Heavy Metals in Indonesian Paddy Soils

by Dedik Budianta

Chapter

Heavy Metals in Indonesian Paddy Soils

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Abstract

Long-term cultivation of paddy soils has resulted in Pb and Cd accumulation that exceeds the WHO tolerance levels of 2 mg kg^{-1} and 0.24 mg kg^{-1} in food. In Musi Rawas, South Sumatra, Indonesia, the paddy soils with the greatest levels of Pb and Cd were those that had been intensively farmed for 80 years, reaching the concentrations of 20.56 mg kg^{-1} Pb and 0.72 mg kg^{-1} Cd for soil, and 3.11 mg kg^{-1} Pb and 0.29 mg kg^{-1} Cd for rice. The lowest concentrations were obtained with 20 years of cultivation at 17.82 mg kg^{-1} and 0.26 mg kg^{-1}, for Pb and Cd in soils, respectively. The Pb content in the paddy fields in Pati, Central Java, ranged from 0.23 to 2.55 mg kg^{-1}, while the Pb content in the lowland watershed of Solo Hilir ranged from 0.20 to 2.94 mg kg^{-1}. The highest concentration of Pb and Cd in rice was found at 80 years old in paddy soils with the value of 3.11 mg kg^{-1} and 0.29 mg kg^{-1}, respectively. The lowest concentrations were found at 20 years old of soils with a value of 2.35 mg kg^{-1} Pb and 0.15 mg kg^{-1} Cd, respectively.

Keywords: cadmium, intensive farming, lead, paddy soil, P fertilizer, rice intensification

1. Introduction

Rice is a staple food that globally provides calories to more than 3.5 billion people. It has contributed almost 19% of global human per capita energy and 13% of per capita protein [1]. Paddy soils used for rice growth are contained by embankments, called galengan in Indonesia, or canal to hold water. Indonesian farmers have per capita paddy fields of only <0.5 ha, which decreases over time due to population growth and conversion to non-agricultural activities. The total area of rice fields in Indonesia is around 7,483,948 ha [2]. The average production is around 7–8 tons ha^{-1} when the soil is relatively fertile. However, when the soil is less fertile, rice production is very low, below 4 tons ha^{-1}. The fertility of paddy soils has decreased, as indicated by the decreasing availability of macro- and micro-nutrients, low organic matter content, and slightly low pH (**Table 1**) [3, 4].

The fertility of paddy fields continuously decreases with the time of land use due to harvest and irrigation.

According to **Table 2**, paddy soils in Musi Rawas, South Sumatra, have a pH ranging from 5.40 to 5.56 with a low organic C content of 1.75–1.85%, moderate to low soil CEC ranging from 15.31 to 19.58 cmol(+)kg^{-1}, moderately available P between 14.10 and 20.80 mg kg^{-1}, medium K-exchangeable of 0.58 cmol(+)kg^{-1}, exchangeable Na between 0.33 and 0.70 cmol(+)kg^{-1}, exchangeable Ca ranging from 2.10 to 6.48

No.	Soil pH	Org-C	N	C/N	CEC
		%			Cmol(+)kg^{-1}
1	6.14	2.58	0.13	19.85	5.41
2	6.36	2.37	0.13	18.23	4.97
3	6.72	1.19	0.12	9.9	6.07
4	5.71	0.96	0.11	8.7	6.61
5	5.85	1.52	0.15	10.1	5.42
6	6.35	0.89	0.10	8.9	7.26
Mean	6.19	1.58	0.12	12.61	5.96
Criteria	slightly acidic	low	low		low

Table 1.
Some characteristics of paddy soils in Sidoarjo (East Java) near the industrial area [3].

