7
Heavy Metal Removal by Crops from Land Application of Sludge

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Contents

Abstract This chapter describes the application of phytoremediation in removing heavy metals from contaminated soils. The types of crops used as well as the characteristics and application of sludge in Malaysia are described. The standards and regulations of sludge application are also discussed. The chapter gives a detailed discussion of principles of phytoremediation and design parameters used in the design of the treatment systems. Moreover, a few case studies and design examples are covered in the chapter.

1. INTRODUCTION

The use of plants to treat wastes was investigated as early as 1967 as researchers investigated plants ability to uptake and translocate contaminants (1). Since then, this technique has evolved into a cost effective technology to remediate hazardous constituents from contaminated land. The present development of phytoremediation technology is being driven primarily by the high cost of many other soil remediation methods as well as a desire to use a "green" sustainable process. Some specific examples include the successful application of Phytoremediation technology in a small pond at Chernobyl with uranium contamination,

From: *Handbook of Environmental Engineering, Volume 11: Environmental Bioengineering*
Edited by: L. K. Wang et al., DOI: 10.1007/978-1-60327-031-1_7

an engineered wetland at Milan, Tennessee for TNT removal and a riparian zone buffer strip at Amana, and Iowa for nitrate and atrazine removal from agricultural runoff. Applications at small sites were also successful in their agricultural cooperatives with pesticide and ammonia spills. It is also found that long-term monitoring and evaluation of phytoremediation technology are essential to demonstrate efficacy and further define suitable plants and applications in order to gain acceptance from regulatory agencies (2). In Malaysia, phytoremediation application in general has been limited to research only on a small-scale basis. As to date, actual remedy of heavy metals and organics has not been applied on real sites in Malaysia. Nevertheless, the Ministry of Science, Technology and Environment (MOSTE) encourages the phytoremediation research in terms of providing research funds, while the local Institution of Engineers (IEM) takes a leading role in bringing awareness of this technology to the related professional bodies and regulatory agencies.

1.1. Definition of Phytoremediation

Phytoremediation is an emerging technology that uses plants and their associated rhizospheric microorganisms to remove various pollutants from contaminated soils, sludge, sediment, groundwater, and surface water through contaminant removal, degradation, stabilization, or containment of the contaminant (3). It uses living plants for in situ and *ex situ* remediation of contaminated sites. The plants also help prevent wind, rain, and groundwater from carrying pollutants away from sites to other areas (2). It can be used to remediate various contaminants, including metals, pesticides, solvents, explosives, petroleum hydrocarbons, polycyclic aromatic hydrocarbons, and landfill leachate. It is also applicable to point and nonpoint source hazardous waste control (3).

The word "phytoremediation" is from the Greek prefix *phyto*-meaning "plant" and the Latin root word *remidium* – meaning "to correct or remove an evil." In soil, the "evil" could be anthropogenic (man-made) contaminants such as organic solvents, heavy metals, pesticides, or radionuclides (4). In general, the mechanism of phytoremediation is mainly living plants altering the chemical composition of the soil matrix in which they are growing. This is achieved by one of the five applications of phytoremediation, namely Phytotransformation, Rhizosphere, Bioremediation, Phytostabilization, Phytoextraction, or Rhizofiltration.

Phytoremediation is best applied at sites with shallow contamination of organic, nutrient, or metal pollutants. It has been utilized at a number of pilot and full-scale field demonstration tests in the United States. It is an emerging technology that should be considered for remediation of contaminated sites because of its cost effectiveness, aesthetic advantages, and long-term applicability. Phytoremediation is well suited for use at very large field sites, where other methods of remediation are not cost effective or practicable; at sites with low concentrations of contaminants, where only "polishing treatment" is required over long periods of time; and in conjunction with other technologies, where vegetation is used as a final cap and closure of the site (5).

Initially, much interest was focused on hyperaccumulator plants that are capable of accumulating potentially phytotoxic elements to concentrations more than 100 times than those found in nonaccumulators (6–8). These plants have strongly expressed metal sequestration mechanisms and, sometimes, greater internal requirements for specific metals (9). Some species may be capable of mobilizing metals from less soluble soil fractions in comparison with nonhyperaccumulating species (10). Metal concentrations in the shoots of hyperaccumulators normally exceed those in the roots, and it has been suggested that metal hyperaccumulation has the ecological role of providing protection against fungal and insect attack (7). Such plants are endemic to areas of natural mineralization and mine spoils (11). Examples include species of Thlaspi (*Brassicaceae*), which can accumulate more than 3% Zn, 0.5% Pb, and 0.1% Cd in their shoots (12, 13), and Alyssum (Brassicaceae), some species of which have been shown to accumulate over 1% Ni (14).

Exploitation of metal uptake into plant biomass as a method of soil decontamination is limited by plant productivity and the concentration of metals achieved (12). For instance, *Thlaspi caerulescens* is a known Zn hyperaccumulator, but its use in the field is limited because individual's plants are very small and slow growing. The ideal plant species to remediate a heavy metal-contaminated soil would be a high biomass-producing crop that can both tolerate and accumulate the contaminants of interest (15). Such a combination may not be possible; there may have to be a trade-off between hyperaccumulation and lower biomass, and vice-versa. Furthermore, the cropping of contaminated land with hyperaccumulating plants may result in a potentially hazardous biomass (16).

1.2. Heavy Metals in Soil

A heavy metal is a term usually applied to a large group of trace elements with an atomic density greater than 6 g/cm^3. The heavy metals, which tend to give rise to the greatest amount of concern with regard to the human health, agriculture, and ecotoxicology are As, Cd, Cr, Hg, Pb, Tl, and U (17). Heavy metals occur naturally in soils, usually at relatively low concentrations, as a result of the weathering and other pedogenic processes acting on the rock fragments on which the soils develop (soil parent material). Although heavy metals are constantly encountered in soil parent materials, such as igneous or sedimentary rocks, the major anthropogenic source of metals to soils and the environment are as follows:

(a) Metalliferrous mining and smelting
(b) Agricultural and horticultural materials
(c) Sewage sludge
(d) Fossil fuel combustion
(e) Metallurgical industries – manufacture, use, and disposal of metal commodities
(f) Electronics – manufacture, use, and disposal of electronic commodities
(g) Chemical and other manufacturing industries
(h) Waste disposal
(i) Sports shooting and fishing
(j) Warfare and military training.

