each parameter. It aims to determine the area containing the

highest to the lowest heavy metals. Score classification to determine the concentration with the largest classification to the smallest of each sampling point can be seen in Table 2.

Table 1. Concentration Level Classification

| Class     | Colour      | Score |
|-----------|-------------|-------|
| Very Low  | Dark Green  | 1     |
| Low       | Light Green | 2     |
| Medium    | Yellow      | 3     |
| High      | Orange      | 4     |
| Very High | Red         | 5     |

Table 2. Classification and Score of Exposure Concentration

| Class     | Colour      | Score   |
|-----------|-------------|---------|
| Very Low  | Dark Green  | < 7     |
| Low       | Light Green | 8 – 14  |
| Medium    | Yellow      | 15 – 21 |
| High      | Orange      | 22 – 28 |
| Very High | Red         | > 29    |

### 3. Result and Discussion

#### 3.1. Soil in Landfill and Paddy Field

Based on testing of samples taken both soil and rice from paddy field around the land of Gunung Tugel landfill indicate heavy metal content with various concentration. Table 3 and 4 shows that the soil and rice around Gunung Tugel landfill has been exposed with heavy metals. It is known that the concentration range of Cd from 0.17 - 0.42 mg/kg with an average value of 0.32 mg/kg. While the concentrations of other heavy metals are 24.03 – 48.69 mg/kg, 971- 34.71 mg/kg, 18997.26 mg/kg, 342.74 – 834.49 mg/kg and 136.10 – 290.00 mg/kg for Cu, Cr, Fe, Mn, and Zn, respectively. There are parameters of heavy metals in the soil exceeding:

Table 3. Concentration of Heavy Metals in Soil around Gunung Tugel Landfill

| Location | pH   | Concentration of Heavy Metals (mg/kg) |       |       |          |        |        |
|----------|------|---------------------------------------|-------|-------|----------|--------|--------|
|          |      | Cd                                    | Cu    | Cr    | Fe       | Mn     | Zn     |
| Area 1   | 6.00 | 0.37                                  | 40.55 | 34.71 | 18997.26 | 342.74 | 163.76 |
| Area 2   | 6.00 | 0.42                                  | 39.47 | 18.61 | 19217.73 | 636.20 | 136.10 |
| Area 3   | 5.00 | 0.37                                  | 48.69 | 16.41 | 32572.30 | 709.78 | 170.72 |
| Area 4   | 5.00 | 0.25                                  | 28.29 | 6.27  | 31151.74 | 629.69 | 190.19 |
| Area 5   | 6.00 | 0.35                                  | 24.03 | 14.53 | 19564.74 | 552.37 | 188.91 |
| Area 6   | 6.00 | 0.17                                  | 37.12 | 9.71  | 19639.77 | 834.49 | 290.14 |
| Average  | 5.67 | 0.32                                  | 36.36 | 16.71 | 23523.92 | 617.54 | 189.97 |

Table 4. Concentrations of Heavy Metal On Rice from Paddy Plant around Gunung Tugel Landfill

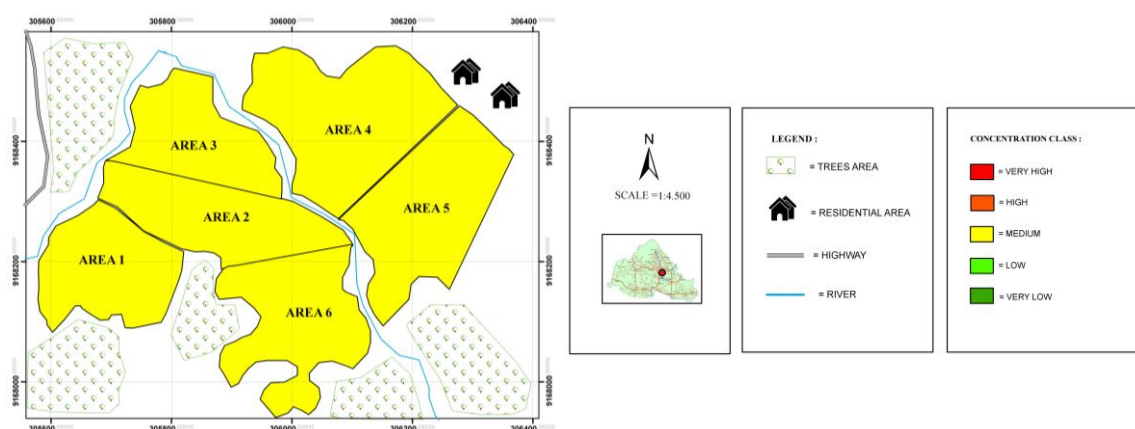
| Location | Concentration of Heavy Metals (mg/kg) |      |       |       |       |        |
|----------|---------------------------------------|------|-------|-------|-------|--------|
|          | Cr                                    | Cd   | Cu    | Fe    | Mn    | Zn     |
| Area 1   | 0.00                                  | 0.05 | 1.37  | 21.06 | 45.66 | 273.22 |
| Area 2   | 0.88                                  | 0.13 | 2.55  | 18.99 | 50.56 | 115.21 |
| Area 3   | 1.70                                  | 0.04 | 10.43 | 26.80 | 49.73 | 149.82 |
| Area 4   | 0.76                                  | 0.00 | 2.05  | 20.12 | 39.16 | 88.06  |
| Area 5   | 1.32                                  | 0.04 | 1.06  | 61.46 | 21.10 | 87.27  |
| Area 6   | 0.57                                  | 0.26 | 0.79  | 13.88 | 18.73 | 87.43  |
| Average  | 0.87                                  | 0.09 | 3.04  | 27.05 | 37.49 | 133.50 |

