

Identification of Pb distribution and pollution potential on agricultural land in Poncokusumo Sub-district, Malang Regency, East Java, Indonesia

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Abstract. This study aims to identify the distribution of Pb concentrations in several areas of intensive vegetable plantations in the use of agrochemical materials in the Poncokusumo sub-district and their absorption potential in vegetable crops grown in the area. This study used a survey method in three villages known as the center of vegetable production in Poncokusumo sub-district. The results showed that the average distribution of Pb concentration in Wonorejo village namely in the range of 11.35-11.62 mg kg⁻¹ identified was higher than in Belung village (range 0.73-3.15 mg kg⁻¹) and Pandansari (range 1.85-9.49 mg kg⁻¹) either during the land use range of 5, 10 and 15 years. Some sources of production inputs used by farmers from irrigation water, organic fertilizers, inorganic fertilizers and pesticides are also identified to contain Pb metal, although the average Pb concentration in these materials is still below the maximum threshold specified. The average Pb concentration in some types of vegetables grown in the area, is above the maximum limit of Pb contamination in fruits and vegetables namely 0.5 mg kg⁻¹). In Belung village, Pb concentrations found in plant samples of tomato was 27.38 mg kg⁻¹, in cabbage 27.38 mg kg⁻¹, in sweet corn 29.12 mg kg⁻¹, in green beans 29.12 mg kg⁻¹, in leaf/spring onion 26.51 mg kg⁻¹, in curly chili 33.45 mg kg⁻¹ and in wild spinach 31.72 mg kg⁻¹. In Pandansari village, Pb concentration found in the sample of cayenne/hot chili plant was 30.85 mg kg⁻¹, in chicory/chinese cabbage 26.51 mg kg⁻¹, in mustard greens 26.51 mg kg⁻¹, and in peanuts 26.51 mg kg⁻¹. In Wonorejo village. Pb concentration found in plant samples of sweet corn was 30.85 mg kg⁻¹, in cabbage 29.12 mg kg^{-1} , in leaf/spring onion 31.72 mg kg^{-1} . Further research is needed to test how much influence has the use of agrochemical materials that still contain heavy metals on the level of metal absorption in vegetable plants for food security of human health.

Key Words: vegetables, Pb concentration, production inputs, horticultural lands.

Introduction. Heavy metal contamination on agricultural land has long become a serious concern because of the issue of food security and potential risks to human health (Khan et al 2008; Lu et al 2015; Mahar et al 2016; Gori et al 2019; López et al 2019; Tao et al 2019). Although the composition of heavy metals in agricultural lands tends to be covered by host rock, inputs from various sources such as the application of organic matter, contaminants in fertilizers and other agrochemical materials such as pesticides can be a major source of significant heavy metals in the soil (Alloway 2013).

The reality in the field is that, some farmers use several different types of agrochemical materials that are applied intensively to plants. The results of laboratory analysis for agricultural lands taken randomly in the Poncokusumo sub-district showed a range of Pb content of $7.86-10.20 \text{ mg kg}^{-1}$. The preliminary data raises allegations that the use of various production inputs intensively has the potential to cause heavy metal contamination, especially Pb. How does this affect vegetable crops planted on the land, so this research needs to be done to identify them.

The intensive use of various agrochemical materials as production input within a certain period of time on the land is thought to cause mineralogical changes in the soil

(Lal 2004; Six et al 2000; Amaibi et al 2019). As stated by Stevens & Walker (1970), every soil is affected by the interaction of five or more soil-forming factors, namely climate, biotic factors, relief, parent/host material, time and other factors. The series that are related to land that is different from one another, especially certain properties as a result of time as a soil forming factor known as a *chronosequence*. Most soil properties are functionally related to time. If the age of the soil from *chronosequence* is known, the level of similarity of soil formation can be derived. Successions of plants that have been colonized at the surface of the soil, and in their *chronosequence* in the soil have developed together. Deposition of organic matter and inorganic materials during plant growth in soil initiates gradient depth in soil profiles from various soil characteristics such as pH, bulk density, organic matter content, N, various P fractions, and certainly other minerals. Processes of weathering, leaching in soil profiles leads to large gain, loss, transformation, and redistribution of various organic and inorganic components.

This study aims to identify the distribution of Pb in vegetable planting lands in Poncokusumo sub-district with a land use period of between 5 to 15 years and potential for Pb absorption in various types of vegetables grown, and potential of Pb content from various input sources such as irrigation water, fertilizers and pesticides used by farmers to increase vegetable production.