Plants have the potential to enhance remediation of the following types of contaminants:

(a) Petroleum hydrocarbons
(b) Benzene, toluene, ethyl benzene, and xylene (BTEX)
(c) Polycyclic aromatic hydrocarbons (PAH)

(d) Polychloroethene biphenyls (PCB)
(e) Trichloroethene (TCE) and other chlorinated solvents
(f) Ammunition wastes and explosives
(g) Heavy metals
(h) Pesticide waste
(i) Radio nuclides
(j) Nutrient waste (such as phosphates and nitrates) (18).

1.2.1. Natural Content of Heavy Metals in Soil

Trace elements are those inorganic chemical elements that in very small quantities can be essential or detrimental to plants and animals. Trace elements occur as trace constituents of primary minerals in igneous and sedimentary rocks. Since soil is considered the products of in situ weathering of all rock types, the trace element concentrations in soil may then be linked to the types of parent material and the interactions with climate, organisms, and time. Table 7.1 shows the mean concentrations of selected trace metals in a range of representative types of igneous and sedimentary rocks, while Table 7.2 shows the mean concentrations of various types of surface soils.

Although Table 7.1 illustrates that trace metals are commonly present in soil parent materials, anthropogenic sources may also increase the background concentration of the soil sometimes to dangerously high levels. Table 7.2 shows that the creation of lead free housing is not nearly sufficient, as lead in soil has been shown to be a major contributor to the high lead levels present in children. There is a need for a plan, which will eliminate the soil lead exposure pathway because there is major health associated with lead exposure, especially in children (4).

Table 7.1
Mean selected trace metal contents of major rock types (mg/kg) (17)

Elements	Earth's crust	Igneous rocks			Sedimentary rocks		
		Ultrabasic	Basic	Granitic	Limestone	Sandstone	Shales and clays
Arsenic	1.5	1	1.5	1.5	1	1	13
Cadmium	0.1	0.12	0.13	0.09	0.028	0.05	0.22
Chromium	100	2,980	200	4	11	35	90
Cobalt	20	110	35	1	0.1	0.3	19
Copper	50	42	90	13	5.5	30	39
Lead	14	14	3	24	5.7	10	23
Manganese	950	1,040	1,500	400	620	460	850
Mercury	0.05	0.004	0.01	0.08	0.16	0.29	0.18
Molybdenum	1.5	0.3	1	2	0.16	0.2	2.6
Nickel	80	2,000	150	0.5	7	9	68
Selenium	0.05	0.13	0.05	0.05	0.03	0.01	0.5
Tin	2.2	0.5	1.5	3.5	0.5	0.5	6
Zinc	75	58	100	52	20	30	120

Table 7.2
The mean concentrations of various types of surface soils (17)

Element	Sandy soils	Silty and loamy soils	Organic soils
Arsenic	4.4	8.4	9.3
Cadmium	0.37	0.45	0.78
Chromium	47	51	12
Cobalt	5.5	10	4.5
Copper	13	23	16
Lead	22	28	44
Manganese	270	525	465
Mercury	0.05	0.1	0.26
Molybdenum	1.3	2.8	1.5
Nickel	13	26	12
Selenium	0.25	0.34	0.37
Zinc	45	60	50

1.3. Heavy Metals from Sludge

All sludge contains a wide range of metal and other contaminants in varying concentrations. Industrial sludges usually contain higher metal contents than suburban domestic sludges. However, domestic inputs of metals to the sewerage system are still not insignificant, being derived from the corrosion of metal plumbing fittings, excretion of metals in the human diet, cosmetics, healthcare products, and other domestic products. It has been estimated that in the UK, 62% of the Cu and 64% of the Zn were from domestic sources. The heavy metals most likely to cause problems for crop production on sludge-amended soils are Cd, Cu, Ni, and Zn (17). The ranges of values found in the literature for the concentrations of heavy metals in sewage sludges are given in Table 7.3.

1.4. Land Application of Sludge

1.4.1. Sewage Sludge Generation

Increasing industrialization and urbanization have resulted in a dramatic increase in the volume of wastewater produced around the world. Treatment of this wastewater resulted in various pollutants being concentrated or thickened into a sludge containing between 1 and 2% by weight dry solids. The dramatic increase in the volume of wastewater treated also resulted in large volumes of sludge, which required proper disposal in a safe manner. In the early 1990s, the UK produced 1.1 million tones dry sludge solids per year, while the USA produced 5.4 million tones. In the whole of European Community, 6.3 million tones of sewage sludge were being produced, including West Germany producing 2.5 million tones, France 0.7, the Netherlands 0.28, and Switzerland 0.215 million tones. Japan produced 1.1 million tones of dry sludge solids per year, while Australia produced 0.3 million tones.

In 1984, 45% of the sludge produced in the UK was used in agricultural land, compared to West Germany 32% and in the USA 25%. Japan on the other hand incinerates 55% of the sludge produced (20). Land disposal of sludge is a simple physical operation with the main

Table 7.3
Trace elements in sludge

Metal	United States[a]		European Union[a]		Malaysia[b]	
	Range	Mean	Range	Mean	Range	Mean
Arsenic	1.1–230	10				
Cadmium	1–3,410	10	158–1,770		1.2–6.6	2.1
Chromium	10–99,000	500			7.2–1,326	15
Cobalt	11.3–2,490	30				
Copper	84–17,000	800	500–17,000		123–769	153
Iron	1,000–154,000	17,000			10,000–31,300	22,000
Lead	13–26,000	500	800–8,030		15.3–338	32
Manganese	32–9,870	260			297–460	367
Mercury	0.6–56	6			2.2–7.1	3.5
Molybdenum	0.1–214	4				
Nickel	2–5,300	80	100–1,000		14.1–162	19
Selenium	1.7–17.2	5				
Tin	2.6–329	14				
Zinc	101–49,000	1,700	1,000–15,000		669–7,110	1,090

Units: mg/kg dry sludge.
[a] Ref. (17).
[b] Ref. (19).

variations centering on the rates and techniques of application. Land spreading, soil injection, and landfill are the three main options, with environmental and safety considerations dictating application rates and the degree of pretreatment.

