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Mechanical Methods of Soil Cleaning

Abstract

Soil cleaning technologies are becoming more and more important especially in industrialized countries. Application of soil washing i.e. physical separation results in cleaned soil which can be reused at the origin or the other location. Separated impurities (metals, hydrocarbons) are considered as special waste and further processed and/or disposed on special landfills. Technologies that were originally developed for mineral processing, due to similarity of operative goals – concentration/separation of certain matter adhering to a certain class of grain collective – have a high potential for use in soil cleaning. The purpose of this paper is to give a review of mineral processing methods which can be applied in soil washing technology, and to present some of the processes successfully applied in practice.

Key words: soil cleaning, soil washing, mechanical methods

1. Introduction

Soil cleaning (soil remediation) has been gaining more and more importance over the last decade, especially in industrial countries with considerable soil pollution due to intensive industrial activities. The sources of soil pollution are chemical, petrochemical, pharmaceutical and graphic industry, agriculture, metallurgy, mining, fossil power plants, transport, landfills, waste recycling plants, etc. Croatia, as a country with low industrial activity, has a relatively clean soil. However, due to lack of soil protection strategy, the soil as a resource was neglected until recent time and its quality is yet to be determined. According to the available data, 1,056 potentially polluted sites were recorded in Croatia, 69 of which were confirmed as polluted sites, but the number of potentially polluted sites is very likely higher (AZO, 2007)¹. Also, 11 so called “black spots” were registered in 2005 (AZO, 2007)². Two of them were remediated (power plant Plomin and PVC factory “Ina Vinil”) and one is still in the process of remediation (coke plant in Bakar). Other sites with critical soil pollution due to industrial activities and improper disposal of hazardous technological waste are: asbestos-cement waste dumps of the asbestos products factory “Salonit” (Vranjic and Mravinačka kava), ex alumina factory in Obrovac, ex electrodes and ferro-alloy factory in Šibenik, Croatian Railroads station for washing and disinfection in Botovo, hazardous technical waste dump “Lemić brdo” near Karlovac, tar and waste oil dump in Jama Sovjak, phosphorous-gypsum waste dump of “Petrokemija” in Kutina (Figure 1).

Not so long ago, depending on the area of contaminated site and level of contamination, the main methods of dealing with contaminated soil problems were: the isolation of contaminated area with restricted admittance, covering the contaminated area with clean soil and vegetation, or excavation and controlled disposal of contaminated soil.

Today, there is a wide range of soil cleaning methods. Soil washing is a method of soil cleaning in which the contaminant is removed from the soil, and after that cleaned soil can be used again at the same or different location with no threat to humans or environment. Removed contaminants are further treated and/or disposed as a special waste.

2. Soil Contaminants

From the viewpoint of pedology, soil is an autonomous natural and historical creation as well as a capital good of agriculture and forestry (Martinović, 1997). Key factors of soil genesis and development are soil matrix, living organisms, climate, geomorphology, time and human activities. One of the essential properties of soil is its fertility which is closely linked to the development of vegetation and living organisms in soil. Soil is a system consisting of four phases – solid particles, solutions, air and living organisms, structured by different levels of hierarchy: atoms, molecules, particles, horizons, profiles, pedosystems, soil associations and pedosphere.

Contaminated soil is a soil that contains pollutant above a certain level causing a deterioration or loss of one or more soil functions (EC-JRC, 2010). The functions of soil are: production (food, biomass), biotopic function (biological habitat), environmental interaction (transformation, regulation, filtration, storage), source of raw materials, building platform (Martinović, 1997). Anthropogenic contamination typically arises from



Figure 1. Black spots (AZO, 2007)

the underground tanks leakages, extensive use of pesticides and fertilizers, infiltration of contaminated surface water to the subsurface, landfill and waste dumps leachate infiltration, industrial wastewater discharge, spilling and industrial waste disposal directly on the ground, etc. Soil contaminants can further contaminate a groundwater and air (EC-JRC, 2010; EPA, 2010).

