Chapter 4

Remediation of Heavy Metals Polluted Soils in Indonesia

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A. Persistent Organic Pollutants (POPs) in General Understanding

Soil is a natural element that is the same important as water and air. Soil is a natural object, part of the Earth's surface that can be overgrown by plants and has characteristics as a result of the work of climatic factors and living things on the parent material, which is influenced by topographical conditions within a certain period of time. Soil, as a natural resource for agricultural purposes, has two main functions, as a source of nutrients for plants and as a medium where plant roots are anchored, water and nutrient is stored.

Excessive use of natural resources such as the conversion of agricultural land to built-up land for settlement or industry to sup-

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port human needs at the current level puts excessive pressure on the environment. Increased development, including industry, is not only able to absorb labor but also causes negative impacts if it is not managed properly. One example is industrial activities that dispose of waste into rivers/water bodies used as sources of irrigation water.

Waste that is disposed of into water bodies and becomes a source of irrigation water for agricultural land that is still productive has an impact that is not only detrimental to the sustainability of farming on that land but can result in damage to the ecosystem. One of the impacts that occur on agricultural land is soil contamination.

Since 1978 in the Bandung region, Indonesia, to be precise, the Rancaekek sub-district and its surroundings have developed into a textile industrial area, and up to now, there are more than thirty textile factories located between Rancaekek–Cicalengka (Adji, 2006). A large number of factories has an impact on the wastewater that is flowed through the surrounding rivers, especially if the wastewater treatment plant from the factory is not functioning optimally.

Cikijing river water and rice fields in Rancaekek District, Bandung, Indonesia, which are close to the industrial area, contain heavy metals Ni, Zn, Cu, Cd, Co, Cr, and Pb (Suganda et al., 2002). In addition, the rice fields in Rancakeong Block, Linggar Village, Rancaekek District contain heavy metals Zn, Ni, Cd, Co, Cr, Cu, and Pb (Adji, 2006). Both total and available (ionic) values in the soil. The presence of all available values of these heavy metals has been successfully absorbed in plant roots and translocated into plant tissue.

Various contaminants that enter the soil will result in decreased soil function as one of the causes of soil damage or soil degradation. Soil degradation will be followed by a decrease in land productivity. This can cause serious problems because it can harm farmers and hinder efforts to increase the production and safety of agricultural products, which in turn can threaten national food security.

Heavy metal was all metal elements that have a specific gravity with a value of more than 5 g/cm^3 (Pierzynski et al., 2015). In the earth's crust, metals are divided into macro metals and micro metals

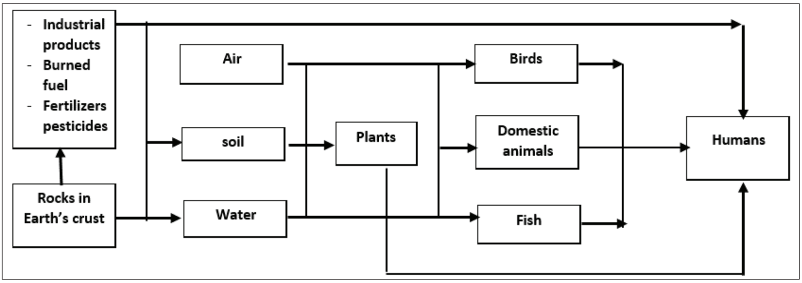
(Darmono, 1995). Macro metals consist of iron (Fe), calcium (Ca), magnesium (Mg), sodium (Na), manganese (Mn), potassium (K), and aluminum (Al). Micro metals include barium (Ba), lead (Pb), zinc (Zn), nickel (Ni), tin (Sn), copper (Cu), uranium (U), cadmium (Cd), silver (Ag), mercury (Hg), and gold (Au). Some of these metals are categorized as metals that are more potentially toxic to humans, are Pb, Ni, As, Hg, Zn, Cu, and Cd.

When plant parts that absorb heavy metals such as leaves and fruit/seeds are consumed by animals or humans, it will gradually cause poisoning for those who consume them. Therefore, it is necessary to improve the nature of the contaminated soil so that the plants can reproduce adequately. Efforts that can be done in restoring soil pollution are remediation. Remediation can be defined as the process of recovering from a contaminated condition so that it is clean again using plants (phytoremediation) or microbes (bioremediation).