In general, land disposal is considered the ideal option for sewage sludge disposal for a number of reasons. If suitable land, which is located less than 20 km from the treatment plant, is available, then excessive processing of the sludge can be avoided and benefit gained from the nutrient content of the sludge as well as its soil conditioning properties. However, one of the major limiting factors on the application of sewage sludge to agricultural land is the presence of heavy metals. Even domestic sludge may contain high amounts of zinc, copper, lead, and cadmium. Table 7.3 gives comparative data from selected countries on the maximum allowable concentrations of heavy metals in sludge.

1.4.2. Land Application of Sludge in Malaysia

There are three types of sludge produced in Malaysia, namely septic tank sludge, drying bed sludge, and lagoon sludge. Different types of sludge will exhibit different physical, chemical, and biological properties and thus need to be classified prior to utilization.

Sewage sludge has many characteristics that are good for soils and plants, if applied properly. Research has shown that the organic matter in sludge can improve the physical properties of soil. Treated sludge is also known as biosolids, a slightly more attractive name used as a soil additive. Sludge improves the bulking density, aggregation, and porosity of the soil. In other words, if added properly, sludge enhances soil quality and makes it better for vegetation. Plants also benefit from the nitrogen, phosphorus, and potassium in sludge. When applied to soils at recommended volumes and rates, sludge can supply most of the nitrogen and phosphorus needed for good plant growth, as well as magnesium and many other essential trace elements like zinc, copper, and nickel within existing approval levels.

The application of treated sewage sludge to agricultural land is generally the most economical means of waste disposal and also provides an opportunity to recycle beneficial plant nutrients and organic matter to soil for crop production. However, sewage sludge also contains varying amounts of heavy metals that may pose hazard to metal toxicity in crops and to consumers of the crops. Thus, the uptake of heavy metals by crops and the fate of these heavy metals in soils need to be monitored.

1.4.3. Characteristic of Sludge

Sludge consists of organic solids, grit, and inorganic fines. Sewage sludge comprises lumpy, flaky, and colloidal solids interspersed with water. The volatile organic substance of the sewage sludge is either solid or liquid. If water is totally removed, the remaining organic volatile matter and inorganic matter (ash) are known as dry solids (DS). Tables 7.4 and 7.5 present the sludge characteristics for selected cities in Malaysia. Table 7.6 provides the sludge characteristics of three residential estates compared with an industrial estate.

1.4.3.1. Physical Properties

The application of treated sludge on soil has shown to alter the physical properties of the soil texture. Hydraulic properties like porosity, permeability, and flow velocity can influence

Table 7.4
Sludge characteristics in Malaysia for selected cities (19)

Selected city in Malaysia	Dry matter (% DS)	Organic matter (% DS)	pH 25°C	Total (N %)	Total (P %)	Zn (mg/kg DS)
Alor Setar	3.25	55.32	7.1	2.82	0.43	963.3
Gombak	4.27	62.36	7.1	2.87	0.28	912.2
Ipoh	12.12	67.35	7.4	3.04	0.57	1,178.9
Klang	2.20	62.57	7.1	2.47	0.46	1,123.4
Kluang	5.76	70.12	7.1	2.37	0.34	1,068.3
Kuala Terengganu	1.02	69.16	7.3	3.25	0.37	1,240.3
Kuala Lumpur	3.8	66.83	7.3	2.92	0.27	1,101.5
Kuantan	3.25	64.41	7.2	2.7	1.02	1,215.7
Labuan	3.43	63.31	7.2	2.64	0.45	2,156.8
Langkawi	0.71	66.51	7.3	2.92	0.9	1,250.4
Melaka	2.68	75.08	7.2	3.09	0.19	960.2
Penang	1.08	78.35	7.5	3.8	0.17	669.4
Seremban	3.04	67.14	7.3	2.75	0.32	1,096.5
Prai	2.29	73.17	7.2	3.08	0.36	1,167.7
Taiping	2.59	72.05	7.2	3.08	0.31	1,162.5
Ulu Tiram	4.77	57.68	7.2	2.3	0.25	928.4
Range	0.71–12.12	55.32–78.35	7.1–7.5	2.3–3.8	0.1–1.02	669.4–2,156.8

Table 7.5
Physical characteristics of sludge (21)

Parameter	Primary sludge	Secondary sludge	Dewatered sludge
Dry solids	2–6%	0.5–2%	15–35%
Volatile solids	60–80%	50–70%	30–60%
Sludge specific gravity	~1.02	~1.05	~1.1
Sludge solids specific gravity	~1.4	~1.25	~1.2−1.4
Shear strength (kN/m^2)	<5	<2	<20
Energy content (MJ/kg VS)	10–22	12–20	25–30
Particle size (90%)	<200 μm	<100 μm	<100 μm

Table 7.6
Sludge characteristics in Malaysia for selected cities (19)

Selected district in Malaysia	Cu (mg/kg DS)	Ni (mg/kg DS)	Cd (mg/kg DS)	Pb (mg/kg DS)	Hg (mg/kg DS)	Cr (mg/kg DS)
Alor Setar	135.7	21.1	1.8	35.0	4.7	23.3
Gombak	135.4	20.2	1.2	35.7	3.5	13.5
Ipoh	171.1	19.0	3.1	42.8	4.3	13.5
Klang	143.2	24.8	2.5	44.8	7.1	19.1
Kluang	147.1	15.6	2.0	33.0	2.2	14.4
Kuala Terengganu	153.1	17.2	2.5	23.2	2.2	9.3
Kuala Lumpur	153.9	18.6	2.4	29.5	4.2	16.5
Kuantan	151.9	30.5	2.4	42.0	7.1	9.9
Labuan	131.7	20.2	1.7	29.0	2.9	15.6
Langkawi	140.9	26.1	1.7	33.9	2.4	16.6
Melaka	155.3	27.2	2.2	20.2	4.6	17.0
Penang	131.6	15.8	2.1	19.8	3.3	7.2
Seremban	130.0	14.1	1.9	26.5	2.9	12.1
Prai	165.1	22.6	2.2	36.4	3.1	15.5
Taiping	154.4	17.5	2.2	25.5	3.9	12.5
Ulu Tiram	129.2	14.6	1.8	27.2	3.3	16.2
Range	129.2–165.1	14.1–30.5	1.2–3.1	19.8–44.8	2.2–7.1	7.2–23.3

