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# HEAVY METALS IN THE COAL MINES ENVIRONMENT, THEIR TOXICITY AND REMEDIATION

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**ABSTRACT:** Heavy metals are natural constituents of the earth's crust, resulting from the natural activities such as weathering, volcanic activity and anthropogenic activities like mining, manufacturing and industrialization. Coal mining is also considered one of the sources of heavy metal contamination in the surrounding environment. Heavy metals, particularly those used in excess, have raised concern for human health. Their impacts on the environment too have been studied extensively. Some of the heavy metals like Cr, Cd, Pb, Se, As, Hg, Co, Ni and Zn are of severe concern for the researchers in terms of their environmental load and health effects. Moreover, due to their non-biodegradability, it is very challenging to remove metals from the environment. Several researcher groups, chemical engineers and environmentalists are taking their efforts to mitigate the excessive release of heavy metals from coal mines to the environment. Therefore, numerous remediation strategies such as adsorption, ion-exchange, flocculation, coagulation, precipitation, electrochemical, ultra-filtration, phytoremediation and bioremediation methods have been developed and applied in order to remove the toxic metals from the coal mine environment. This review article briefly summarizes coal mining released heavy metals, their adverse impacts on living beings as well as on environment and their remediation approaches in detail.

**Key words :** - Coal mines, Heavy metals, Toxicity, Remediation

## INTRODUCTION :

The transformation of soil caused by human activity continues to expand worldwide. In the wake of the industrial revolution, coal, the black gold of the earth, has become an increasingly valuable resource around the world. There is more than 7 billion tons of coal in the world today and it makes up about 75% of the world's total energy demand. India currently ranks second after China in coal production globally (I.E. Agency, Coal Information, Paris, 2018.). Environmental pollution is now posing a serious threat to biodiversity and ecosystem processes due to the rapid pace of industrialization, increased demand for energy and reckless exploitation of natural resources throughout the past century. One of the leading causes of environmental degradation is coal mining, particularly in opencast mining areas (Chandra et al.2015). As a consequence of opencast coal mining, variations in the composition and properties of soil and water have occurred in the surrounding area (Quadros et al. 2016). Thus, opencast coal mining negatively impacts ecosystems, pedogenesis, revegetation and environmental restoration (Singh et al.2007). In addition to degrading the natural landscape, coal and overburden dumps interfere with the natural drainage system and delay normal plant growth (De & Mitra, 2002). In coal mining processes, iron-sulfide minerals are exposed to atmospheric conditions, which promote acid formation (Banerjee, 2014). A majority of water contamination comes from coal leachate and runoff from overburdened dumps ( Singh, 2005). Especially in opencast mining, coal and other minerals extraction processes transform topsoil dramatically. In the process of drilling and blasting, massive amounts of dust are released, which contains toxic pollutants, including heavy metals and carcinogenic gases(benzo(a)pyrene) (Ye et al.2019). There is a significant amount of effluent discharged into surface waters during coal mining. Inorganic and organic pollutants are discharged into the water bodies, which further contribute to soil and sediment pollution and heavy metals are among the most hazardous pollutants affecting the environment. The effluent from coal is also associated with a load of TSS, TDS, calcium carbonate and trace metals, which can contaminate the soil and aquatic environment and retards plant growth (Pandey et al.2014). Over the last few decades, heavy metal ions have been widely used, resulting in an increase in metallic substances in aquatic and terrestrial environments (Knopf and Konig, 2010). There are many factors contributing to the metal load in all segments of the environment, including acid mine drainage, industrial and domestic effluents, agricultural runoff and acid rain (Haynes et al.2009). As non-biodegradable and persistent contaminants, heavy metals pose particular risks. A large number of metals have existed on earth for a long time and about 75% of the elements in the periodic table are metals. According to the USEPA, trace inorganic contaminants are mostly heavy metals (Wuana et al.2010). The long-term toxic effects of dissolved metal ions in water have increased public awareness in recent years. A natural process rarely eliminates heavy metals from an aquatic ecosystem, unlike most organic pollutants. A unique property of these contaminants is that they accumulate along a food chain over an extended period of time and they can become toxic to aquatic and human lives (Papageorgiou et al 2006). As a result, the heavy metals concentration increases and causes physiological and biological problems.

This review article briefly summarizes coal mining released heavy metals, their adverse impacts on living beings and the environment and their remediation approaches in detail.