Soil properties	Age of soil cultivation (year)				Unit
	20	40	60	80	
pH (H_2O)	5.56 sa	5.52 sa	5.44 a	5.40 a	
C-organic	1.85 l	1.80 l	1.75 l	1.80 l	%
P-available	14.10 l	14.50 l	16.40 m	20.80 m	mg kg^{-1}
K-exchangeable	0.58 m	0.58 m	0.58 m	0.58 m	cmol(+)kg^{-1}
Na-exchangeable	0.44 m	0.44 m	0.33 l	0.70 m	cmol(+)kg^{-1}
Ca-exchangeable	2.10 l	2.50 l	6.08 m	6.48 m	cmol (+)kg^{-1}
Mg-exchangeable	0.57 l	0.35 vl	0.68 l	0.50 l	cmol (+)kg^{-1}
CEC	17.40 m	16.31 m	19.58 m	15.31 l	cmol (+)kg^{-1}

Note: a = acidic, m = moderate, sa = slightly acidic, vl = very low and l = low.

Table 2.
Characteristics of soils based on the age of use of paddy soils [5].

cmol(+)kg^{-1}, and low exchangeable Mg of 0.35–0.68 cmol(+)kg^{-1}. The low organic C content of 1.75 to 1.85% can increase the solubility of Pb and Cd in paddy soil while increasing the uptake by plant roots. Organic matter is vital as a regulating agent for heavy metal mobility in the soil [6]. Furthermore, Pb and Cd can form complex and chelate compounds with organic materials [7]. The complex form is a reaction between metal ions and ligands through electron pairs [8]. Paddy soils have low pH ranging from 5.56 to 5.4. The high soil acidity or low pH can increase the solubility of Pb and Cd in the soil with the uptake by plant roots [9]. Soil acidity is an essential factor that determines metal transformation and controls the chemical properties of Pb and Cd and other processes in the soil. The decrease in pH increases the availability of heavy metals except for Mo and Se. At low pH, the availability of Pb and Cd increases, and the more acidic the soils, the greater the heavy metal affects the rice [1, 10]. To increase the fertility of paddy soils, farmers intensively apply inorganic fertilizers such as urea, SP-36/TSP, and KCl. These inorganic fertilizers are essential to provide adequate nutrients for crop growth and ensure successful harvests [11]. This is supported by the data in **Table 3**, which indicate that the average lowland rice

No	Subdistrict	Fertilizer (kg ha^{-1})			
		Urea	SP36/TSP	KCl	NPK
1	Tugumulyo	166.37	144.68	67.50	123.70
2	Megang Sakti	189.65	156.25	74.37	141.66
3	Purwodadi	181.94	162.85	76.84	131.57
4	Muara Beliti	191.27	156.90	64.54	133.22
5	Sumber Harta	192.85	130.58	69.23	131.42
	Total	922.20	751.26	352.48	661.57
	Mean	184.44	150.26	70.49	132.31

Table 3.
Average fertilizer usage in five sub-districts of Musi Rawas Regency, Indonesia [5].

farmer uses around 150.26 kg ha^{-1} P fertilizer in each growing season, exceeding the recommended dose of 100 kg ha^{-1} [5].

Paddy soil is not a typical soil classification term but indicates how to manage various soil types for rice cultivation. There are four paddy soil ecosystems: (a) flood-prone rice ecosystem, characterized by a flat to slightly wavy or basin surface; it is flooded due to high tides for more than 10 consecutive days as deep as 50–300 cm during plant growth; (b) aerobic to anaerobic and rice cultivation is carried out by transferring or spreading seeds on dry plowed soil; (c) rainfed lowland rice ecosystem, characterized by a flat to the slightly wavy land surface, bordered by bunds, and inundated due to discontinuous tides with varying depths and periods; and (d) aerobic–anaerobic soil alternating with varying frequency and period, where rice planting is carried out by transferring seeds to silted soil [12]. Intensive management of paddy soils in the long term can reduce soil productivity and environmental quality. High inputs of agrochemicals can deplete nutrients in the soil and cause negative impacts in the form of increased residues of materials. Additionally, consumer demands for food or agricultural products that are safe and hygienic, have a high nutritional value, and are free of contamination are a public concern for the quality of the environment and human health [13]. Furthermore, [13] reported that around 21–40% of paddy soils in the Pantura of West Java were contaminated with these two types of heavy metals; even 4–7% of them were contaminated in the heavy metals category, which was Pb > 1.0 mg kg^{-1} and Cd > 0.24 mg kg^{-1}.