soil moisture content and aeration respiration. The color varies from the source of sludge i.e., individual septic tanks (IST), Imhoff tanks, and mechanical process. Shear strength of sludge is a relevant parameter for consideration as sludge is being more and more disposed of on land. In the case of landfills (specifically mono-landfill), sludge should have a DS $> 35\%$ and a shear strength of more than $30\,kN/m^2$. However, there is some doubt as to the ability of sludge to retain shear strength as some research indicates that sludge loses its strength over time (i.e., more than 2 years) as shown in the Tables 7.4 and 7.5.

1.4.3.2. Chemical Properties

Chemical properties of sludge include metals, polymers, pH, alkalinity, and nutrients. The organic volatile matter may be characterized by its net calorific value. Chemical properties such as pH, alkalinity, and organic content of sewage sludge vary with industrial discharge into the system. The inorganic content of sewage sludge also varies widely, but for waste activated sludge, it is typically 20–35% DS, and for primary sludge, 30–45% DS.

1.4.4. Some Statistics on Sludge

Malaysia produces about 5 million cubic meters of sewage sludge per year, and this is expected to increase continuously. By the year 2022, the amount has been estimated to reach 7 million cubic meters per year (1). This is a tremendous amount of waste that has to be disposed off. World wide, the traditional means of sludge disposal is on land and into the sea. Due to increasing environmental awareness, disposal of sewage sludge will be costly, and an alternative disposal method that will enhance soil property and plant life without land contaminating them needs to be studied. Thus, pressures exist for useful or beneficial utilization of treating this waste. Research on utilization of treated domestic sewage sludge on crop lands has been in progress, and it is one aspect of sludge utilization that needs to be studied in detail in order to reduce rising costs. An efficient and economical disposal or safer application of this waste as a fertilizer is eminent.

Since 1994, individual septic tanks in Malaysia are desludged on a 2-year routine basis. Treated sludge from sewage treatment plants has been periodically taken to drying beds in regional plants. Table 7.7 shows the volume of sludge managed by Indah Water per month, while Table 7.8 gives the different sludge facilities adopted to treat the current sludge produced in Malaysia. The characteristic of sludge taken from several existing sewerage treatment plants in Negeri Sembilan and Klang Valley, Malaysia are provided in Table 7.9.

Table 7.7
Liquid sludge handled in Malaysia per month (19)

Sludge type	Quantity (m^3)
IST sludge	7,500
STP sludge	18,000
Pour flush sludge	4,500
Total	30,000

Table 7.8
Sludge treatment facilities used in Malaysia in treating sludge (19)

Treament/disposal Method	Quantity (%)	Volume (m^3)
Trenching	10	3,000
Drying beds	20	6,000
Sludge lagoons	10	3,000
STPs (with spare capacity)	60	18,000

Table 7.9
Characteristic of sludge taken from several existing sewerage treatment plants in Negeri Sembilan and Klang Valley (19)

Sludge Characteristics	Residential estates			Industrial estates
	Tmn Tasik Jaya, Seremban	Tmn Sri Gombak, Selangor	Tmn Sg. Besi Indah, KL	Tmn Perindustrian Puchong Utama
Total dry solids (TS, %)	46.2	36.7	54.5	94.3
Volatile solids (VS, %)	50.0	59.5	57.5	51.0
pH	6.87	5.16	6.00	6.06
Ash content (%)	50.0	40.5	42.5	49.0
Moisture content (%) (wet, wt)	53.8	63.3	45.5	5.7
Organic material (%)				
Carbon (C, %)	29.0	34.51	33.35	29.58
Nitrogen (N, %)	1.4	3.23	4.39	3.19
C/N Ratio	20.71	10.68	7.59	9.27
Phosphorus (P, %)	1.52	0.71	1.09	1.98
Inorganic material (% or mg/kg)				
Potassium (K, mg/kg)	696.73	539.50	521.10	861.61
Sodium (Na, mg/kg)	246.21	433.70	235.22	401.38
Calcium (Ca, %)	1.27	0.89	2.06	1.01
Iron (Fe, %)	2.8	2.01	1.02	3.13
Copper (Cu, mg/kg)	122.83	171.21	258.72	768.56
Zinc (Zn, mg/kg)	1,280.30	1,316.99	7,110.10	5,752.92
Lead (Pb, mg/kg)	73.60	93.55	223.40	338.28
Magnesium (Mg, mg/kg)	1,769.50	667.57	1,766.05	2,982.50
Silika (Si, mg/kg)	406.92	423.70	430.73	224.02
Chromium (Cr, mg/kg)	15.15	112.17	90.83	1,325.56
Cadmium (Cd, mg/kg)	3.35	3.77	6.28	6.57
Nickel (Ni, mg/kg)	28.14	25.88	43.12	162.25
Aluminum (Al, %)	0.91	1.8	1.18	1.67
Manganese (Mn, mg/kg)	389.07	296.55	322.48	460.23

2. PRINCIPLES OF PHYTOREMEDIATION

2.1. Types of Crops and the Uptake Relationship of Heavy Metal

The US EPA's Phytoremediation Resource Guide definition of the six types of phytoremediation and their application in listed below (22):

2.1.1. Phytoaccumulation

Also called phytoextraction, refers to the uptake and translocation of metal contaminants in the soil by plant roots into the aboveground portion of the plants. Certain plants, called hyperaccumulators, absorb unusually large amount of metals in comparison with other plants and the ambient metal concentration. These plants are selected and planted at a site based on the type of metal present and other site conditions. After the plants have been allowed to grow for several weeks to months, they are harvested. Landfilling, incineration and composting are options to dispose of or recycle the metals, although this depends upon the results of the Toxicity Characteristic Leaching Procedure (TCLP) and cost. The planting and harvesting of plants may be repeated as it is necessary to bring soil contaminant levels down to allowable limits. A plan may be required to deal with the plant waste. Testing of the plant tissue, leaves, roots etc., will determine if the plant tissue is a hazardous waste. Regulators will play a role in determining the testing method and requirement for the ultimate disposal of plant waste.