2. Heavy metals and their toxicity:

The metals which have high atomic weight and density are termed as heavy metals. These are toxic for organisms even at very low metal ions concentration (Suganya & Kumar 2019). In some cases metalloids are also considered as heavy metals; which have the ability to act as both metals and non-metals. Different sources of heavy metals in soils, their toxicity and their permissible limits in drinking water set by United States Environmental Protection Agency (USEPA) and Indian Standards Institution (ISI) are listed in Table 1.

A major contributor to coal mine pollution is the leaching of acidic mine tailings known as 'Acid Mine Drainage (AMD). AMD results in the leaching of metalloid, arsenic and heavy metals like manganese, aluminium, chromium, cadmium and silting of coal and sand, which may cause environmental hazards (Johnson and Hallberg, 2005).

MATERIAL AND METHODS:

**a.** 3. Heavy metals impact on human health and environment:

**b.** Metals play an essential role in the ecosystem, whose biologically available concentrations depend mainly on geological and biological processes. In nature, there are 92 elements, of which 81 are essential for human body (Fe, I, Cu, Zn, Co, Cr, Mo, Ni, V, Se, Mn, As, F, Si and Li)and four are con-ditionally essential (Cd, Pb, Sn and Rb). As a result of bioaccumulation and biomagnification, heavy metals in soil adversely affect living organ-isms (Szynkowska et al. 2018). Moreover, heavy metals can replace the ions of the main metals needed by humans and living systems. As a re-sult, metabolic processes and biochemical reactions are disrupted during food consumption followed by metabolites are also removed from the body. By accumulating excessive amounts of heavy metals, protein compounds begin to break down, peptide bonds are ruptured, free radicals are re-leased and vulnerable organs (kidneys, liver, brain and blood vessels) are severely damaged. During coal transportation, several pollutants are also deposited on the routes along with dust. Heavy metals accumulate in soils for a long time and their excessive amounts affect plant growth, metabo-lism, physiology and aging (Ashraf et al.2019).

**c.** Microorganisms are one of the first group of biota to be directly or indirect-ly affected by heavy metals. At low concentrations, certain metals (e.g. Fe, Zn, Cu, Ni, Co) are essential for microbial activity. In contrast, high con-centrations of heavy metals may have toxic or inhibitory effects on living organisms. Metals adversely affect soil microbes, causing decreased de-composition of organic matter, reduced soil respiration, decreased diversity and declined activity of several enzymes (Tyler, 1974). Furthermore, heavy metals alter protein and nucleic acid conformational structures, which render them inactive by forming complexes with them. These effects dis-rupt the integrity of the membrane of microbial cells or cause them to die (Bong et al.2010).

**d.** 4. Remediation approaches for heavy Metal Contaminated Soils:

**e.** In order to prevent the spread of toxic heavy metal contaminants in soils and bodies of water, engineering and biological remediation approach are employed. Each approach has its pros and cons, which is addressed in this section.

**f.** 4.1 Engineering remediation: Physical and Chemical method has been routinely employed in this engineering remediation approaches.

**g.**

**h.**

**i.** 4.1.1Physical method:

**j.** 4.1.1.1 Soil Replacement:

**k.** Often called "dig-and-haul," this method involves replacing of metal-contaminated soil with clean soil. As a result of this method, the contami-nated soil is moved from a more risky area to a safer one. The process is very tedious, costly and it's best applied to smaller, heavily polluted areas only (RoyChowdhury et al. 2018).

**l.** 4.1.1.2 Soil Washing:

**m.** Heavy metal contamination in severely contaminated soil can be mini-mized by soil washing. This process can be carried out either in-situ by forcing washing solution through the soil matrix or ex-situ by excavating soil and washing it in a reactor (Zhou et al. 2004). This method makes heavy metals soluble in washing reagents and separates them from soil. Sand and granular soils are more effective for soil washing than clay-rich soils, since clay creates problems with solid-liquid separation. In order to remove heavy metals from soil, a variety of chemicals are used, including chelating agents, surfactants, organic acids and cosolvents. Several re-searchers reported the successful removal of heavy metals like Ni, Zn, Pb, Cd and Cu, from contaminated soils in a significant amount using soil washing methods (Zhou et al.2004).Nonetheless, this method is expensive and the washing solutions need to be handled and treated properly after-ward.