Soil contaminants are usually divided into organics and inorganics. Further, organic contaminants are classified by volatility and presence of halogens in their chemical structure (Reis et al, 2008; FRTR, 2010):

1. Organics

- Nonhalogenated Volatile Organic Compounds (VOCs)
 - a. Light Hydrocarbons
 - b. Benzene, Toluene, Ethyl benzene, Xylene (BTEX)
 - c. Oxygenated Hydrocarbons
 - d. Other compounds
- Halogenated Volatile Organic Compounds (X-VOCs)
 - a. Chlorinated Hydrocarbons
 - b. Other X-VOCs
- Nonhalogenated Semi-Volatile Organic Compounds (SVOCs)
 - a. Heavy Hydrocarbons
 - b. Nonhalogenated Pesticides
 - c. Polycyclic Aromatic Hydrocarbons (PAHs)
 - d. Nitrogenated PAHs and Amines
 - e. Nonhalogenated Phenols
- Halogenated Semi-Volatile Organic Compounds (X-SVOCs)
 - a. Polychlorinated Byphenyls (PCBs)
 - b. Halogenated Pesticides
 - c. Other X-SVOCs
- Dioxins i furans

2. Inorganics

- Heavy metals
 - a. Volatile heavy metals and compounds
 - b. Nonvolatile heavy metals and compounds
- Radionuclides
- Other inorganic elements and compounds (azbestos, fluorine, cyanide, etc.)

3. Soil cleaning methods

Soil cleaning can be applied at contaminated sites using mobile plants or in stationary plants after transportation of excavated contaminated soil. Soil cleaning technologies are classified by main mechanism of cleaning into: physical, chemical, biological and thermal treatments (Martin et al, 2004). Since an operational goal of certain soil cleaning method can be separation of contaminant, detoxication of soil by contaminant destruction or contaminant immobilization, treatments of contaminated soil are classified into three groups: separation (physical and chemical methods), detoxification/ destruction (chemical, biological and thermal methods) and immobilization (chemical and thermal methods).

Physico-chemical methods based on physical and/or chemical properties of contaminant or contaminated medium are used for the separation or concentration of contaminant, or for its destruction i.e. the change of contaminant chemical properties. Chemical treatments can be divided into two groups: the group of separation methods used for improvement of physical separation (conditioning) and the group of destruction/detoxification methods. Chemical destruction/detoxification methods are based on destruction of contaminant by changing its molecular and structural properties when reacting with certain reagents, resulting in less toxic or non toxic compounds. Physico-chemical treatments are low in cost and energy consumption, treatment duration is relatively short, equipment is widely available and less demanding in terms of qualifications and number of personnel. Application of some treatments directly in the ground can meet the difficulties due to variations of soil parameters (e.g. clay and humus can cause the variations of soil horizontal and vertical hydraulic properties resulting in unevenly cleaned soil).

Biological methods, based on microbial activities, are used for degradation of organic contaminants products like CO_2 , H_2O , fatty acids and biomass. Application is possible both on site and ex site in special reactor tanks. Usually, treatment duration is long but after contaminant destruction soil remains bio-active. Bio-treatments are applicable to soil, sediments, sludge and water. The costs of implementation are low. Contaminant destruction is almost complete and additional treatments are rarely required. Difficulty with these treatments is to determine when complete contaminant is destroyed, and additional problem can be the sensitiveness of microorganisms to toxins and high concentrations of contaminants.

Thermal methods are classified in the group of detoxification/destruction methods, but some of them are used for separation too. Using the high temperature in controlled environment, physical, chemical or biological properties and composition of contaminated material (soil, waste) are changed, i.e. contaminant is volatilized, incinerated, degraded, destroyed or melted. Thermal treatments are used for cleaning of soil contaminated with organics like oil and oil derivatives (soil near underground tanks, spilling). Destruction of contaminant is fast (depending on oven capacity and volume of contaminated soil), but energy consumption and equipment costs are high, as well as equipment maintaining and personnel requirements. In incineration devices, e.g. rotating ovens, at the temperature of 500-600 °C, organics are volatilized and oxidized, producing carbon-dioxide, water and toxic compounds like SO₂, NO_x, HCl, products of incomplete incineration and ashes. After thermal treatment, the soil is biologically dead. Heating the soil above 1600°C (vitrification) produces inert vitrified mass, i.e. slag. Because of high energy consumption, these treatments are used only if other treatments are not applicable or effective.