2.1.2. Phytodegradation

Also called phytotransformation, is the breakdown of contaminants taken up by plant through metabolic processes within the plant, or the breakdown of contaminants external to the plant through the effect of compound (such as enzymes) produced by the plants. Pollutants are degraded, used as nutrients, and incorporated into the plant tissues. In some cases, metabolic intermediate or end products are re-released to the environment depending on the contaminant or plant species (see Sect. 2.1.4).

2.1.3. Phytostabilization

Phytostabilization is the use of certain plant species to immobilize contaminants in the soil and groundwater through absorption and accumulation by roots, adsorption onto roots, or precipitation within the root zone, and physical stabilization of soils. This process reduces the mobility of the contaminant and prevents migration to the groundwater or air. This technique can be used to re-establish a vegetative cover at sites where natural vegetation is lacking due to high metal concentrations. Metal tolerant species may be used to restore vegetation to such sites, thereby decreasing the potential migration of contamination through wind erosion, transport to exposed surface soils, and leaching of soil contamination to groundwater.

2.1.4. Phytovolatilization

Phytovolatilization is the uptake and transpiration of the contaminant by a plant, with release from the plant. Phytovolatilization occurs as growing trees and other plants take up water and the organic and inorganic contaminants. Some of these contaminants can pass through the plants to the leaves and volatilize into the atmosphere at comparatively low concentrations. Many organic compounds transpired by a plant are subject to photodegradation.

2.1.5. Rhizodegradation

Rhizodegradation, also called phytostimulation, rhizosphere biodegradation, enhanced rhizosphere biodegradation, or plant-assisted bioremediation/degradation, is the breakdown of contaminants in the soil through microbial activity that is enhanced by the presence of the rhizosphere. Microorganisms (yeast, fungi, and/or bacteria) consume pollutants to degrade or transform organic substances such as nutrient substances. Certain microorganisms can degrade organic substances for use as nutrient substances such as fuels or solvents that are hazardous to human and eco-receptors and convert them into harmless products through biodegradation. Natural substances released by plant roots – such as sugars, alcohols, and acids – contain organic carbon that act as nutrient sources for soil microorganisms, and the additional nutrients stimulate their activity. Rhizodegradation is aided by the way plants loosen the soil and transport oxygen and water to the area. The plants also enhance biodegradation by other mechanisms such as breaking apart clods and transporting atmospheric oxygen to the root zone.

2.1.6. Rhizofiltration

Rhizofiltration is the absorption or precipitation of contaminants onto plant roots or the absorption of contaminants into the roots when contaminants are in solution surrounding the root zone. The plants are raised in greenhouses (with their roots in water rather than in soil). Once a large root system has been developed, contaminated water is diverted and brought in contact with the plants or the plants are moved and floated in the contaminated water. The plants are harvested and disposed as the roots become saturated with contaminants (22).

Plant species are selected for use according to their ability to treat the contaminants of concern and achieve the remedial objectives to redevelopment, and for their adaptability to other site-specific factors such as adaptation to local climates, depth of the plant's root structure, and the ability to the species to flourish in the type of the soil present. Often the preferred vegetation characteristics include the following:

(i) The ability to extract or degrade the contaminants of concern to nontoxic or less toxic products
(ii) Fast growth rate
(iii) Adaptability to local condition
(iv) Ease of planting and maintenance
(v) The uptake of large quantities of water by evapotranspiration.

Several types of plants and sample species frequently used for phytoremediation are listed below:

(i) Hybrid poplars, willow, cottonwood, and aspen trees
(ii) Grasses (rye, Bermuda grass, sorghum, and fescue)
(iii) Herbaceous plants such as legumes, clover, alfalfa, and cowpeas
(iv) Aquatic and wetland plants (water hyacinth, reed, bulrush, and parrot feather)
(v) Hyperaccumulators for metals (such as alpine pennycress for zinc or alyssum for nickel). Other plants that are being investigated for their potential to remediate heavy metals contaminated soil include Indian mustard (*Brassica juncea*), oats (*Avena sativa*), barley (*Hordeum vulgare*), and alfalfa (*Medicago sativa*).

2.1.7. Impact of Heavy Metals on Plants

Zinc (Zn) and Cadmium (Cd) concentration exceeded normal values reported for these two elements in the leaves of all crops studied except maize. Cd tends to accumulate in leafy vegetables (17) like lettuce and spinach as well as in potato leaves. Sugar beet has been reported to accumulate Zn (23). Result shows how much metal uptake from the same soil can vary between different crops and within different parts of the same plant. Because of the low availability of the metals in relation to the high total loads, no phytotoxicity was observed, but metal accumulation was still high enough to make crop products on the highly polluted plots unacceptable for consumption by humans or animals according to the current legal standards in Switzerland.

The fate and effects of sewage sludge constituents in a soil–plant system are influenced by factors such as climate (rainfall and temperature), management (irrigation, drainage, liming, fertilization, addition of amendments), and composition of the sewage sludge. In addition, soil properties affect the chemical reaction and process, which occur after the application of sewage sludge to a soil. Soil properties that affect the reaction and resultant plant uptake of sewage sludge constituent include pH, organic matter, cation exchange capacity (CEC), iron and aluminum oxides, texture, aeration, specific sorption sites, and water availability. Many of these factors are interrelated and thus create a rather complex medium involving chemical and microbial reactions. The factors, such as pH, water content, and aeration (relates to water content), vary frequently or are easier to adjust. For example, soil pH can be increased by lime additions.