Material and Method

Location and time of the research. The survey research was carried out in three locations namely Belung, Wonorejo and Pandansari villages which are famous as centers of vegetable production in Poncokusumo sub-district, Malang regency, East Java Province. Research started in July 2017 and lasted until May 2018. The determination of study location was conducted purposively (purposive sampling) at the vegetable production centers in the three villages. On average, the three villages have flat land slopes with a slope of 3-8%, and few have slopes >40% (Figure 1). The types of agroecosystems in the three villages in general are areas of various leaf vegetables and fruit vegetables as well as a few food crops such as rice and corn.

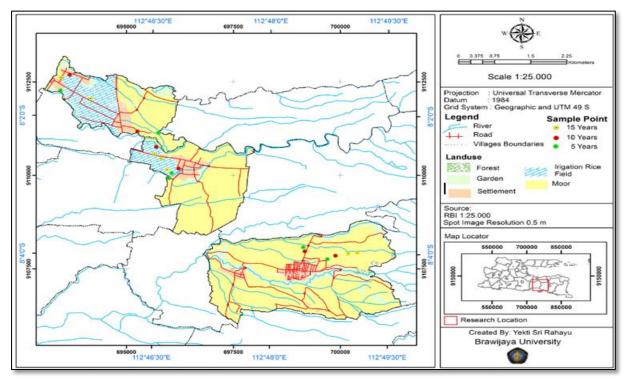


Figure 1. Location of three survey areas of vegetable production and soil sampling locations for vegetable planting in Poncokusumo sub-district.

Determination of sampling methods. The research method used in this study was survey method. Soil sampling was carried out intentionally (purposive sampling) in vegetable production centers in Wonorejo village, Poncokusumo sub-district, Malang Regency, East Java Province. Each of the 30 composite soil samples from a depth of 0-20 cm was collected from the study area in the villages of Belung, Wonorejo and Pandansari. Soil samples were taken from the study area planted with growing vegetables.

Method of collecting data. The survey begins with data collection by gathering information from several farmers and farmer groups as a source of data and also village officials, about the land use period and the history of the use of various production inputs, especially fertilizers and pesticides in the period of 0-5 years, 6-10 years and more than 11-15 years. The data collection technique was done by interviewing direct sources and location observations as well as documentation in the field. After the historical information on the use of various production inputs in the vegetable land was obtained, we proceeded with the determination of the location of land/soil sampling in several locations according to the period of 0-20 cm evenly at five different points and stored in polyethylene plastic bags and then composite soil samples were prepared to analyze the soil chemical properties.

In the study area we also identified production input sources used by farmers for the production of their vegetable crops. Identification was done by collecting and recording several production input samples among other namely: 1) water used as an irrigation source starting from the main source, namely from the main dam, then sample the front water of the land canal before entering the inner land, until irrigation water flowing inside the land; 2) sampling some manure commonly used by farmers in the village of Wonorejo; 3) inorganic fertilizers both solid and liquid forms commonly used by farmers for plant maintenance; and 4) pesticides both solid and liquid forms which are commonly used by farmers for the maintenance of cultivated vegetable crops. All materials used as input for the production of vegetable crops were then analyzed for its Pb content.

Chemical analysis. Analysis of soil chemical properties was tested among other soil pH which was measured by pH meter in suspension of 1:1 sample soil with H_2O that has been shaken for 60 minutes. Analysis of organic C was measured by the titration method used by Walkley & Black (1934), N-total with the method of Kjedhal (Bremmer & Mulvaney 1982), P-total according to Olsen & Sommers (1982), Cation Exchange Capacity (NH₄OAC pH 7.0), and Pb heavy metal content with Atomic Absorption Spectrometry (AAS) wet digestion method with HNO₃ and HClO₄ (AOAC 2002).

Statistical analysis. Data from laboratory analysis result on the Pb content at the locations of soil sampling, and Pb content in production input materials such as fertilizers, pesticides and irrigation water, as well as the Pb content from samples of vegetable plants at several soil sampling points were tabulated and presented in table form. Furthermore, data analysis was carried out descriptively, and the patterns of data obtained are illustrated in graphical form.