Immobilization (stabilization/solidification) is a group of treatments for immobilization of toxic compounds by their binding to materials like soil, sand or building materials. Toxic compounds are physically bonded or encapsulated in solid mass which has a high compressive strength and low permeability (cement or silicate based solidification, thermoplastic solidification, polymer based encapsulation, vitrification) or they are immobilized by chemical reactions with stabilizers (stabilization). Immobilization methods are applied mainly for inorganic contaminations, but they can be applied efficiently to volatile organics and pesticides.

Soil washing is a technological process of separation of highly contaminated soil particles from the remaining soil, based on difference in physical and physical-chemical parameters of soil and contaminant. The separation is conducted in water as a medium. After washing, clean soil is safe for environment and ready to transport to the original or other location. Soil washing is applicable to soil and sludge contaminated with heavy metals, oil derivatives, PAHs, cyanides, phenols etc. Soil washing refers to the group of methods based on physical separation and/or chemical extraction. Physical separation is used for the concentration of contaminants into a smaller volume of soil, based on difference in physical properties (particle size, density, magnetic, electrostatic and surface properties) between contaminated soil particles (contaminant carrier) and soil particles. The aim of chemical extraction process is to dissolve the contaminant using the water solution of reagents. If the soil contamination

is in the form of particles, physical separation is applied, and if it is in adsorbed forms, then chemical extraction is applied. Definitions and use of expressions “soil washing”, “physical separation” and “chemical extraction” can vary from author to author, however, the prevailing technique in the process of cleaning has the main influence on choice of expression. In the USA and Europe, the expression “soil washing” usually refers to the soil contaminant removal by technologies commonly used in mineral processing (Dermont et al, 2008). The efficiency of separation methods in soil washing depends on soil characteristics: particle size distribution, particle shape of soil matrix and contaminant, clay content, humus content, soil moisture, heterogeneity of soil matrix, difference in density between soil matrix and contaminant, magnetic and electrostatic properties of soil and contaminant, surface properties of particles (wetting), etc. Soil washing for heavy metal removal can be difficult, especially in the following cases:

- Metal is firmly bonded to soil particle surface;
- Soil matrix and metal carrier particles have similar densities;
- Metal is present in wide variety of chemical forms;
- Metal carrier particles are present in wide size range;
- Contaminated soil contains more than 30-50% of clay, large amount of humus and/or very viscous organics.

Separation methods are applied primarily on urban and industrial soils contaminated by human activities. They are not so efficient for cleaning of agricultural soil because it contains relatively low concentrations of adsorbed metals, high amounts of clays, fines and organic matter, although some treatments like attrition scrubbing can be used for the improvement of chemical extraction of adsorbed metals. Some recent experiments have shown high efficiency of separation methods for the removal of hydrocarbons too. Only by water washing, 24% of *n*-alkanes is removed from the soil of particles size > 0.8 mm, contaminated with diesel fuel at the concentration of 7% by weight, and the use of surfactants in washing process results in 97% removal of the contaminant (Khalladi et al, 2009). Attrition scrubbing experiments conducted on sand samples of particle size 0.1-0.5 mm, contaminated with diesel fuel 5-15% by weight, resulted in 96% contaminant removal (Bayley & Biggs, 2005). Flotation of sand (particle size 0.4-1.2 mm) also resulted in high percentage removal of diesel fuel ($> 90\%$) (Couto, 2009). Mineral processing technologies can be efficiently applied not only for the removal of metal and organic contaminant, but also for radionuclide removal (Gavrilescu et al, 2009).

4. Technological process of soil washing

In mixture of water and contaminated soil, mechanical energy is introduced via certain machine or device and, if necessary, a reagent is added and/or mixture is heated. In that process, soil matrix is disintegrated, individual soil particles are liberated and contamination dissolved and redistributed. After classification, separation and dewatering, most of the processed material is “washed soil” that can be returned to the original location. Small amount of separated material contains most of the contaminants in the form of sludge, light products (wood, coal, coke, porous building material, etc.), flotation froth or water solution. Separated contaminated products are treated as special waste or further processed and disposed.