**n.** 4.1.1.3 Thermal Desorption:

**o.** Thermal desorption relies on the volatility of the metal compounds in pol-luted soils. Using steam, ultraviolet radiation, or microwaves, polluted soil is heated first to make the pollutant compounds volatile (Dhaliwal et al.2020). The volatile contaminants are then extracted using vacuum neg-ative pressure or a carrier gas, allowing the pollutants to be separated from the soil. Depending on the temperature, thermal desorption can be classified as high temperature desorption (320~560°C) or low temperature desorption (90~320°C). This technique is much simpler than the soil re-placement method, the equipment is transportable and the remediated soil can be reused. Nevertheless, it has limitations, such as drastic changes in soil quality, expensive devices and long desorption times (Li et al.2019).

**p.** 4.1.2 Chemical Method:

**q.** 4.1.2.1 Chemical Precipitation: It is one of the oldest and most widely used methods for the

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removal of metals from wastewater. It involves the addition of chemicals to modify the physical state of dissolved or suspend-ed solids in order to facilitate the sedimentation of these solids. Metals can be precipitated by adding coagulants such as alum, lime, iron salts and polymers (Wuana et al. 2010). Chemical precipitation is employed for most of the metals and the most common precipitants used for the precipitation are hydroxide, sulfide, carbonate and phosphate (Pandya, 2015).

**r.** 4.1.2.2 Membrane Process: Heavy metals are removed efficiently from wa-ter bodies with membrane filtration, which is categorized into ultrafiltra-tion and nanofiltration based on the particle size. UF (ultrafiltration) sepa-rates metal ions based on pore sizes of 5-20 nm and molecular weights of 1-10 kDa. More than 90% of metal is removed by UF when the metal con-centration is between 10 and 112 mg/L. Nanofiltration comprises a semi-permeable organic membrane with pores between 0.1 and 2 nm in size and a molecular weight of about 200 Da (Pandya ,2015). Nanofiltration is driven by the pressure difference over the membrane. Another important examples of membrane process applicable to inorganic wastewater treat-ment include reverse osmosis and eletrodialysis. This process involves the use of selective membranes with a specific driving force to concentrate ions. In reverse osmosis (RO), the membrane has minute pores that can be used for water recycling and metal recovery. The process involves reversing the osmotic pressure of a solution in order to push it through a membrane that retains the solution on one side while allowing the solutes to pass through pure water on the other (Azimi et al.2017). The electrodialysis process is based on the movement of ions through selectively permeable membranes in response to current applied to electrodes. These membrane processes are costly and sophisticated, requiring a skilled person to han-dle.

**s.** 4.1.2.3 Adsorption:

**t.** As a surface phenomenon, adsorption includes drawing and holding ad-sorbate molecules to the surface of an adsorbent, either through physical interaction (physisorption) or through chemical forces (chemisorption) be-tween adsorbate and adsorbent. Adsorption method is based on the fact that almost all heavy metal ions can be fixed and adsorbed by adsorbent like activated carbon, clay minerals like bentonite, zeolite, etc, (Yuan et al. 2009), microbial biomasses (Fomina and Gadd, 2014), metal oxides and polymeric materials (Zare et al. 2016) Adsorption is commonly used to re-move heavy metal ions due to its high efficiency, remarkable simplicity and low cost.

**u.** 4.1.2.4 Ion Exchange:

**v.** Ion exchange is an effective process for the removal of heavy metals in the water treatment industry containing low effluent concentrations. In this process, a special ion exchanger containing cations or anions requires for the removal of metal ions from the solution. The typical matrices used in ion exchange are synthetic organic ion exchange resins (Pandya, 2015). Cationic resins may absorb nickel, copper and zinc ions, whereas anion exchange resins are ideal for minimal soil or water contamination and are used to absorb chromate, sulphate, nitrate and cyanide ions. Following the separation of the resin from the elution, valuable heavy metals can al-so be recovered using ion exchange.

**w.** 4.1.2.5 Immobilization:

**x.** Soil heavy metals are partitioned into soluble, exchangeable, Fe/Al bound, Ca/Mg bound, organic matter bound and residual form. Metals that are soluble in water are the most toxic to living organisms. Immobilization re-duces the solubility of metals in soil and thus their mobility. A number of organic and inorganic immobilizing agents like organic composts, clay, cement, phosphates, minerals and zeolites are utilized to reduce metal toxicity by precipitation, sorption and complexation (Fin_zgar et al.2006). Metals are precipitated in soil matrix as metal hydroxides as a result of this process. Since toxic organic vapors can form in the presence of organic contaminants, makes this method ineffective (Farrell et al. 2010). Also, physicochemical techniques for heavy metal remediation are generally ex-pensive and cause side effects. Consequently, continuous efforts have been made to develop techniques that are economical, sustainable and easy to use.