Results

Soil characteristics of the study area. The chemical properties of soil in the study area is shown in Table 1. According to general criteria, the average soil pH in the study area tended to be acidic to slightly acidic namely an average of 4.9 (Wonorejo village), 5.1 (Belung village) and 6 (Pandansari village). Availability of nutrients, namely Total N in the study area showed a low average value for Wonorejo villages (0.11%) and Pandansari (0.15%), and very low (0.1%) in Belung village. Availability of nutrients, namely Total N in the study area showed a low average value for Wonorejo villages (0.11%) and Pandansari (0.15%), and very low (0.1%) in Belung village. The average total P content in the three study villages showed a very high value, while the average K

content in Wonorejo and Pandansari villages tended to be low (0.36 and 0.21 cmol kg⁻¹) and in the village of Belung tended to be moderate (0.41 cmol kg⁻¹). Cation exchange capacity (CEC) in Belung, Wonorejo and Pandansari villages tended to be low at 22.26 cmol kg⁻¹, 20.03 cmol kg⁻¹ and 21.15 cmol kg⁻¹ respectively.

Table 1

Soil chemical characteristics of vegetable production centers in the three villages of Poncokusumo Subdistrict

Soil chemical characteristics	Village			
	Belung	Wonorejo	Pandansari	
pH 1:1 (H ₂ O)	5.1	4.9	6	
pH 1:1 (KCl 1N)	4.6	4.5	5.7	
Organic C (%)	0.55	0.91	1.36	
Total N (%)	0.1	0.11	0.15	
C/N	5	9	9	
Organic material (%)	0.94	1.57	2.36	
P.Bray1 (mg kg ⁻¹)	119.65	95.11	47.55	
K (cmol 100 g ⁻¹)	0.41	0.36	0.21	
CEC (cmol 100 g ⁻¹)	22.26	20.03	21.15	

Source: primary data.

Pb concentration in the soil of vegetable planting areas. The average Pb concentration in the vegetable planting area in the three villages is presented in Table 2. The average Pb concentration from each village on land use starting from 5, 10 to 15 years is still below the threshold limit value for the protection of agricultural lands. Information from the Bogor Land and Agro-Climate Research and Development Center shows that soil quality standards in Indonesia are not yet available because they are difficult to define. In Indonesia environmental quality monitoring and recovery have not been carried out in an integrated manner because there are only air and water quality standards. In addition, the level of soil weathering in Indonesia is quite intensive so that the possibility of soil bearing capacity against heavy metals is lower than in industrialized countries.

Table 2

Study sites	Land use period (Vear)	Dissolved Pb (ppm)	Total Pb (ppm)
Study sites	Land use period (Year)	HCI 0.1N	HCI 25%
Belung	0-5	0.17	1.10
	6-10	0.36	0.73
	11-15	0.36	3.15
Pandansari	0-5	1.10	1.85
	6-10	1.10	4.83
	11-15	4.27	9.49
	0-5	1.47	11.53
Wonorejo	6-10	1.47	11.35
	11-15	4.08	11.62

Soil Pb concentration with different land use periods

Source: primary data processed.

Although in general the Pb concentration of some soil samples was below the threshold, some samples, especially in the village of Wonorejo, show higher total Pb compared to the villages of Belung and Pandansari. The soil chemical characteristics are shown in Table 1, Soil Pb concentration Table 2, and comparison of average soil Pb concentrations according to the land use period of vegetable crop production in Figure 2.

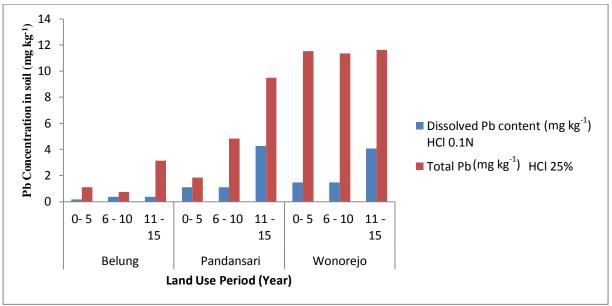


Figure 2. Comparison of the average soil Pb concentration according to the period of land use for vegetable crop production.

Table 1 show that the total Pb concentration in the soil in Wonorejo village was generally higher than the total Pb concentration in the soil from the other two villages. While the available average Pb concentration in Wonorejo and Belung villages, was higher than in the other three villages. Based on the survey results of land use history for vegetable crop production in three villages, the average Pb concentration in soil in all three locations for periods of use over 11 years shows a greater value than the period of use under 10 and 5 years. And the average Pb concentration in the production area of vegetable crops in Wonorejo village showed higher value compared to Belung and Pandansari villages (Figure 2). The Pb concentration in several production input sources is presented in Table 3, and the Pb concentration in several types of leaf vegetables and fruit vegetables in Table 4.