4.1. Laboratory characterization of soil samples

Material that enters the soil washing cycle is not just a soil in the pedological sense, it is a complex mixture of soil forming minerals, organic matter and impurities such as small pieces of brick, concrete, wood, slag, etc. Because of the large number of possible impurities in the mixture, their different physical and chemical properties, types of intergrowth between impurities and soil components, and differences in soil texture, it is not possible to define universal soil washing process. Each case must be thoroughly investigated and analyzed. Depending on determined specific values and technological indicators, decision about the most suitable process is made. Usual analyses of contaminated soil samples include (Salopek et al, 2004):

- determination of density, particle size distribution, mineral and chemical composition,
- determination of contamination content in each particle size class, mass content of light and heavy components (light – less than 1800 kg/m³, heavy – above 2800 kg/m³),
- determination of the type of intergrowth between contaminant and soil components,
- determination of chemical composition and concentration of contaminant, the possibilities of attrition, deagglomeration, dispersion, emulgation and solubility,
- determination of classification and/or separation cut (Tromp curve, HR-diagram), recovery and contaminant concentration in washed products.

4.2. Soil preparation

Contaminated soil can contain pebbles, rock, slag, concrete or brick pieces which, due to their size, are not appropriate for washing. Usually, grains larger than 30 (50) mm are separated on vibrating screens. If contaminant concentration in separated material is below a certain required value, then the material is treated as clean. If not, then separated material is crushed, e.g. in impact crusher, and returned to the washing process with the rest of the soil.

4.3. Liberation

The purpose of grain liberation is to physically separate the mineral grain from contaminant. It is a process of breaking the bonds between soil matrix constituents and contaminant in the soil matrix. Only successful liberation of contaminant particles guarantees successful separation. Choice of liberation process depends on the type of intergrowth between mineral particle and contaminant. The three main type of intergrowth are presented in Figure 2. Type 1 is easily achieved by mixing

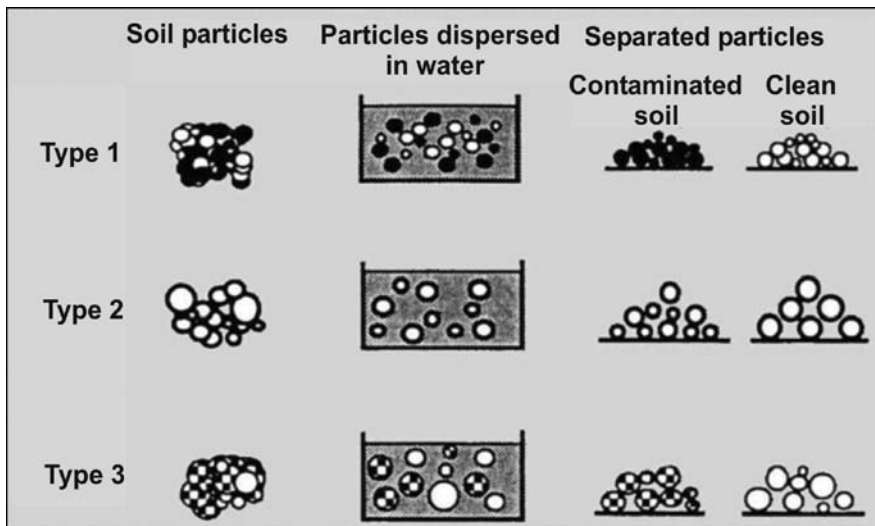


Figure 2. Main types of intergrowth between mineral particle and contaminant: type 1 – contamination in the form of free grains (coal, coke, brick or concrete particles mixed with soil particles), type 2 – contamination in the form of film on soil particle surface (particles enveloped with oil or oil derivatives film), type 3 – contamination dispersed inside the mineral particle (penetration of contaminant inside the grain).

with water in washing drum. Type 2 is more complex and requires larger input of mechanical energy. For film stripping attrition scrubber is used. If deagglomeration is required, high-pressure jet washing is applied. Attrition scrubbing products are cleaned particles, oil-water emulsion and sludge.

Type 3 of intergrowth is the most complex and liberation can rarely be achieved only by mechanical methods. Chemical treatment such as leaching, possibly after grinding, is usually required to convert the type 3 into type 2 or 1.