The purpose of this paper is to further describe the problem of heavy metals and the influence of anthropogenic activities on soil pollution, describe remediation techniques for heavy metal polluted soil based on the results of recent research in Indonesia, and provide recommendations for phytoremediation modeling.

B. Heavy Metals Problems and its Remediation Technique

Mining activities have affected health due to contamination of local water sources and have a detrimental effect on the environment, mining of coastal sand which causes erosion, or long-term effects in reducing biodiversity or increasing the level of fish mortality (WHO, 2008). The disposal of tailings is carried out in the environment, which is usually disposed of in artificial reservoirs, rivers or lakes, and the sea. In mineralogy, tailings can consist of various minerals such as silica, iron silicate, magnesium, sodium, potassium, and sulfide. Among these minerals, sulfides have chemically active properties and, when in contact with air, will undergo oxidation to form acidic salts, and the acid stream contains a number of toxic metals such as



Source: Farcasanu et al. (2018)

Figure 4.1 Various Sources of Heavy Metal Pollution and Their Cycle

cadmium, mercury, arsenic, lead, and mercury which can pollute or damage the environment (UNO. 1995).

The research results reported that heavy metal contamination occurred in gold mining areas, industrial waste disposal, and agriculture (fig. 4.1.). Community gold mining waste such as Pongkor, West Java contains up to 240 ppm Hg and 0.1 ppm Cd and is simply wasted in the surrounding environment both in the rice fields and the Cikaniki River (Hidayati N et al., 2004). Textile industrial waste containing heavy metals reaches 296.5 thousand tons per year which pollutes the rice fields and Cikijing River, Bandung (Rija S, 2000). Meanwhile, heavy metals Pb and Cd from motor vehicles contaminated rice fields in North Coast Java (Pantura) area, covering 40% of the 105,557 ha of rice fields in the Karawang-Bekasi area (Kasno et al., 2000). In the Cirebon–Palimanan area, Pb contamination in rice fields reaches 30.08 ppm, causing the Pb content in rice approach the danger threshold for consumption (Miseri & Santoso, 2000).

The existence of industrial activities in coastal areas causes the disposal of a number of organic chemicals, heavy metal compounds, and compounds from other industrial wastes. Industrial wastes containing toxic compounds released into the environment will have a negative and toxic effect on organisms in food webs (Dembitsky & Rezanka, 2003). Chemical properties and biological functions toxicity varies depending on the concentration and type of metal. Cr, Pb, Cu, Zn, Ni, Hg, Cd, and Co are very toxic both in the basic form and in the form of dissolved salts. The presence of these compounds in aquatic, soil, and atmospheric environments causes very serious problems for organisms. The most dangerous cause for human health is its bioaccumulation into the food chain. The most common cause of heavy metal contamination is the consumption of food and drinking water.

Identification of carcinogenic compounds such as arsenic takes a long time. Pollution of arsenic compounds in the soil has resulted from human activities, including pesticides, burning, wood preservation, and mining. Around ten thousand sites worldwide are contaminated with arsenic with the highest concentration being 26.5 mg/kg (Hingston et al., 2001)

C. Heavy Metals Remediation Technique

Several techniques used to remediate heavy metal polluted soil are containment, solidification, vitrification, soil washing, soil flushing, pyrometallurgy, electrokinetic, and phytoremediation (Chen et al., 2016). *Containment* measures are often carried out as measures to prevent or significantly reduce the migration of contaminants in soil or groundwater. *Solidification/stabilization* using cement is an alternative waste treatment with the aim of reducing the environmental pollution.

Vitrification is a remediation technique in which a substance is transformed into glassy materials. *Soil washing* is a remediation technique based on the theory that prone contaminants bind to fine-grained soils (silt and loam), which tend to bind to coarse-grained soils. These techniques include ex-situ remediation, which involves removing harmful pollutants from the soil by washing the soil with liquid, scrubbing the soil, and then separating the clean soil from contaminated soil and washing water (Dellisanti et al., 2009).