2. Heavy metals

Various sources and causes of contamination of paddy fields that can lead to soil degradation include agrochemicals, industrial waste, mining activities, and household waste. The two sources of heavy metals are natural and anthropogenic [1]. The use of synthetic fertilizers (inorganic fertilizers) and industrial activities play an important role as a source of pollution in rice fields [14]. There are many reports of contamination of rice fields, especially areas adjacent to factories [15–17]. The amount of waste generated from industrial processes causes water sources to be polluted. Furthermore, materials consisting of toxic compounds can settle in the rice soil. This process is repeated over time, accumulating these materials and heavy metals in the soil. Therefore, there will be undesirable changes in the physical, chemical, and biological properties of the soil. Productivity decreases with the ability to support plant growth [18]. Heavy metal contents in agricultural soils can directly affect human health by consuming crops grown in contaminated soils [17]. These metals are nonessential elements but can accumulate in plants and adversely affect human health [19]. Contaminated soil adversely affects the whole ecosystem when these toxic metals migrate into groundwater or are taken up by plants, which may threaten ecosystems [20]. In general, the metals are accumulated mostly in the root compared to the stem, leaf, and grain [1]. The occurrence of these metals in paddy field soils ranks in the order Mn > Zn > Pb > Cr > Cu > Cd [19]. Heavy metals are potentially toxic to crop plants, animals, and humans when contaminated soils are used for crop production [21]. Environmental contamination of the biosphere due to intensive agricultural and other anthropogenic activities poses severe problems for the safe use of agricultural land [22]. Heavy metals such as Cd and Pb are of primary concern in soil and food contamination because of their toxicity, particularly in the rice cropping system [23]. These toxic elements accumulate in the soils, contaminating the food chain, endangering the ecosystem's safety, and causing soil degradation.

Degraded soil will have properties that do not support rice growth. It will lose the topsoil or arable layer, lose nutrients needed by rice plants, and result in reduced levels of organic carbon. In addition to these observable characteristics of degraded soils, it can also be distinguished by plants that typically do not thrive in such conditions. The performance of plants is reduced when planted in soil with degraded physical, chemical, and biological qualities. The parameters used to evaluate the level of soil degradation are decreasing base saturation, available nutrients including N, P, K and trace elements, bulk density, soil permeability, and organic carbon [24].

Soil properties influence rice growth and development. The characteristics supporting plant growth should be maintained, one of which is soil conservation measures to prevent chemical damage/degradation. Degraded soil can also lose the top layer, impacting the loss of nutrients needed by plants, changes in soil structure, and reduced levels of organic carbon. The organic carbon has a major role in improving the physical, chemical, and biological properties of the soil [25]. It can also be identified by using plants with poor growth performance. In this regard, the plant can be used as an indicator of soil degradation. Many definitions of soil degradation have been reported, showing a decrease in soil chemical properties compared to non-degraded soil. Land degradation results from one or more processes that decrease the actual or potential ability to produce food and fiber and provide ecosystem services. This definition shows a general understanding of agriculture's broad scope [26]. Land or chemical degradation is often associated with a use that does not follow the aspects of the balance of inputs and outputs. Inputs are related to soil improvement or fertilization in cultivation activities. In contrast, the output is associated with plant nutrient uptake and the possibility of leaching through erosion mechanisms. The phenomenon of land degradation is found in areas of land that promote agricultural activities. Land degradation can be indicated by symptoms of poor plant growth or the growth of weeds on the soil. The marginalization will continue with low inputs for farming and dry land management technology, which ultimately causes physical and chemical degradation. On sloping land, land degradation will occur quickly due to erosion, which reduces the quality of the physical and chemical properties of the soil. Consequently, the soil will be damaged or degraded due to acidification, accumulation of salts (salinization), and contamination of heavy metals, organic compounds, and xenobiotics such as pesticides or oil spills.