Table 5 shows the concentration level score of the map results of each parameter of heavy metals according to quality standard:

Table 5. Score and Classification of Heavy Metal Concentration Level on Soil in Paddy Plant Area

| Location | Each Parameter Score |    |    |    |    |    |    | Total | Class  |
|----------|----------------------|----|----|----|----|----|----|-------|--------|
|          | Cd                   | Cu | Cr | Fe | Mn | Zn | Pb |       |        |
| Area 1   | 3                    | 4  | 3  | 2  | 1  | 4  | 1  | 18    | Medium |
| Area 2   | 3                    | 4  | 2  | 2  | 2  | 3  | 1  | 17    | Medium |
| Area 3   | 3                    | 4  | 2  | 3  | 2  | 4  | 1  | 19    | Medium |
| Area 4   | 2                    | 3  | 1  | 3  | 2  | 4  | 1  | 16    | Medium |
| Area 5   | 3                    | 2  | 2  | 2  | 2  | 4  | 1  | 16    | Medium |
| Area 6   | 2                    | 4  | 1  | 2  | 2  | 5  | 1  | 17    | Medium |

Figure 3 shows a map of the overlay and classification of heavy metal concentration levels on soils in the landfill and paddy field according to score level. The concentration of heavy metals in the soil is in the medium grade with a score range of 16 to 19, indicating that the overall heavy metal concentration on the soil in the paddy field area is evenly distributed. The concentration of heavy metals in the soil in the paddy fields is not only from the landfill, but can result from excessive use of fertilizers and pesticides. This can be seen in area 1 which is not adjacent to the landfill but has a score of 18 higher in areas other than in area 3. Area 1 uses more river water adjacent to the area, but there is a possibility that leachate water polluting area 3 flows into area 1 and accumulates in the area, so that the concentration of heavy metals in the soil in area 1 results from the accumulation of fertilizer and pesticide use and irrigation water that has been contaminated with leachate water. While in area 3, most likely pollution occurs due to leachate water from landfills that contaminate surface water used for irrigation. The exposure of heavy metals in the paddy fields is evenly distributed, since concentration scores are not too far between areas and have only moderate classes overall. The concentration level of heavy metals in the soil in the area of medium paddy fields can also be caused by the absorption of heavy metal content to rice crops, so that heavy metal content can be more accumulated and pollute rice plants.