Soil flushing is an in-situ remediation technology that can occur in an unsaturated zone, a saturated zone, or both. The flushing solution increases the mobility or solubility of the adsorbed contaminants to the soil matrix. *Pyrometallurgy* is a process of metallurgical extraction using heat energy. The temperature reached up to 2000°C. *Electrokinetic* remediation is a technology for restoring soil contaminated with heavy metals and organic compounds through an in-situ process using low voltage and constant direct current (DC).

Phytoremediation is a technique to use plants and their parts to decontaminate waste and environmental pollution problems either ex-situ using artificial ponds or reactors or in-situ or directly in the field on soil or areas contaminated with waste (Subroto, 1996). Phytoremediation is also characterized as a form of contamination retention that is intervened by non-food plants, including trees, aquatic plants, and grasses.

D. Remediation of Polluted Soils in Indonesia

The remediation techniques commonly used in Indonesia are phytoremediation and bioremediation. Both of these techniques are considered more efficient and do not require large costs. Phytoremediator plants used are derived from local plants, some are tree species. Examples of phytoremediator plants used are *Ipomoea aquatic, Pennisetum Purpuroides, Boehmeria nivea, Cordyline fruticosa, Sansevieria Trifasciata, Celosia Plumosa*, and *Chrysopogon zizanioides*. Whereas bacteria used in remediating heavy metal contaminated soil are using indigenous bacteria and mycorrhizal bacteria. Recent research related to remediation of heavy metal contaminated soil in Indonesia is presented in Table 4.1.

Table 4.1 Recent Research on Heavy Metal Remediation Polluted Soil in Indonesia

Recent Research	Author, Year
Effect of Compost and Biochar on Phytoremediation of Soil Contaminated with Cadmium from Lapindo Mud Volcano Using Water Spinach	(Lestari & Aji, 2020)
Study on The Addition of EDTA to the Phytoremediation of Lead	(Nadhila et al., 2021)
Potential of Phytoremediation of Lead Contaminated Soil (Pb) with EDTA Addition Using King Grass (Pennisetum Purpuroides)	(Anugroho et al., 2020)

Recent Research	Author, Year
The Potential of Rami (Boehmeria nivea) for Phytoremediation of Copper Contaminated Soil	(Lestari & Pratama, 2020)
Phythoremediation Lead Metals Contaminated Soils (Pb) Using Sansevieria Trifasciata and Celosia Plumosa	(Ratnawati & Fatmasari, 2018)
Bioremediation of Lead Using Indigenous Bacteria Isolated from Leachete Contaminated Soil	(Rahadi et al., 2020)
Remediation of Cobalt (Co) polluted Soil Using Bioremediator and Ameliorant	(Purbalisa & Dewi, 2019)
Bioremediation of Pb by Agar Liquid Waste Indigent Bacterial	(Ikerismawati, 2019)
Bioremediation of Pb in Textile Waste with Staphylococcus Aureus and Bacillus Subtilis	(Maulana et al., 2017)
Heavy Metals Remediation Polluted Soil by Using Biochar	(Hidayat, 2015)
Bioremediation of Soils Contaminated Heavy Metals Cd, Cu, and Pb by Using Endomikoriza	(Chairiyah et al., 2013)

One of the sources of heavy metal pollutants in Indonesia comes from gold mining waste.. The main activity in gold mining is the process of separating gold ore from rock using metals which in turn will produce waste. The waste obtained can be in the form of solids (tailings) and liquid waste. Kuranchie et al., (2013) explained that obtaining or it will produce several forms of waste, namely tailings, slugs, dust, and liquid waste. This waste can spread widely to the surrounding area if there is no proper handling.

Gold mining waste in the form of solids or tailings has characteristics such as sand with a fine texture, low organic matter content, acidity, and high heavy metal content (Fashola et al., 2016). Heavy metals contained in these wastes include Hg, As, Pb, Cd, Cr, and Cu (Ngure et al. 2017; Xiao et al. 2017). Lead is a heavy metal with a high enough concentration in gold mine tailings waste (Siregar & Zakiyah, 2016).