3. Characteristics of Pb and Cd

3.1 Lead (Pb)

Lead (Pb) is accumulated in plant organs, namely, leaves, stems, roots, and tubers (shallots), and the transfer depends on the soil composition and pH. High Pb concentrations (100–1000 mg kg^{-1}) have a toxic effect on photosynthesis and growth [27]. Pb is one of the nonessential heavy metals that are toxic to living organisms. It causes stunted growth, irritates the eyes, and contributes to lung [28] and kidney [29] damage. The highest accumulation in roots was proven by [30] through a study of Pb in kale (*Brassica oleracea* var. *sabellica*). In the 6-week-old kale plant, Pb concentration in the roots reached about 3360 mg kg^{-1}, and in other parts of the plant, it reached 2090 mg kg^{-1}. In 3-week-old kale, the Pb content in the roots was 1.860 mg kg^{-1} in the sample but 1.130 mg kg^{-1} in other parts. These data indicate that most Pb in water spinach is accumulated in the roots.

The largest Pb pollution comes from burning gasoline, which produces PbBrCl and $PbBrCl_2PbO$. The pollution can come from Pb components in dissolved air or water, such as $PbCO_3$ [31]. According to [32], heavy metals in the media are rapidly absorbed by plants at very low concentrations. The mechanism of absorption and accumulation can be divided into three continuous processes: (a) Absorption by roots: metals should be brought into the solution around the roots (rhizosphere) in several ways to be absorbed. Water-soluble compounds are usually taken up by the roots with water, while the surface absorbs hydrophobic compounds. (b) Translocation of metals from roots to other plant parts: After penetrating the root endodermis, metal or other foreign compounds follow the transpiration flow through the transport tissue (xylem and phloem) to other parts. (c) Metal localization in cells and tissues: This aims to keep metals from inhibiting plant metabolism. Plants have detoxification mechanisms in certain organs, such as roots, to prevent metal poisoning of cells. Metals in the root cells are transported to other plant parts through the xylem and phloem network when translocation occurs in the plant body. At low concentrations, heavy metals do not affect plant growth but cause damage to the soil, water, and plant at high concentrations.

Satpathy et al. [33] argued that Pb originating from air/atmosphere pollution is in the form of dust particles, which will stay on the plant's surface. Clouds and rain can cause Pb to be dissolved and enter the plant through the stomata, which can cause damage and contaminate food. Air pollution by Pb mainly comes from exhaust fumes from motor vehicles, and this metal is the remnant of combustion between the fuel and the vehicle engine. The presence of Pb in motor vehicle fuel functions as an anti-knock agent. The Pb element is released into the air through the exhaust of the vehicle's gasoline. Some will form particulates in the free air with other elements, while others will stick and be absorbed by the leaves of plants along the way. Soil contamination by Pb is more extensive than other heavy metals. This is because the largest contribution is from anthropogenic sources. The research results [34] showed that the Mn, Co, Cr, and Ni on the soil surface come from lithogenic and anthropogenic sources. These results indicated a significant need for developing pollution prevention and reduction strategies for heavy metal pollution. Accumulation of heavy metals can degrade soil quality, reduce crop yields and agricultural product quality, and negatively impact humans, animals, and the ecosystem. The solution can be achieved by identifying the source and measuring the concentration of heavy metals and the spatial variability in the soil. The results revealed could be used to determine the increase in Cd and Pb concentrations [35].

3.2 Cadmium (Cd)

Soil Cd in igneous, metamorphic, and sedimentary rocks is 0.100–0.300, 0.100–1.00, and 0.300–11 mg kg^{-1}. In general, the Cd content in the soil from the weathering process of rocks is 1.00 mg kg^{-1} or lower. The elements Cd and Zn have almost similar chemical properties, and only their function in the plant body is different. Cd levels in plant tissues range from 0.100 to 1.00 mg kg^{-1}. Excessive Cd accumulation can occur from other materials, with a detrimental effect on plant growth. This is because it breaks down nitrate absorption and inhibits the activity of the enzyme nitrate reductase. The critical limit of Cd in plants is 5–30 mg kg^{-1} [36], and the content in the 0–20 cm layer, on average, is close to 0.5 mg kg^{-1}, which is the critical limit concentration of the metal [13]. Cadmium in the soil is an anthropogenic byproduct of fertilizer and garbage dumps. Most of the soil's Cd is affected by pH, organic materials, metallic oxides, clay, and organic and inorganic substances [28]. The average level of natural Cd in the earth's crust is 0.1–0.5 mg kg^{-1}.