Soils cation exchange capacity (CEC) is dependent on soil properties such as organic matter, pH type, and percentage of clay. Thus, it serves as an easily measured; integrating parameasured soil property, which provides background information on soil, pH measured in the laboratory is the representation of that site in the soil may be significantly different from the pH of other sites. For example, the pH at the root–soil interface may be lower because of exuded organic acids. Due to differential uptake of cations and anions, the pH in the root cylinder of active root hairs may be lower than that in order parts of the root system. Also, pH reductions with time in sludge-treated soils are due to the protons generated during the oxidation of reduced forms of N and S mineralized from sludge organic matter. Similar pH reductions occur after the addition of fertilizers, particularly those containing ammonium.

Plant uptake of elements from soil solution initially requires positional availability to the plant root. Either the element must be moved to the root through diffusion or mass flow processes, or the root must grow to the element. The element must then occur in a form, which can move into the plant via the uptake mechanism. This transfer requires that the element move through a solution phase, thus water solubility and a variety of complexation, chelation, and other chemical reaction become important.

In general, researchers agree that effects of organic compounds, certain pesticides, and metals are not dangerous when managed properly at regulated levels. However, they caution that additional study of organic compounds and longterm fate of materials is needed before unlimited application of sludge can occur safely on all lands.

2.2. Design Parameters

The design consideration includes

(a) Contaminant levels
(b) Plant selection
(c) Treatability
(d) Irrigation, agronomic inputs (P, N, P, salinity, Zinc, etc.), and maintenance
(e) Groundwater capture zone and transpiration rate
(f) Contaminant uptake rate and clean-up time required.

The design of a phytoremediation system varies according to the contaminants, the conditions at the site, the level of clean up required, and the plants used. Contaminant and site conditions are perhaps the most important factors in the design and success of a phytoremediation system. Other factors that influence the selection and design of a phytoremediation system are as below:

(a) Technical factors
(b) Strategies for contaminant control
(c) Innovative technology treatment trains
(d) Design team (soil science or agronomy, hydrology, plant biology, environmental engineering, regulatory analysis, cost engineering and evaluation, risk assessment and toxicology, and landscape architecture) (4).

Design of a phytoremediation system includes

(a) Plant selection
(b) Treatability
(c) Planting density and pattern
(d) Irrigation, agronomic inputs, and maintenance
(e) Groundwater capture zone and transpiration rate
(f) Contaminant uptake and clean up time required
(g) Analysis of failure modes.

2.2.1. Monitoring Plan

Usually, a monitoring plan is also submitted to the authorities before the approval of a project is given. The success of a phytoremediation project would depend on the monitoring of the reduction of contaminant levels in the soil or the accumulation of contaminant concentrations in the plants. Pilot studies should be performed before field-scale phytoremediation project are implemented. Information collected during monitoring may indicate the need for design modification. The monitoring plan should include evapotranspiration, erosion control, contaminant reduction in soil or contaminant accumulation in plants, and the process of succession. Evapotranspiration is measured by assessing the hydrologic balance of the site, while erosion control is usually measured by the quality of water runoff. The effectiveness of the contaminant reduction may be estimated through soil nutrients data, soil oxygen content, root development, and the measured levels of contaminants. The change of site vegetation over time must also be considered in the monitoring plan. The ideal species for remediating a site may be replaced by naturally occurring plants or pioneer species. The ideal species should be introduced to the site prior to pioneer species invasion.

2.2.2. Limitations

Phytoremediation has its own limitations. The presence of the contaminants in plants may be bioavailable to the food chain at an unacceptable concentration. The potential absorbed dosages must be estimated and compared with maximum safe food intake limits. Authorities such as the Department of Environment or Local Government may impose certain restrictions, for example, by erecting fencing around contaminated sites and/or by having buffer zones around such sites.

Table 7.10
Pollutant limit for land application (24)

Pollutant	Ceiling concentration limits for all biosolids applied to land (mg/kg) (Dry wt.)	Cumulative pollutant loading rate limits for CPLR biosolids (kg/ha)	Annual pollutant loading rate limits for APLR biosolids (kg/ha) APLR 365-day period
Arsenic (As)	75	41	2.0
Cadmium (Cd)	85	39	1.9
Chromium (Cr)	3,000	3,000	150
Copper (Cu)	4,300	1,500	75
Lead (Pb)	840	300	15
Mercury (Hg)	57	17	0.85
Nickel (Ni)	420	420	21
Selenium (Se)	100	100	5.0
Zinc (Zn)	7,500	2,800	140
From Part 503	Table 1, Section 503.13	Table 2, Section 503.13	Table 4, Section 503.13

2.3. *Empirical Equations*

Determination of the annual whole sludge application rate is given by the following formulae (22):

$$\text{AWSAR} = \frac{\text{APLR}}{0.001\,C}, \tag{1}$$

where, AWSAR is the annual whole sludge application rate (dry metric tons of biosolids/ha/year), APLR is the annual pollutant loading rate (kg of pollutant/ha/year) from Table 7.10, C is the Pollutant concentration (mg of pollutant/kg of biosolids, dry weight), and 0.001 is the conversion factor.

2.4. *Health Effects*

Heavy metals, including lead, are present in soils either as natural components or as the result of human activity. Metal-rich mine tailings, metal smelting, electroplating, gas exhausts, energy and fuel production, downwash from power lines, intensive agriculture, and sludge dumping are the human activities that introduce the largest quantities of lead into soils.

Today, more is known about the effects of lead and the pathways of exposure. Currently, lead is listed as a known or suspected carcinogen in the EPA's Toxic Release Inventory (TRI). If ingested, lead can accumulate in body organs, including the brain, and result in various degrees of lead poisoning. At high levels of exposure, lead can not only damage the brain and kidneys of adults and children severely, but also cause death.

Major pathways of exposure to lead:

(a) The inhalation of lead-containing car exhausts or industrial emission
(b) The ingestion of lead-based pain

(c) The ingestion of contaminated soil or dust from hand-to-mouth activities of those living in lead polluted environment
(d) The inhalation of leaded dust carried on clothing or by the wind.

Children face the most devastating effects of lead poisoning. The effects of lead are listed as below:
Effects for fetuses,

(a) Premature births
(b) Smaller birth weight
(c) Decreased mental ability in the infant
(d) Abortion.