Textile waste is waste generated in the testing process, starch removal, bleaching, cooking, dyeing, printing, and refinement processes. The cotton refinement process produces more and stronger waste than the waste from the synthesis material refinement process. Waste originating from the textile dyeing process is a major factor in environmental pollution problems. Water pollution from the textile industry can come from wastewater from the production process, waste from lubricants and oil, chemical waste from the production process, waste from scraps of cloth, and others. Some water pollutants include dyes, high salt concentrations, and heavy metals. The resulting waste includes heavy metal arsenic, lead, cadmium, zinc, chromium, copper, halogenated hydrocarbons (from the dressing and finishing process), pigments, dyes, tensioactives (surfactants), and organic solvents (MEI, 2005).

Acid Mine Water (AAT) is wastewater from mining with high acidity (low pH, <4). AAT arises from the reaction results of the oxidation of rocks exposed to water, resulting in sulfuric acid and iron hydroxide deposits (Achterberg et al. 2003). The yellowish color that settles at the bottom of the mining channel or on the walls of the silt deposition pond is a visual representation of iron hydroxide deposits (Aubé, 2004).

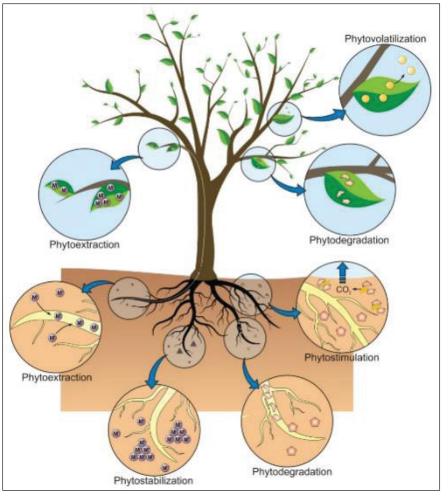
E. Case Study of Phytoremediation and its Uptake Model

1. Phytoremediation of Heavy Metals using Vetiveria zizanioides

Phytoremediation is a technique for recovering polluted land by using plants to clean, absorb, degrade, transform and immobilize pollutants, both heavy metals, and organic and inorganic compounds. Vetiver or nard is a type of grass plant originating from India. This plant can be grown throughout the year and is known to many people for a long time as a source of perfume. In Indonesia, the vetiver plant is used as an herbal product in the form of essential oils which are useful in relieving inflammation, aromatherapy, floor cleaner, preventing bacterial infections, increasing immunity, treating insomnia, and accelerating wound healing (Mulyono et al., 2012). This plant is categorized as a non-food crop and not for consumption by living things, with the result that this plant is very suitable to be used to remediate pollutants.

Plants generally have several mechanisms for removing pollutants in the soil (Figure 4.2). Phytoremediation runs naturally with a six-stage process carried out by plants for contaminants or surrounding pollutants. Phytoextraction is the absorption of pollutants by plants from the air or soil through the roots which are then stored in the plant canopy. This type of plant is known as a hyperaccumulator which have the ability to store specific metals in high concentrations in the aerial portion (0.01% to 1% dry weight, depending on the metal). Phytofiltration is a process that uses plants to absorb, collect, and precipitate contaminants, especially heavy metals or radioactive elements, from water media through root systems or other submerged organs.

Phytostabilization, organic or inorganic contaminants, are introduced into the lignin in the cell wall from the root of the cell to

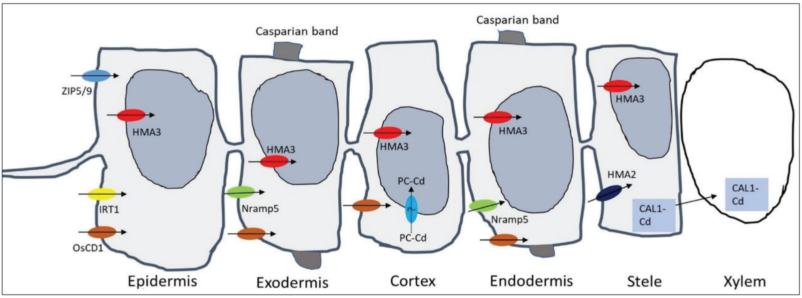


Source: J.C. et al. (2014)

Figure 4.2 Schematic Representation of Phytoremediation become humus. The metal is precipitated in an insoluble form by root exudate and is then trapped in the ground matrix. In rhizodegradation growing roots can encourage the development of damaging rhizosphere microorganisms and utilize exudates and plant metabolism as a source of carbon and energy. In addition, plants can release their own biodegradation enzymes. The use of rhizodegradation is limited to organic contaminants. In phytodegradation, organic contaminants are degraded (metabolized) or mineralized in plant cells by specific enzymes including nitroreductase, dehalogenase (degradation of chlorinated solvents and pesticides), and laccases (degradation of aniline) (Ryllot et al., 2008).