The Cd content is influenced by the reaction of the soil and fractions capable of binding the ions. Due to the rise in the hydrolysis process, the adsorption complex, and the charge of the soil colloid, Cd concentration in soil solution reduces with increasing pH. Sarwar et al. [37] stated that there was a reduction in root and shoot length of about 45 and 35% in maize plants grown on media containing 28.1 and 11.2 mg kg^{-1} Cd(II) ions, respectively, at the age of 18 days. The contribution from atmospheric deposits occurs in industrial areas that use coal and oil as fuel. Cd is added to the soil through phosphate fertilizers, manure, incinerator waste (furnace), and sewage sludge [23, 36]. In addition, the increase in Cd can occur through phosphate fertilizers, whose levels vary greatly depending on the type of phosphorite as an industrial material for phosphate fertilizers [38]. Cadmium has chemical properties similar to those of Zn, especially in the process of absorption by plants and soil. However, Cd is more toxic, which can interfere with enzyme activity. Excessive levels of Cd in food can damage kidney function, interfere with Ca and P metabolism, and cause bone disease [39].

4. Sources and causes of Pb and Cd pollution in agriculture

4.1 Phosphate fertilizer

P fertilizer is regularly applied to the soil in intensive farming systems to increase plant growth. The compounds used contain heavy metals Pb and Cd [14, 40]. Triple super phosphate (TSP) fertilizer supplied excessively in rice fields in the long term will be accumulated and cause pollution because of heavy metals. These metals are also present in natural phosphate rock used as the raw material for the manufacture of fertilizer P [14, 41]. Pb and Cd are known to have no physiological activity/function, and applying certain phosphate fertilizers adds Cd and other potentially toxic elements to the soil, such as F, Hg, and Pb [42]. Phosphate fertilizers can significantly contribute to hazardous trace elements such as arsenic (As), Cd, and Pb in croplands. These trace elements have the potential to accumulate in soils and be transferred through the food chain [11]. Various inorganic fertilizers and those derived from phosphate rock contain heavy metals (**Table 4**). The results of different phosphate fertilizers showed the presence of P_2O_5; secondary Ca and Mg; microelements Fe, Mn, Cu, and Zn; and heavy metals Cd, Cr, Pb, Cu, and Hg in varying amounts, namely, Cd (0.1–170 mg kg^{-1}), Cr (66–245 mg kg^{-1}), Pb (40–2000 mg kg^{-1}), and Cu

Phosphate rock (PR)	Cd	Cr	Pb
		mg kg^{-1}	
PR Christmast	38	-	60
PR Tunisia	76	-	42
PR Marko	57	-	113
PR Jordan	5	344	im
PR China Huinan	3	-	im
PR Ciamis	28	20	im
PR Sukabumi	65	-	65
SP-36	11	4	im

im = immeasurable.

Table 4.
Heavy metals in various types of natural phosphate rock and SP-36 [43].

(1–300 mg kg^{-1}) [44]. Long-term P fertilizer applications are likely sources of heavy metals (Pb and Cd) in agricultural soils and crops [45, 46].

From the analysis of P fertilizer, TSP fertilizer contains 120.60 Pb mg kg^{-1} and 4.90 mg kg^{-1} Cd, while single superphosphate (SP)-36 contains 5.3 mg kg^{-1} Pb and 10.43 mg kg^{-1} Cd. Rai et al. [47] explained that using P fertilizer could cause the soil accumulation of Pb and Cd. Furthermore, it contains heavy metals Pb and Cd from the raw material for making P fertilizer. Natural phosphate rock has various associated elements such as Pb and Cd in high enough quantities. Meanwhile, the elements can be dissolved in soil solution, adsorbed by organic and inorganic colloidal surfaces, firmly bound in soil minerals, deposited by compounds in the soil, and contained in living materials.