4.4. Classification

The purpose of classification is to separate the groups of particles of different size into classes. Separated particle classes can be the final product or input material for the next step of processing (separation). For example, after the first step of washing in washing drum, material is sieved on vibrating screen where the oversize (grains larger than 2 (4) mm) is a final product. Undersize (grains smaller than 2 (4) mm) is washed in the second step (attrition scrubbing) or further classified (in a hydrocyclone) and prepared for separation treatment (gravity concentration, flotation). Different types of vibrating screens are usually used for the classification of coarse material, and hydrocyclones or sometimes other types classifiers for the classification of fine material.

4.5. Separation (concentration, sorting)

After achieved liberation and separation of appropriate grain size class, the material is further processed by a certain separation method. The choice of separation method depends on physical and chemical properties of soil particles and soil contaminant. The properties are: density, magnetism, electrical conductivity and/or wetting.

In the case of type 1 intergrowth, with no firm physical bond between contaminated soil particles, separation is easily achieved. After grain size analysis of contaminated soil and determination of classes with the highest contaminant concentration, impurities could be removed by screening, providing they are present in the soil in the form of coarse particles. If the contaminant is concentrated in finer grain size classes, it can be removed by classifying in a hydrocyclone or hydraulic classifier. Therefore, in simple cases of contamination, classifying equipment could be used for

contaminant separation. If there is a significant difference in density of soil particles and contaminant particles (wood, coal, coke, porous building material), soil particles are separated as a “heavy” component and contaminant particles as a “light” component using gravity concentration.

Separation of particles intergrown as type 2 or 3 is much more complex. In such cases, contaminant is separated together with bonded soil particles because complete liberation is impossible to achieve. For successful separation, most of the contaminant must be concentrated in a relatively narrow soil particle size range. In the case of type 2 intergrowth, the characteristic property of contaminated particles is particle size. Due to their large specific surface area and adsorption capacity, fine particles (e.g. clay particles in soil) adsorb a relatively larger quantity of contaminant than coarser particles. By separating fine particle size classes, most of the contaminant can be removed. Hydrocyclones are commonly used for that purpose. If contaminant is adsorbed on organic matter or has a distinct hydrophobic properties, separation of contaminated particles can be achieved by gravity concentration or flotation. In the case of type 3 intergrowth, where contaminant penetrates inside the soil particle, separation is only possible if there is a significant difference in one or more physical properties between contaminant and soil particles. In such case, gravity concentration, magnetic separation or flotation is applied. To obtain more efficient separation, additional treatments such as attrition scrubbing and/or comminution are usually required. If contaminant is concentrated in coarser particles (e.g. heavy metals in concrete particles), separation on sieves could be applied. In special cases, contaminant is directly separated by leaching.

4.6. Dewatering

Soil washing process ends with dewatering. Coarse grain size classes (sand, gravel) are dewatered on dewatering screens, and fine classes (sludges) in thickeners, filters or centrifuges, depending on particle size distribution and required final water content in product. Washed clean products can be returned to the original location or used for municipal landfill covering, road construction, concrete elements production, etc. Separated sludge (concentrated contaminant) is further thermal or biologically treated, or immobilized and disposed as special waste. Wastewater from washing process is cleaned as required, mixed with fresh water and returned to the process.

4.7. Examples of soil washing flowsheets

Figure 3. shows the general flowsheet of soil washing process (Mann, 1999). Contaminated soil is washed and sieved on vibrating screen. Screen oversize is the first clean soil product. Screen undersize enters the hydrocyclone for separation of fines. Hydrocyclone overflow is thickened in lamella thickener. Thickener overflow returns to the process, and the sludge is further thickened in a filter press. Filter cake contains the most of the contaminant. After attrition, hydrocyclone underflow is processed by flotation. Flotation overflow is