Effects for children,

(a) Impair development
(b) Result in a lower IQ
(c) Shortened attention span
(d) Cause hyperactivity
(e) Cause progressive mental deterioration (includes a loss of motor skill, severe aggressive behavior disorders, and poorly controlled convulsive disorder).

Effects for adults,

(a) Decrease reaction time
(b) Possibly affect the memory
(c) Cause weakness in fingers, wrists, and/or ankles
(d) Cause anemia, weakness, lassitude, insomnia, facial pallor
(e) Weight loss, anorexia, malnutrition, constipation, nausea, abdominal pain, and vomiting
(f) May increase blood pressure in middle-aged man
(g) High levels of exposure may damage the male reproductive systems.

Future research direction,

(a) Determination of uptake rate of contaminant among different species of plants
(b) Determination of heavy metals uptake by harvestable crops such as palm oil.

3. STANDARDS AND REGULATIONS

3.1. Sludge Application on Land

Sewage sludge has many characteristics that are good for soils and plants, if applied properly. Research has shown that the organic matter in sludge can improve the physical properties of soil. Treated sludge, also known as biosolids, is a slightly more attractive name used as a soil additive. Sludge improves the bulking density, aggregation, and porosity of the soil. In other words, if added properly, sludge enhances soil quality and makes it better for vegetation. Plants also benefit from the nitrogen, phosphorus, and potassium in sludge. When applied to soils at recommended volumes and rates, sludge can supply most of the nitrogen and phosphorus needed for good plant growth, as well as magnesium and many other essential trace elements such as zinc, copper, and nickel within existing approval levels.

Fifteen chemical elements are considered essential for plant growth, that is, plants will not complete their lifecycle if they are not supplied. Each of these essential elements has defined physiological functions (25). Macronutrients are needed in concentrations greater than 0.15% of dry matter or are present at greater than 5 kg/ha in the mature plant tops. Micronutrients are those in concentration less than 0.01% of dry matter or are present at less than about 1 kg/ha in the mature plant tops.

Some plant species may require other elements for growth. Sodium may be necessary for plants with the C_4 photosynthetic pathway, and some halophytes, such as saltbush (*Atriplex* spp.). Silicon is important for the strength of stem tissue of some crops, such as rice and sugar cane. Legumes fixing N symbiotically require cobalt. This is a requirement of the *Rhizobium* spp., but has been observed also in growth responses by subterranean clover (Trifolium subterranean) to cobalt sulfate applications.

The application of treated sewage sludge to agricultural land is generally the most economical means of waste disposal and also provides an opportunity to recycle beneficial plant nutrients and organic matter to soil for crop production. However, sewage sludge also contains varying amounts of heavy metals that may pose hazard to metal toxicity in crops and to consumers of the crops. Thus, the uptake of heavy metals by crops and the fate of these heavy metals in soils need to be monitored. The following sections highlight the probable contamination levels of untreated domestic sludge on land and the advantages of nourishing the soil by safe and controlled application of treated domestic sludge or biosolids. Besides, there is a need for regulatory body to formulate the rate and frequency of application of biosolids to different types of soil and recommended characteristics of treated biosolids.

3.2. Standards and Regulations of Sludge Applications in Malaysia, the USA, and Europe

For advanced countries such as Europe and the United State, there are regulations on the criteria of waste disposal to be protected of an environment quality, including heavy metals content in those wastes. The USEPA and European Community Limit have regulated a guideline on heavy metals content for biosolids disposal as shown in the Tables 7.10 and 7.11. There are some differences on concentration of heavy metal allowable limit between them as an example; allowable concentration limit of cadmium is 85 mg/kg (dry wt.) for USEPA instead allowable concentration limit of cadmium range of 20–40 mg/kg for European Community Limit.

In Malaysia, the Government has regulated the *Environmental Quality* (*Sewage and Industrial Effluents*) (*Amendment*) *Regulations 2000* [*P.U (A) 398/00*], but has not mentioned about the allowable concentration limit of heavy metals in the sludge before it can be disposed off into the landfarming. Malaysian's domestic sludge is processed separately from industrial and commercial sludge; thus the heavy metals content are very low, as shown in the result of this study. Anyway, sludge generation would increase tremendously, so the government should revise and update an existing regulation. Hopefully, this study would help the government agencies to revise a regulation, especially on heavy metal allowable limit.

Table 7.11
European community limit (after CEC 1986) [a] (26)

Pollutant	Concentration in soil (mg/kg)	Concentration in dry sewage sludge (mg/kg)	Annual application rate (kg/ha/year)[b]
Cadmium	1–3	20–40	0.15
Copper	50–140	1,000–1,750	12
Nickel	30–75	300–400	3
Lead	50–300	750–1,200	15
Zinc	150–300	2,500–4,000	30
Mercury	1–1.5	16–25	0.1

[a] Assume soil pH range of 6–7.
[b] Based on average 10 years.

4. CASE STUDIES AND RESEARCH FINDINGS

Researches showed that majority of crops were able to adsorb almost heavy metals and concentrated in the tissues with or without effect to the crop's yield depending on the types and concentration of heavy metals applied. One of the factors that influences an uptake of heavy metal by the crops is soil pH. Normally, maximum yield of crops are achieved in the soil pH range of 5.5–6.5 and decrease below or above the range. Based on the study, the soil pH falls slightly below this range (pH = 5.2). However, there are exceptions in the case of lupines and treacle performing well in more acidic soils, whereas medics such as Lucerne prefer alkaline soils. The problem of low soil pH occurs in regions of excess rainfall of 500 mm per annum and irrigated areas. The problems of high pH are common in lower rainfall environments with calcareous sands and cracking clays as well as with many nonsaline sodic soils. Soil composition varies widely, and it reflects the nature of the parent material. The principle factors determining these variations are the selective incorporation of particular elements in specific minerals during igneous rock crystallization, the relative rates of weathering, and the modes of formation of sedimentary rocks.

Studied showed that most of pH values of all treatment ponds was in the normal range (pH $\approx$ 7.0), while COD, TS, and TVS parameters vary from each other. Domestic sludge sample from Community septic tank treatment plant was the highest concentration to COD, TS, and TVS, which were 79,900, 16.0, and 12.54 mg/L, respectively, while Activated sludge was the lowest concentration to TS and TVS, which were 1.34 and 0.28 mg/L as shown in the Table 7.12.