4.2 Pesticide

Generally, pesticides are widely used in agriculture and horticulture, containing heavy metals. For example, about 10% of pesticides in England contain compounds of Cu, Hg, Mn, or Zn. Fungicides are pesticides containing Cu, such as a mixture of Bordeaux (copper sulfate) and copper oxychloride. Lead arsenate has been used in orchards for many years to control several parasitic insects. Arsenic-containing compounds are also extensively used to control cow lice and banana pests in New Zealand and Australia. Wood preservation using formulations of Cu, Cr, and As pollute the air due to excessive concentrations of heavy metals [27, 48]. In Indonesia, the use of pesticides on vegetable crops is very intensive, especially on cash crops with high economic value. Based on research, 30–50% of the total production cost is used for pesticides [49]. Intensive use can increase soil and plant residues and even enter the bodies of animals, fish, or other aquatic biotas. Pesticides with a long half-life of degradation can harm the health of humans that consume products containing these residues.

Organic waste (biosolid) is a solid product produced through a wastewater treatment process and can be recycled. In the United States, it is estimated that more than 50% of the approximately 5.6 million tons of dry waste is used or distributed annually on land. Biosolids are applied to agricultural land in every region of the state. In European society, more than 30% of this waste is used as fertilizer. In Australia, 175,000 tons of dry biosolids are produced annually by local governments. Other biosolid materials are sawdust, rice/corn straw, or plant residues [50]. Heavy metal contamination of the soil may result from the continued application of biosolids. The most common heavy metals found in biosolids are Pb, Ni, Cd, Cr, Cu, and Zn, and the concentration depends on the intensity of industrial activity [51]. The application can be leached into the soil profile and potentially contaminate groundwater. It was shown that continued application of biosolid to several soils in New Zealand resulted in increased concentrations of Cd, Ni, and Zn in drainage water [52].

4.3 Industrial waste

It is estimated that 20 million hectares of agricultural land are irrigated with wastewater from industry or households. In several cities in Asia and Africa, studies show that agriculture based on wastewater irrigation accounts for 50% of the vegetable supply to urban areas. Farmers are less concerned about environmental benefits or harms, maximizing yields and profits. Even though the concentration of heavy metals in wastewater is relatively low, long-term irrigation can lead to its accumulation in the soil [53]. In Indonesia, heavy metal pollution has been identified in watersheds. According to [54], paddy soils in the Solo downstream watershed have been contaminated with Pb. Furthermore, paper mill effluent irrigation water has polluted the soil with Cu, Pb, and Cd. The Juwana sub-watershed indicates that heavy metal Cr contamination has the potential to contaminate agricultural land because the irrigation uses sugar industry waste. **Table 5** shows the industrial type producing some heavy metals.

Industry type	Hg	Pb	Cd	Cr	Cu	Zn	Ni	Al	Fe	Co	Mn
Plastic/resin	+	-	+	-	-	+	+	-	-	-	-
Pharmacy/cosmetic	+	+	-	-	-	+	-	-	-	-	-
Chlorine	+	-	-	-	-	-	-	-	-	-	-
Control/measurement tools	+	-	-	-	-	-	-	-	-	-	-
Electronics/electrical	+	-	-	-	-	-	-	-	-	-	-
Electroplating	-	-	-	+	+	+	+	+	-	-	-
Anti-rust paint	+	-	-	-	-	-	-	-	-	-	-
Textile	+	-	+	+	+	+	-	-	+	+	-
Ceramic	-	+	+	-	-	+	-	-	-	-	-
Skin tanning	+	-	-	+	-	-	+	-	-	-	-
Pulp and paper	+	+	-	+	-	-	-	-	-	-	-
Batteries	+	+	+	-	-	-	+	-	-	-	+
Soap/detergent	+	+	-	-	+	+	+	-	+	-	-
Metal/metal product	+	+	-	+	+	+	+	+	+	-	+
Pesticide	-	+	+	-	-	+	+	-	-	-	-

+ exist; - no exist.