Phytovolatilization is the process of absorbing pollutants by plants and these pollutants are converted to evaporative properties which are then transpired by plants. Here, the absorbed pollutants will be released by the plants into the air. In the periodic table, some ions in group IIB, VA, and VIA elements, especially mercury, selenium, and arsenic are absorbed by plant roots and converted into non-toxic forms which are then volatilized into the atmosphere (Ali et al., 2013).

Vetiver (*Vetiveria zizanioides*) is a metal hyperaccumulator plant that has high absorption or accumulation properties in its plant tissues (MacGrath et al., 1993). Although in Indonesia it is included as a spice plant, the vetiver plant does not require special growing requirements



Source: Luo & Zhang (2021)

Figure 4.3 Heavy Metals Uptake and Transport Schematic in Roots like other plants. With a massive root system, this plant can grow well in extreme soil and environmental conditions (Truong, 2001). This plant has a very high tolerance to drought conditions or water stress, flooding conditions, hot environmental temperatures, tolerance to very acidic soil pH conditions, resistance to aluminum and manganese toxicity, and very tolerant to various metals such as arsenic, cadmium, copper, chrome, and nickel (Benavides et al., 2021). Recent research also supports that vetiver can survive in acid mine drainage soils (Kiiskila et al., 2019).

Heavy metals in the soil such as cadmium are in an insoluble form and are not available to plants. However, the solubility of heavy metals in soil can be increased by plants through the release of exudate substances in plant roots which will change the pH conditions in the rhizosphere (Dalvi et al., 2013). Two pathways lead to the physiological absorption of heavy metals through apoplastic and symplastic routes. The apoplastic pathway is a passive absorption process by diffusion. While the symplastic pathway is an active transport absorption process and an energy-consuming plasma membrane transport process that requires a gradient potential and electrochemical concentration (Peer et al., 2013).

Heavy metal complexes with various chelating agents will be formed when the heavy metals enter the root system. As a result, the toxicity will be lost because it does not move from the cytoplasm or vacuole, the cell wall (Ali et al., 2013). Plant cell walls or vacuoles are the sites of the detoxification of heavy metals. Heavy metal toxicity will disappear after storage in the root vacuole, this will also reduce its transportation which is quite far to the top of the plant (Thakur et al., 2016).

Until now, in most living organisms, the biological activity of certain heavy metals such as cadmium has not been known. Some heavy metals in hyperaccumulator plants such as nickel and zinc have the function of plant resistance to herbivorous organisms (Jiang et al., 2005). Cadmium in the plasma membrane will be absorbed as a carrier of essential elements through the roots, which have low substrate selectivity (Clemens et al., 2001).

2. Phytoremediation Uptake Model

The absorption of heavy metals in the phytoremediation process can be described through a simple system uptake plant model. This model is used to predict the contribution of various pathways through which heavy metals travel on different plant parts such as leaves, stems, and roots. This model includes three paths, namely the soil-root-leaf path, the soil-air-leaf path, and the path (Ouyang & Wan, 2008). The Simple Absorption Model is represented by four types of models, namely the soil model, root model, stem model, and leaf model (Ibrahim & el Afandi, 2020).

Soil Model, the bioaccumulation factors (BCF) approach which is commonly used in soil models is actually easy to use, but the limitation of this approach is that the lack of accuracy will greatly reduce its usefulness as a tool for risk evaluation and decision-making. Therefore, another parameter used besides BCF is the distribution coefficient between water and soil in dry conditions (Mouchet, 2008).