The results of laboratory analysis on several production inputs used by farmers in several villages indicated that there were several production inputs with higher Pb content (Table 3). From chemical fertilizers there is one type of fertilizer with a Pb content of around 21 ppm, while from pesticides there is one type of pesticide with a Pb content of around 99 ppm. While the input in the form of organic material from several samples analyzed in general had very low Pb content even unmeasurable. Irrigation water samples taken at the survey location generally also showed a very low Pb content on an average of 0.01-0.02 ppm (Pb quality standard for water quality management and water pollution control based on Government Regulations No. 82 of 2001 equal to 0.3-1 mg/L).

In general, the total Pb concentration in several samples of vegetable plants taken in the three villages surveyed locations showed an average concentration that exceeds the maximum limit of heavy metal contamination in vegetables of 0.5 mg kg⁻¹ (Indonesian National Standart/SNI:7378:2009) (BSN 2009). The results of laboratory analysis of Pb concentrations in each plant organ from the initial survey showed that for the leaf vegetable crops group, chicory/chinese cabbage showed a higher range of Pb concentrations in the canopy section where the canopy of mustard plants was an edible part of this plant. While for the fruit eaten vegetable crops group, beans, and chilli, showed higher concentrations of Pb.

Table 3

	Pb (mg kg ⁻¹)	Quality Standard Value
Production Input	$HNO_3 + HClO^4$	(Based on the applicable legislation in Indonesia)
Irrigation Water		0.3–1 mg/L (Government Regulation No. 82 of 2001)
Wonorejo dam	Nd	
Wnrjo inner irrigation	0.02	
Wnrjo outer irrigation	0.02	
Belung inner irrigation	Nd	
Belung outer irrigation	0.01	
		Granules and crumbs = 50 ppm (Regulation of the
Organic fertilizer		Ministry of Agriculture
Organic rentilizer		No.70 / Permentan /
Hugh L broilers	F	SR.140 / 102011))
Husk + broilers	5	
Duck manure	9	
Goat manure	6	
Cow manure	1	
Laying hens manure	Nd	
Rabbit manure	Nd	
Broilers manure	Nd	
Swallow manure	Nd	
Laying hens manure	7	
Goat manure	Nd	
		POC = 12.5 ppm
Liquid Organic Fertilizer (cow urine) / POC	Nd	(Regulation of the Ministry of Agriculture No.70/ Permentan/SR.140/102011)
Inorganic fertilizer		, ,
SP (superphosphate)	21	
KCI Ý	Nd	
NPK Mutiara	Nd	
NPK Phonska	Nd	
ZA	Nd	
Urea	Nd	
Leaf Fertilizer	1	
Lime	16	
Pesticide		
Fungicide (11)	Nd - 8	
Insecticide (22)	Nd – 99	
Acaricide (1)	3.77	
Herbicide (4)	Nd - 2	
Adhesive (5)	Nd – 0.71	

Pb concentration in several production input sources

Nd - not detected.

Source: Primary data processed (2018).

Table 4

		Total Pb		Total Pb
Village		$(mg kg^{-1})$		(mg kg ⁻¹)
<i>illa</i>	Vegetables	$HNO_3 + HClO_4$	Vegetable organs	HNO₃ +
2		Total vegetable		HClO₄
	Spring onion (Allium fistulosum)	organs 31.72	leaf	1.87
Wonorejo	Sweet corn (<i>Zea mays</i>)	30.85	leaf + stem	0.14
	Sweet com (Zea mays)	20.02		0.14
	Cabbage (<i>Brassica oleracea</i> var. Capitata)	29.12	leaf + stem	4.46
	Tomato (Lycopersicum esculentum	27.38	fruit	1.00
	Mill.)		leaf + stem	2.73
	,	20.12	cob	1.00
	Sweet corn (Zea mays)	29.12	leaf + stem	5.33
ם מי	Ctring hoon (Phagoalus yulgaria)	20.12	pod	6.19
Belung	String bean (<i>Phaseolus vulgaris</i>)	29.12	leaf + stem	7.92
Be		22.45	fruit	2.73
	Curly chili (Capsicum annuum L.)	33.45	leaf + stem	3.60
	Spring onion (Allium fistulosum)	26.51	Leaf	1.87
	Cabbage (<i>Brassica oleracea</i> var. Capitata)	27.38	leaf + stem	7.06
Ē	Chinese cabbage (<i>Brassica rapa pekinensis</i>)	26.51	Leaf	19.18
ISa	Peanuts (Arachis hypogea)	26.51	pod	12.25
dar			leaf + stem	9.66
σ	Mustard greens (Brassica chinensis)	26.51	leaf + root	26.51
	Tomato (<i>Lycopersicum esculentum</i> Mill.)	32.59	leaf+stem+fruit+r oot	32.59
	Peas (<i>Pisum sativum</i>)	30.85	leaf + stem	2.73
		50.05	pod	0.14
Wonomulyo	Chinese cabbage (<i>Brassica rapa pekinensis</i>)	29.98	leaf + stem	3.60
	Long bean (Vigna unguiculata)	28.25	leaf + stem pod	5.33 7.06
			fruit	1.00
Ś	Hot chili (<i>Capsicum frutescens</i> L.)	30.85	leaf + stem	5.33
		20.25	fruit	0.14
	Eggplant (<i>Solanum melongena</i>)	28.25	leaf + stem	1.00