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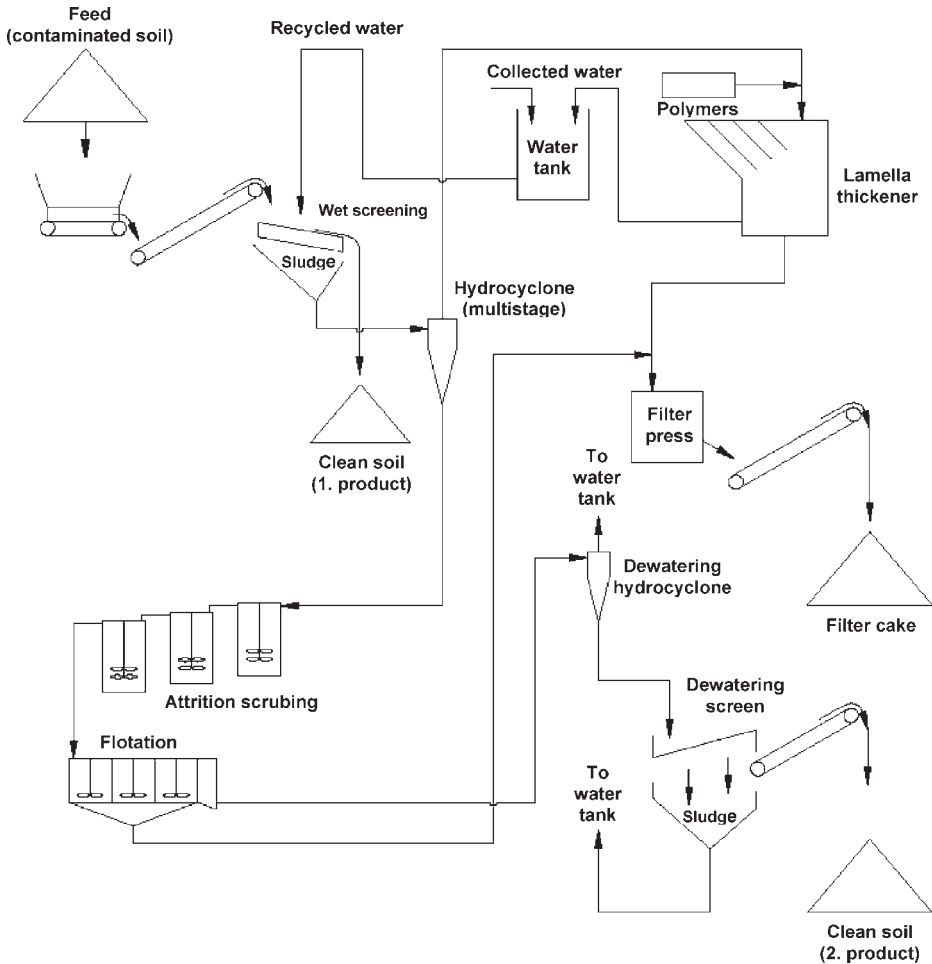


Figure 3. General soil washing flowsheet (Mann, 1999)

thickened in a filter press together with lamella thickener sludge. Flotation underflow, after dewatering in a hydrocyclone and on a screen, is the second clean soil product.

Figure 4. shows technological process of washing soil (14,200 t) contaminated with pesticides and organic mercury (Bunge et al, 1995). Feed material (grain size class -150 mm) is wet screened on vibrating screen with mesh size of 2 mm. Screen oversize is introduced to the washing drum and screen undersize to the hydrocyclone. After washing in drum and additional sieving on screen with mesh size of 2 mm, screen oversize is disposed as a clean product (mixture of coarse sand and gravel), and screen undersize together with the first screen undersize is classified in a