Table 5.
Types of industries whose raw materials produce heavy metals [55].

5. Accumulation of Pb and Cd in soil and plant

5.1 Pb content in the soil

The results for Pb based on paddy soil use can be seen in **Tables 6** and **7**. The data show that the highest and lowest soil Pb content was found in 80-year-old rice fields and control areas, namely, 20.56 and 1.20 mg kg^{-1}, respectively. Compared with 20-year-old paddy fields, Pb content in 80 years was increased by around 16-fold. The Pb content in Pati, Central Java, and the lowland watershed of Solo Hilir, Lamongan Regency, ranged from 0.23 to 2.55 mg kg^{-1} [54] and 0.20–2.94 mg kg^{-1} [59]. In the highland rice fields of Wonosobo, Central Java, Gowa, South Sulawesi, and India, Pb content was 9.32–14.82 mg kg^{-1} [60], 0.0151 mg kg^{-1} [61], and 5.3–19.8 mg kg^{-1} [19]. Meanwhile, [17] reported that Pb content in paddy soil in Tanzania ranged from 8.0 to 28.5 mg kg^{-1}. The heavy metal content of Pb in the paddy field of Semarang Regency of Central Java is still below the standard provisions of India (250–500 mg kg^{-1}) and Europe (300 mg kg^{-1}) [62]. Possible sources of Pb include pesticide spraying by farmers and car exhaust near the village road [63].

Soil cultivation (year)	Soil Pb (mg kg^{-1})	Percentage increase (%)
Control (0)	1.20	
20	17.82	1.385,00
40	19.48	1.523,33
60	20.46	1.605,00
80	20.56	1.613,33

Table 6.
Soil Pb content and percentage increase based on the age of use of paddy fields [5].

No	Heavy metals (mg kg^{-1})			
	Soil	Pb	Cd	Hg
1	1	1.48	0.48	0.93
2	2	1.44	0.42	1.02
3	3	1.3	0.24	0.7
4	4	1.57	0.35	1.04
5	5	1.57	0.17	0.66
6	6	1.26	0.14	0.58
	Threshold value	0.5^{a}	3-8^{b}	0.3^{c}

[a] [56]
[b] [57]
[c] [58].

Table 7.
Content of various heavy metals in rice fields in Sidoarjo, East Java [9].

The relationship between soil Pb and time follows a logarithmic pattern, where the content increases with land use. The mathematical equation for soil Pb content as a function of time is Y = 11.88 + 2.02 Ln (x) and R^2 = 0.956, where Y = Pb content in soils (mg kg^{-1}) and x = age of land use (year).

5.2 Cd content in soil

The soil Cd analysis results based on paddy fields can be seen in **Table 8**. The table shows that the highest and lowest Cd content was found in 80- and 20-year-old rice fields, namely, 0.72 and 0.26 mg kg^{-1}. Compared with 20-year-old paddy fields, the highest soil Cd increase was in 80-year-old fields by 1340%. Pradika et al. [64] also reported that P fertilization could add Cd metal to agricultural land. This is because the raw material for making P fertilizer comes from phosphate rock, which naturally contains Cd metal. The concentrations in surface soils range from 0.06 to 1.10 mg kg^{-1} with an average of 0.41 mg kg^{-1} [65]. Satpathy et al. [19] reported that Cd content in Indian paddy soils ranged from 0.02 to 0.6 mg kg^{-1}.

The relationship between soil Cd content and time follows an exponential pattern, where the content increases with the age of land use. The mathematical equations as a function of time are Ln (Y) = 0.17 + 0.02 x and R^2 = 0.913, where Y = Cd content in soil (mg kg^{-1}) and x = age of land use (year). The presence of heavy metals Pb and Cd in the soil can be caused by the intensive use of P fertilizer and exceeding the recommended dose. Heavy metals Pb and Cd can increase in line with the age of paddy fields.