**Figure 3.** Map of Heavy Metal Exposure of Soil on Paddy Plant

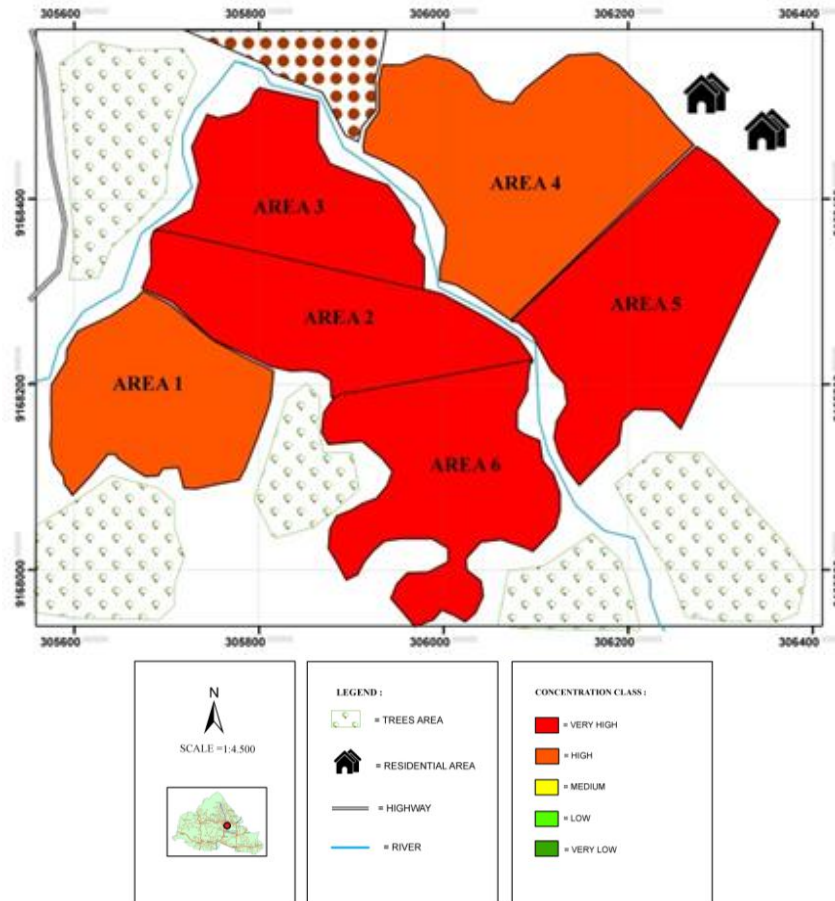
This mapping aims to show which areas have the highest contamination levels to the smallest, it is based on map overlay of each heavy metal parameter that has been classified using the scoring method. The following table shows the concentration level score based on the map of each heavy metal parameter in the rice sample:

**Table 6.** Scoring Result of Heavy Metal Concentration Level on Rice Samples

| Area   | Each Parameter Score |    |    |    |    |    |    | Total | High      |
|--------|----------------------|----|----|----|----|----|----|-------|-----------|
|        | Cr                   | Cd | Cu | Fe | Mn | Zn | Pb |       |           |
| Area 1 | 1                    | 1  | 5  | 5  | 5  | 5  | 3  | 25    | High      |
| Area 2 | 4                    | 3  | 5  | 5  | 5  | 5  | 3  | 30    | Very High |
| Area 3 | 5                    | 1  | 5  | 5  | 5  | 5  | 3  | 29    | Very High |
| Area 4 | 4                    | 1  | 5  | 5  | 5  | 5  | 3  | 28    | High      |
| Area 5 | 5                    | 1  | 5  | 5  | 5  | 5  | 3  | 29    | Very High |
| Area 6 | 3                    | 5  | 3  | 5  | 5  | 5  | 3  | 29    | Very High |

Based on Table 6, area 2 is the area with the highest number of scores of 30, included in the category of very high contamination level. However, other areas of 3, 5 and 6 are also in the same category. This is further illustrated by the map in Figure 4 which shows that all areas have a red

color which means the area has a score between 29 – 35. The relatively similar hazard level indicates that the source of heavy metal contamination in rice is not only from landfill, but from other sources such as excessive use of fertilizers and pesticides by farmers. Evident from the level of hazard area 1 located in zone A with irrigation location is before the landfill is equal to area 4 located in zone B with irrigation location is located after the landfill landslide marked with high color orange category.



**Figure 4.** Map of Heavy Metal Exposure of Rice in Paddy Plant

#### 4. Conclusions

Based on the results of analysis and discussion of this study can be concluded the following points:

- 1, Soil and paddy plants around Gunung Tugel landfill have been exposed to heavy metals with varying degrees. The average concentration in soil are 0.3236.36, 16.71, 23523.92, 617.54, 189.97 mg/kg and paddy plant are 0.87, 0.09, 3.04, 27.05, 37.49, 133.50 for Cd, Cu, Cr, Fe, Mn, Zn mg/kg respectively.
- 2, Overlay of heavy metal content on the soil in the landfill area has medium class and the paddy plant has high to very high. Exposure of heavy metals in the soil tends to be higher in the center of the landfill area, but in the paddy plants more evenly distributed. It can be caused the absorption of heavy metal to rice plants, so that accumulated in rice plants

## 5. Acknowledgement

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