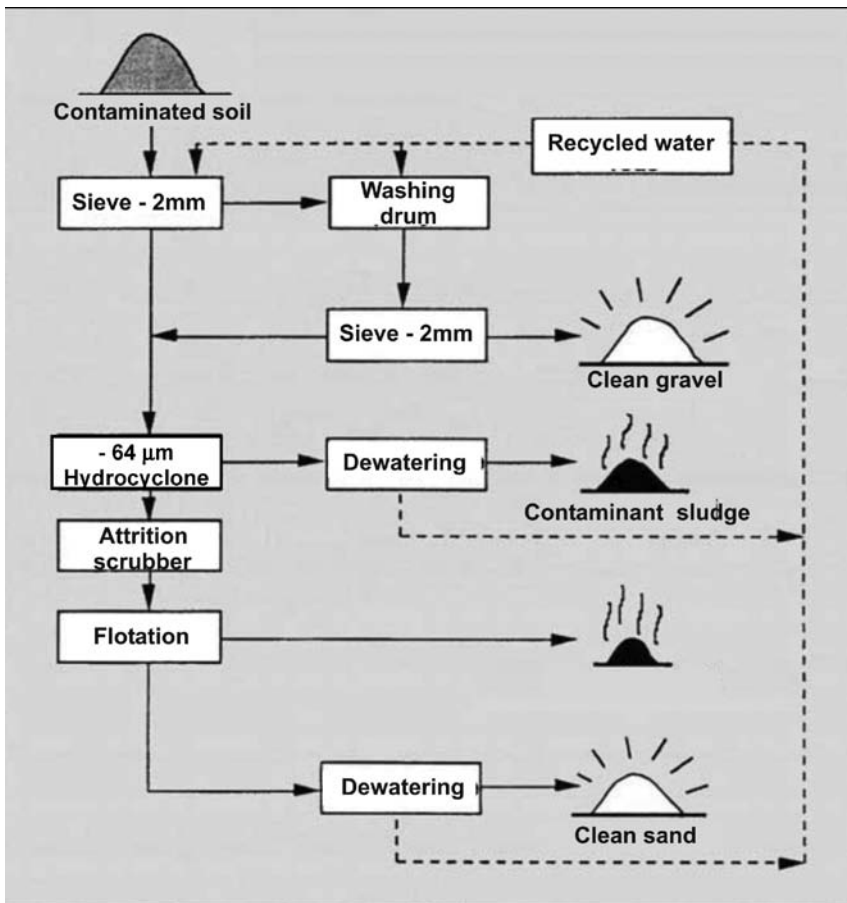


Figure 4. Soil washing flowsheet for the treatment of soil contaminated with pesticides and organic mercury (MBT Environmental Eng. Ltd.)

hydrocyclone. Hydrocyclone cut point is circa 0.064 mm. Hydrocyclone overflow is contaminated sludge which is further treated after dewatering. Hydrocyclone underflow is processed by attrition scrubbing and then by flotation. Contaminated flotation sludge is separated in overflow, and the underflow is disposed, after dewatering, as clean sand. Hydrocyclone and flotation overflows are treated with lime, sealed in barrels and disposed as special waste. Cleaned sand and gravel (screen overflow and flotation overflow) are returned to the original site. Water collected in dewatering process is, after fines removal, returned to the process. Total mass of clean products in presented soil washing process were: 7900 t (56% of input material) of material with grain size larger than 2 mm and 5400 t (38% of input material) of material with grain size from 0.064 mm to 2 mm. The mass of contaminated product (particle size smaller than 0.064 mm) was 900 t, i.e. 6% of input material. At the end of the washing process, 85% of contaminant is separated into 6% of total material mass. In addition, during process water treatment a part of contaminant is neutralized, therefore the amount of separated contaminant was 95% of total contamination. Soil washing plant capacity was 10 t/h.

Figure 5. shows the Deconterra process developed by Lurgi Company and used for washing soils contaminated with oil and oil derivatives (Hankel et al, 1992; Schneider, 1992).