5.3 Pb content in rice

The results of the Pb analysis of rice based on the age of the paddy fields can be seen in **Table 9**. The data show that the highest and lowest Pb content was found in 80- and 20-year-old rice fields, namely, 3.11 and 2.35 mg kg^{-1}. The Pb content has exceeded the critical limit set by WHO, which is 2 mg kg^{-1}. The Pb content in grain from paddy fields in Pati, Central Java, and Wonosobo highlands ranged from 0.23 to 1.23 mg kg^{-1} [54] and 0.28–1.32 mg kg^{-1} [60]. In the present study, the concentration of Pb was found to be higher in roots than in shoots and grains [19].

The relationship between the Pb content of rice with time follows an exponential pattern. The mathematical equations as a function of time are Ln (Y) = 2.06 + 0.005 x and R^2 = 0.928, where Y = Pb in rice (mg kg^{-1}) and x = age of land cultivation (year).

Soil cultivation (year)	**Soil Cd (mg kg^{-1})**	**Percentage increase (%)**
Control (0)	0.05	
20	0.26	420
40	0.32	540
60	0.39	680
80	0.72	1,340

Table 8.
Soil Cd content and percentage increase based on the age of use of paddy fields [5].

Age of soil (year)	Pb in rice (mg kg^{-1})	Percentage increase (%)
20	2.35	
40	2.40	2.12
60	2.86	21.70
80	3.11	32.34

Table 9.
Pb levels in rice based on the age of use of paddy fields [5].

5.4 Cd content in rice

Cadmium can affect enzyme activity in plants, leading to lower photosynthesis. Therefore, growth and development, including germination, root elongation, and leaf expansion, can be decreased [65–67]. The Cd analysis of rice based on the age of the paddy fields can be seen in **Table 10**. It shows that the highest and lowest Cd content is found in rice fields aged 80 and 20 years, namely, 0.29 and 0.15 mg kg^{-1}. The content of Cd in 80-year-old rice fields has exceeded the critical limit set by WHO, which is 0.24 mg kg^{-1}. In the shoots, the concentration was higher than that in roots and grains [19]. Jarvis et al. [68] reported that Cd was easily taken up by plants and transported to different parts, but it is nonessential and has no beneficial effects on plants.

The relationship between the Cd content of rice with time follows an exponential pattern. The mathematical equations as a function of time are Ln (Y) = 0.11 + 0.011 x and $R^2 = 0.934$, where Y = Cd in rice (mg kg^{-1}) and x = age of land use cultivation (year).

The high content of Pb and Cd was caused by the content in the soil and the low fertility of paddy fields. Lowland rice plants can absorb dissolved Pb and Cd in the soil and accumulate them in large quantities from their tissues. The elements contained in plant tissue in the vegetative phase will be translocated to fruit during vegetative growth. Aprilia and Purwani [69] added that heavy metals Pb and Cd are more easily absorbed by plant roots in the form of Pb^{2+} and Cd^{2+} ions. Munaf [70] explained that the accumulation of Pb and Cd in plant tissues derived from the absorption of roots could be influenced by several factors. These include the solubility properties of compounds in soil solution, pH, organic C content, cation exchange capacity (CEC), and clay content.

Age of soil (year)	Cd in rice (mg kg^{-1})	Percentage increase (%)
20	0.15	
40	0.16	6.66
60	0.21	40.00
80	0.29	93.33

Table 10.
Cd content in rice based on the age of use of paddy fields [5].

6. Conclusion

Rice fields in Indonesia are used very intensively, and the fertility is negatively affected following the decrease in pH. Furthermore, the paddy fields in Indonesia have experienced heavy metal pollution, especially Pb and Cd, which is indicated by their accumulation in soil and rice. These heavy metals come from phosphate fertilizers and industrial waste. The proposed solution uses organic materials to immobilize the metal or form ligand bonds. Cadmium and lead input from phosphate fertilizers threaten the environment and human health due to soil contamination, crop absorption, and bioaccumulation in the food chain. A decrease in non-polluted recycled and mineral P fertilizer dependence could alleviate the Cd and Pb pollution of the paddy soils.

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