Pb concentration in several types of leaf and fruit vegetables

Discussion. The average distribution of Pb concentration in Wonorejo village was in the range of 11.35-11.62 mg kg⁻¹ identified as higher than in Belung village (range 0.73-3.15 mg kg⁻¹) and Pandansari (range 1.85 -9.49 mg kg⁻¹) either during the land use range of 5, 10 and 15 years. The average concentration of heavy metals contained in the three villages were generally relatively harmless, however, it can be dangerous if heavy metals become soil contamination, through the transfer from the soil to plants and then into the metabolic system of living organisms in amounts exceeding the threshold.

The absorption results in plants showed this potential where the Pb concentration in several types of vegetables grown in the area an average above the maximum limit of Pb contamination in fruits and vegetables namely 0.5 mg kg⁻¹ according to Indonesian National Standart/ SNI 7378: 2009 (BSN 2009). In Belung village, Pb concentration (mg kg⁻¹) in plant samples of tomato was 27.38, in cabbage 27.38, in sweet corn 29.12, in bean 29.12, in spring/leaf onion 26.51, in curly chili 33.45 and in wild spinach 31.72. In Pandansari village, Pb concentration (mg kg⁻¹) in samples of cayenne pepper/hot chili was 30.85, in Chinese cabbage 26.51, in mustard greens 26.51, and in peanuts 26.51. In Wonorejo village, Pb concentration (mg kg⁻¹) in samples of sweet corn plants was 30.85, cabbage 29.12, in spring/leaf onion 31.72. Metal entry can occur indirectly when food ingredients such as vegetables are consumed. Although Pb is not an essential element for plants, plants can absorb it from the soil and accumulate it in edible parts in various concentrations (Kalinovic et al 2019; Ahmad et al 2019; Vongdala et al 2019; Gori et al 2019; Papaioannou et al 2019).

The amount absorbed in parts that can be eaten by humans is the most important thing for soil PB that is harmful to humans. Rahman et al (2014), Byambas et al (2019), Mishra et al (2019), Rai et al (2019), Mishra et al (2019), Kibet et al (2019), Hussain et al (2019), Burden et al (2019), revealed that metal accumulation, toxicity and the presence of heavy metals in agricultural soils can be absorbed by plants and give the toxic effects on humans and animals that eat them.

Although the concentration of Pb in vegetable production lands was below the threshold, however, the use of other input sources which may contain Pb metal can also potentially increase Pb uptake/absorption in cultivated vegetable plants. According to Alloway (2013), Álvarez et al (2018), Mihaljevič et al (2019), Lee et al (2019), Shin et al (2019) the total content of heavy metals in the soil is obtained from minerals in geological host rock where the soil has developed (lithogenic sources) and input from various possible sources of anthropogenic contaminants. This also includes atmospheric deposition from aerosol particles (diameter <30 μ m), raindrops containing heavy metals, or elements in the form of gases, direct application of agricultural fertilizers, agrochemical materials and various organic materials such as animal manure, food waste and compost. The total concentration of metals in the soil is the sum of all these various inputs minus losses through harvested or pruned plants, erosion of soil particles by wind and water, leaching of soil profiles in solution and losses due to volatilization of gas element forms.

The results of chemical analysis of several input sources used by farmers in vegetable production in this study indicated that most production inputs such as irrigation water, organic fertilizer, inorganic fertilizers and pesticides still had Pb content even though was below the threshold limit. However, the continuous application of these inputs can increase the potential for higher Pb metal uptake/absorption in plants.