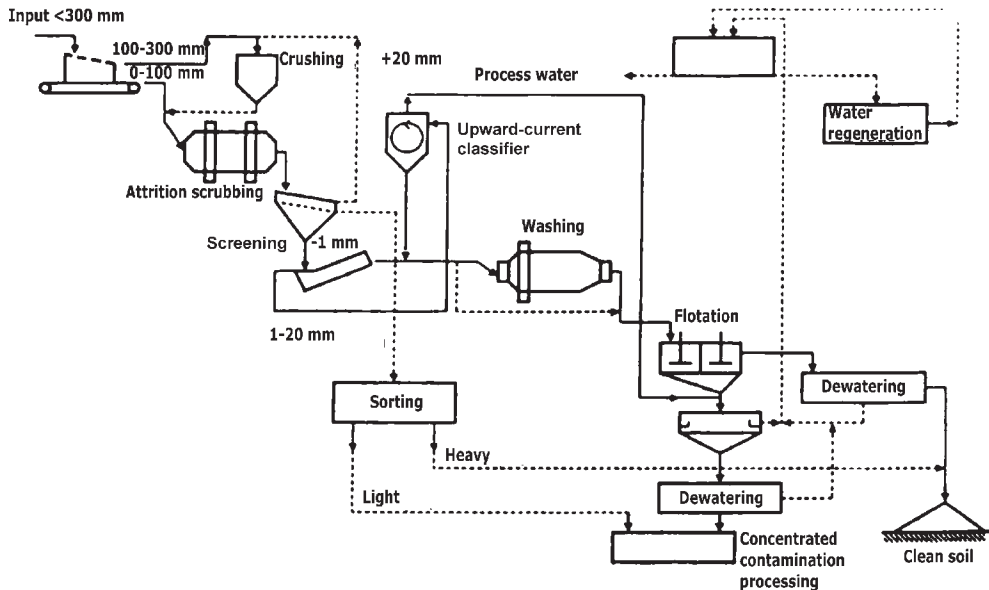


Figure 5. Soil washing flowsheet (Lurgi-Deconterra process)

Material is screened on a vibrating grizzly with 100 mm openings. Grizzly oversize is comminuted in impact crusher, and undersize is washed together with comminuted material in attrition drum. In attrition drum, material is deagglomerated and adhering contaminant is partially scrubbed from particle surface. After that, material is classified on double-deck vibrating screen with mesh sizes of 20 mm and 1 mm. Grain size class +20mm is additionally comminuted in a crusher, size class 20/1 mm concentrated in a jig, and size class -1 mm classified in spiral classifier. Classifier overflow is classified in upward-current classifier. Upward-current classifier underflow together with sand from spiral classifier is additionally washed in attrition drum. Drum output material is introduced to the flotation cell. After dewatering, flotation underflow is disposed as a clean product. Flotation and classifier overflows are dewatered and further treated. Heavy fraction separated in the jig is disposed as a clean product, and the light fraction is further processed. The water separated during dewatering is returned to the process. In the shown process, hydrocarbons concentration of 400-4000 mg/kg can be reduced to less than 50 mg/kg. More than 96% of the total contaminant is concentrated in the flotation and upward-current classifier overflow and in the jig light fraction. Heavy metal content are also partially removed during soil washing.

5. Research work

In recent years, a number of experiments, which results can be directly applied in studying and designing the soil washing processes, have been conducted in the Laboratory for Mineral Processing and Environmental Protection of the Faculty of Mining, Geology and Petroleum Engineering, University of Zagreb, as part of projects financed by the Ministry of Science, Education and Sport of the Republic of Croatia.

In the field of gravity concentration, the separation of coal particles from sand samples extracted from the Sava River was tested. Coal content in input samples ranged from 1% to 6%. Samples of grain size class 4/1 mm were treated in a jig, and samples of grain size class 4/0.5 mm on a shaking table and Humphreys spiral. The results of performed tests showed reduction of coal content in sand down to between 0.03% and 1.37% with recovery of 70-80% after concentration in a jig, between 0.06 and 1.05% with recovery of 84-95% after concentration on a shaking table and

between 0.16 and 1.25% with recovery of 95-98% after concentration in the spiral.

Hard coal floatability was tested in flotation column. Flotation of coal samples with 27-30% ash content, resulted in a concentrate with 8.9-9.5% ash content and 63.6-64.8% recovery. Underflow ash content was 53.9-61.4%.

Attrition scrubbing of silica sand (solids concentration in the suspension: 77%, impeller speed: 560 min⁻¹, attrition time: 20 min) resulted in the reduction of Fe₂O₃ content by 23% and Al₂O₃ content by 34%, while the content of SiO₂ was increased by 2,44%.

6. Conclusion

Overview of the achieved results in soil washing technology implies the need for further systematic fundamental research. Special attention should be paid to the investigation of liberation mechanism and contaminant redistribution depending on the type of intergrowth between contaminant and soil particles, as well as to the type and amount of energy needed for optimal liberation. Further research of physical separation processes should lead to finding more efficient flotation reagents and highly adsorptive minerals and organics for each type of contaminant.

The field of soil remediation is fast developing. The technologies are very expensive and increasingly demanding, therefore there is a risk that achieved results would not always justify the process costs. Each case should be thoroughly researched and analyzed in order to enable the choice of optimal technology, reliable evaluation criteria and control of the whole system based on scientific research.

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