Studies on heavy metals content in the domestic sludge showed that cadmium range from 0.001–0.100 mg/kg (dry weight), chromium from 0.091–0.285 mg/kg (dry weight), copper from 0.131–0.569 mg/kg (dry weight), lead from 0.212–0.555 mg/kg (dry weight), nickel from 0.300–2.324 mg/kg (dry weight), and zinc from 0.180–3.129 mg/kg (dry weight). The concentration of those heavy metals after application to soil was 1.1211, 54.450, 57.113, 397.62, 844.42, and 183.38 mg/kg (dry weight) for cadmium, chromium, copper, lead, nickel, and zinc as shown in Table 7.13. Metal concentrations of sludge are presented in Table 7.14. Based on the U.S. Environmental Protection Agency (22) Part 503 and European Community

Table 7.12
Characterization of domestic sludge in Malaysia

Parameter	Community septic tank (CST)	Activated sludge (AcS)	Oxidation pond (OP)	Aerated lagoon (AL)
pH	7.22	7.16	6.92	7.03
COD, mg/L	79,900	29,600	31,500	26,400
Total solids, mg/L	16	1.34	3.99	13.61
Total volatile solids, mg/L	12.54	0.28	1.25	10.05

Table 7.13
Heavy metals content in the domestic sludge sample

Subject	Concentration of elements, mg/kg (dry wt.)					
	Cadmium	Chromium	Copper	Lead	Nickel	Zinc
Heavy metal in studied domestic sludge (range)	0.001–0.100	0.091–0.285	0.131–0.569	0.212–0.555	0.300–2.324	0.180–3.129
Heavy metal in soil after applied sludge	1.1211	54.450	57.113	397.62	844.42	183.38

Table 7.14
Comparison of heavy metals content to USEPA and European community limit

Element	Concentration of elements, mg/kg (dry wt.)					
	Cd	Cr	Cu	Pb	Ni	Zn
This study (average)	0.003	0.203	1.202	0.37	1.077	1.46
USEPA, Part 503	85	3,000	4,300	840	420	7,500
European Community Limit	20–40	N.S	1,000–1,750	750–1,200	300–400	2,500–4,000

Limit, the content of heavy metals substance in domestic sludge studies remains well below the limit values.

The heavy metal concentration range was different in the plants after being applied by domestic sludge as shown in the Table 7.15. Three types of plants were chosen to be studied of heavy metal uptake by crops; *Ipomoea aquatica*, *Spinacea oleracea* and *Brassica juncea*. *Spinacea oleracea* has shown a good uptake of metal cadmium and zinc, while *ipomoea aquatica* and *Brassica juncea* have shown a good uptake of metal chromium, copper, lead, and nickel. It also showed a good sign of heavy metal mobility in plant–soil system.

Table 7.16 shows a distribution of heavy metals content in plants cross-section (%). The distribution of heavy metal in different parts of the crops is variable depending on the type of heavy metal. Most metals are more concentrated in root tissues of plants than in stem and leaves tissues, especially for lead, nickel, and copper.

Table 7.15
Heavy metals content in the plants sample

Type of plants	Average concentration of heavy metals content, mg/kg (dry wt.)					
	Cadmium	Chromium	Copper	Lead	Nickel	Zinc
Ipomoea aquatica	0.251	10.83	37.68	32.96	213.2	63.47
Spinacea oleracea	1.26	8.4	17.45	23.05	24.06	118.25
Brassica juncea	0.15	9.27	22.42	26.25	32.24	88.83

Table 7.16
Distribution of heavy metals content in plants cross-section (%)

Cross-section	Type of plant	Cd	Cr	Cu	Pb	Ni	Zn
Leaves	*Ipomoea aquatica*	29.055	38.085	27.644	12.862	31.646	30.586
	Spinacia oleracea	26.928	12.545	24.563	12.508	11.681	18.908
	Brassica juncea	39.092	16.15	13.315	12.803	19.367	26.075
Stems	*Ipomoea aquatica*	40.685	23.363	21.527	13.342	26.806	36.411
	Spinacia oleracea	13.459	32.173	18.76	9.1241	15.702	28.595
	Brassica juncea	22.504	29.608	44.155	23.595	30.74	35.008
Roots	*Ipomoea aquatica*	30.818	38.573	50.959	73.666	41.562	32.927
	Spinacia oleracea	59.652	55.275	56.706	78.378	72.625	52.531
	Brassica juncea	38.403	54.242	42.53	63.602	49.893	38.917

Table 7.17
Design example for sample from Indah Water Konsortium (IWK), Malaysia

Heavy metal	Biosolids concentrations (mg/kg)	APLR (kg/ha/year)	$\text{AWSAR} = \frac{\text{APLR}}{(0.001)\ \text{Conc. In biosolids}}$ (tons/ha)
Cadmium, Cd	2.0	1.9	$1.9/(0.001 \times 2.0) = 950.0$
Chromium, Cr	14.4	150	$150/(0.001 \times 14.4) = 10{,}416.7$
Copper, Cu	147.1	75	$75/(0.001 \times 147.1) = 509.9$
Lead, Pb	33.0	15	$15/(0.001 \times 33.0) = 454.5$
Nickel, Ni	15.6	21	$21/(0.001 \times 15.6) = 1{,}346.2$

5. DESIGN EXAMPLE

By using data from Indah Water Konsortium (IWK), Malaysia for Kluang location as shown in Table 7.6, the determination of the annual whole sludge application rate could be calculated as shown in Table 7.17.

6. FUTURE DIRECTION RESEARCH

Studies show that landfarming method is capable of reducing the concentration of heavy metals in the samples. From the result of this study, landfarming technique is suitably applied to the palm oil farm because the concentration of heavy metals could be reached into an eatable tissue lesser. Anyway, further study should be done to make sure the concentration of heavy metals in an eatable tissue. For future research, the determination of heavy metals uptake rate for several of plants could be done. Further study would be able to gain the range of heavy metal constant uptake rate by the crops (27, 28).

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