According to Sheoran et al (2016) Pb uptake/absorption into plants, depends both on soil factors and also on the factors of the plant itself. Some soil factors that affect the availability of metals in plants include soil types, especially texture, moisture, pH, redox reactions, cation exchange capacity, biochemical processes.

Regarding soil texture, the availability of metals is highest in loamy soil and sandy soils, followed by clayey loam soil and the lowest in soft-textured clay soil. Normally, higher levels of heavy metals can be held in soft-textured soils such as clay and loamy clay compared to coarse-textured soil such as sand (Sheoran et al 2010). The present research results showed that the texture of the soil at the study site was sandy loam, thus supporting the availability of metals in the soil. While the relation to the availability of metals with soil moisture, it was explained that in general metal absorption was observed at higher soil moisture levels. This is in line with the need for plants to be able to produce high biomass, generally requiring high levels of humidity, so that the opportunity to extract a number of metals from the soil can also increase (Marchiol et al 2004). Vegetables in Belung, Wonorejo, and Pandansari villages use flooding irrigation systems for 1-3 days so that they can maintain soil moisture for a long time after the land is irrigated until saturated during plant growth.

While in relation to pH, generally the concentration of heavy metals in soil solutions can be increased by decreasing soil pH because the number of H + ions will increase and the cation exchange capacity between heavy metal cations and H + absorbed to the surface of soil particles will increase after pH decreases. Therefore, at a lower pH a large number of heavy metal ions are absorbed from the surface of the colloid and clay mineral particles and then enter to the soil solution. A decrease in pH can decide precipitation-dissolution equilibrium among the heavy metal ions and promotes the release of heavy metals into the soil solution (Hattori et al 2006). The average pH in the

three villages is below the value of 6 with the acidic to a slightly acidic range (Table 1) which allows the availability of Pb in the soil to be absorbed by plants. This is also supported by the results of Zhang et al (2018) that the nature of soil affects the mobility and bioavailability of heavy metals in agricultural soils which ultimately affects their accumulation in plants from the soil. Zhang et al (2018) reported that soil pH is an important factor that influences the availability of heavy metals that surround the soil where the fraction of the availability of Cu, Hg, Ni, Mn and Pb increases with decreasing pH.

In general, high pH values (>8.0) promote adsorption and precipitation, while low pH (<5.0) weakens the association of heavy metals in the soil, which further changes their mobility in the soil. These results are consistent with the study of Cerqueira et al (2011) that Pb^{2+} absorption is significantly affected by pH. Soil horizon with the highest pH, CECe, Mn and Fe content and various minerals in clay fraction showed the highest competitive sorption capacity for Cu²⁺ and Pb²⁺. The sorption irreversibility reaches the peak in the soil with the lowest acidity. Lots of soils that show high affinity for Pb^{2+} and based on indications of migration, Pb^{2+} are less mobile crossing soil profiles than Cu^{2+} .

The availability of Pb in the soil is also determined by the content of organic matter as a soil constituent, where organic matter has a high affinity against metal cations due to the presence of ligands or groups of metal chelating (Adriano 2001). The general level of affinity of metal cations complexation with organic matter, is namely $Cu^{2+}>Cd^{2+}>Fe^{2+}>Pb^{2+}>Ni^{2+}>Mn^{2+}>Co^{2+}>Mn^{2+}>Zn^{2+}$. Meanwhile, the soil in the study site has very low to low organic matter, 0.94% (Belung), 1.57% (Wonorejo) and 2.36% (Pandansari), so the possibility of Pb metal affinity in organic matter is very low, and is more easily available in the soil.

Conclusions. Based on the results of this study, it was concluded that the average distribution of Pb concentrations in Wonorejo village was in the range of 11.35-11.62 mg kg⁻¹ identified as higher than in Belung village (range 0.73-3.15 mg kg⁻¹) and Pandansari (range 1.85 -9.49 mg kg⁻¹) either during the land use range of 5, 10 and 15 years. Some sources of production inputs used by farmers from irrigation water, organic fertilizers, inorganic fertilizers and pesticides are also identified to contain Pb metal, although the average Pb concentration in these materials was still below the maximum threshold specified. The Pb concentration in several types of vegetables grown in the area was average above the maximum limit of Pb contamination in fruits and vegetables namely 0.5 mg kg⁻¹. The results of the identification in this study provide guidance on several production input sources containing heavy metals and several types of vegetables that have high potential in accumulating heavy metals in horticultural fields. Further research is needed to test how much influence has the use of agrochemicals that still contain heavy metals to the levels of metal absorption in vegetable plants for food security of human health.

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