In this modeling experiment, experiments in the lab were carried out by determining in advance the concentration of heavy metals, namely heavy metals in water used to irrigate plants and soil using polyethylene bottles that had been washed and pre-soaked in the acid solution for 24 hours. The samples were then conditioned in an acid state until they reached pH 2. The concentration of heavy metals was then measured using AAS which was calibrated using a standard solution. Then the calculation follows the following equation:

$$CMn - Kd * Cw, \text{ where:}$$

CM: mass plant concentration; Kd: Distribution Coefficient of Hm; Cw: Hm concentration in water.

$$Cw/Csoils - KWS, \text{ where:}$$

Cw: Hm concentration in water; Csoil: Hm concentration in soil; Kws: Partition coefficient of Hm (between two phases soil and water)

Root Model, determination of the root model. It is represented by the following equation: Mass balance: flux in – flux out is given by:

Cw: Hm concentration in water, Csoil: Hm concentration in soil, Kd: distribution coefficient. Concentration divided by plant mass:

Cw – Csoils / KWS, where:

Cxy: concentration of the Hm inside the xylem; Cr: concentration of Hm in the root; Krw: Partition coefficient of Heavy metals in two phases root zone and water

The steady-state is important to consider because the chemical conditions of the solution in the rhizosphere to which the plants are actually exposed are very different from the chemistry of bulk soil solutions and must be properly quantified (Paquin et al., 2002). On the other hand, the degree of accumulation at the site of action (biotic ligand) is assumed to be related to the toxicological response. Set to steady conditions and settle for heavy metal concentrations in plant roots.

$$((Cw * Q) - \left(\frac{Cr * Q}{Krw * M}\right) - (K * Cr))$$

$$Cr - \left(\frac{Q}{\frac{Q}{Krw}} + KM\right) * \left(\frac{Csoil}{Ksw}\right)$$, where:

Q: Flow of water in the soil (ml/kg); K: Rate of change in time; M: Plant mass; KM: Rate of change in mass of the plant; Ksw: Partition coefficient of heavy metals in two phases of soil and water

Stem model, stem model calculated according to xylem phloem (Briggs & Sculpher, 1998; Dettenmaier et al., 2009) in the following equation:

TSCF – 0.784*EXP ((- logKow – 1.78/2.44)2), where:

TSCF: Translocation stem concentration factor

Leaf Model, Leaf model is calculated in the following equation:

*LogBCF – 0.578*logKow + 1.588,* Where:

BCF: Bioaccumulation factors; Kow: Partition coefficient of heavy metals between two phases (organic compound) and water

$$\text{Outflux: } \left(\frac{Q}{Mr} * Kws\right) * Cs - \left(\frac{Q}{Mr} * Krw\right) * Cr - (Kr * Cr),$$

where:
Q: Flow of the water from the soil to the root (ml/kg); Mr: Mass of the root; Kws: Partition coefficient of Heavy metals between two phase waters; Cs: Concentration of Heavy metals in soil; Kr: Rate of change in the weight of the root.

$$\text{Influx to leaves: } \frac{Cl}{K} - (Q(Ml + Krw) * Cr), \text{ where:}$$

K: Rate of change in days; Q: water flow through the stem to the leaves (ml/kg); Cl: Heavy metals concentration in leaves; Cw: Heavy metals concentration in water; Ml: leaves mass; Kaw: Partition coefficient of Heavy metals (between two-phase air and water); Ca: Concentration of Heavy metals in air sample;

F. Conclusion

Heavy metal pollution of soil in Indonesia is mostly caused by the activities of the mining and the textile industry. Heavy metal pollution that occurs has entered the area of agricultural land for food crops so that heavy metals are found to have accumulated in plant tissues that can be dangerous to humans' health. *Vetiveria zizanioides* plants are the best candidates for heavy metal hyperaccumulator plants in the phytoremediation process based on their high absorption of heavy metals and non-food crops, so they can be further utilized as non-consumable products. A simple Plant Uptake Model (UPM) system has been used successfully in estimating heavy metal uptake in soil and its accumulation in plant tissues (stems, roots, and leaves). This model can be used in the management of phytoremediation related to the type and ability of plant species according to polluted soil and environmental conditions.

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