**y.** 4.2 Bioremediation:

**z.** Bioremediation offers the potential to contribute to achieving this goal in a sustainable and eco-friendly manner. Bioremediation involves the use of microorganism and plant species to eradicate heavy metals from the envi-ronment. Heavy metal environments have enabled microorganisms and some plant to get adapted to them and develop metabolic mechanisms to easily detoxify heavy metals.

**aa.**4.2.1 Microbial remediation:

**bb.** Microorganisms play an essential role in the environmental biogeochemi-cal cycle of metals and their properties are of significant interest in the remediation of contaminated sites. Microbial sorption and transformation of metals is a promising solution to resolving pollution issues (Han and Gu, 2010). Heavy metal-tolerant microorganisms are of great interest as bioremediation agents since they have evolved several types of mecha-nisms to tolerate heavy metal ions and transform them. These mechanisms involve the efflux of metal ions outside the cell, accumulation and complexation of the metal ions inside the cell and its reduction to a less toxic form. Heavy metals are not degraded, but instead transformed into non-toxic forms by altering their physical and chemical properties (Dhali-wal et al.2020). The universality, size and ability to reproduce in harsh environmental conditions makes bacteria important biosorbents in bioremediation. The majority of organisms are capable of detoxification (i.e. mineralization, transformation and/or immobilization of pollutants), thereby maintaining the biosphere's sustainability (Miyata et al. 2007). In order to remove heavy metals, microorganisms use three mechanisms; the first involve the biosorption of metal on the cell surface, the second is intracellular uptake and the third they undergo chemical transformation (Zimmerman, 2010).

Microorganisms play an essential role in the environmental biogeochemi-cal cycle of metals and their properties are of significant interest in the remediation of contaminated sites. Microbial sorption and transformation of metals is a promising solution to resolving pollution issues (Han and Gu, 2010). Heavy metal-tolerant microorganisms are of great interest as bioremediation agents since they have evolved several types of mecha-nisms to tolerate heavy metal ions and transform them. These mechanisms involve the efflux of metal ions outside the cell, accumulation and complexation of the metal ions inside the cell and its reduction to a less toxic form. Heavy metals are not degraded, but instead transformed into non-toxic forms by altering their physical and chemical properties (Dhali-wal et al.2020). The universality, size and ability to reproduce in harsh environmental conditions makes bacteria important biosorbents in bioremediation. The majority of organisms are capable of detoxification (i.e. mineralization, transformation and/or immobilization of pollutants), thereby maintaining the biosphere's sustainability (Miyata et al. 2007). In order to remove heavy metals, microorganisms use three mechanisms; the first involve the biosorption of metal on the cell surface, the second is intracellular uptake and the third they undergo chemical transformation (Zimmerman, 2010). Among the different heavy metal removable tecniques, the biosorption method has been found to be extremely selec-tive for the removal of metals from multi-element systems. In addition, metal-accumulating bacteria also employed for eradicating the heavy met-als from industrial effluents metabolism. Pollution by heavy metals impos-es selective pressure on microbial communities, causing them to develop resistant strains with a reduction in their extracellular enzyme activities. For studying the harmful effects of metals at the cellular level, microorgan-isms, such as bacteria, algae, fungi and yeast, serve as constructive mod-els (Wiatrowski et al.2006). Moreover, these microorganisms are effective for the eradication of toxic metals from contaminated sites since they can accumulate heavy metals efficiently. Different microbial remediation ap-proach and microorganisms has been used to remove heavy metals from the environment which is represented in Table 2. Several microbial genera and species show potential for metal biosorption; fungi were found to be efficient biosorbent organisms as they contain a high percentage of cell wall material in their cells. The mixed culture is metabolically preferable for bioremediation in the field, since bacteria survive and are more stable in a mixed culture.

4.2.2 BIOREMEDIATION USING PLANT SPECIES (Phytoremediation):

Phytoremediation involves extracting pollutants from contaminated soil with the roots of trees, shrubs and herbaceous plants (Shah and Daverey, 2020). Plants absorb very small amounts of iron, manganese, copper, nickel and zinc. Furthermore, certain plants are capable of absorbing toxic heavy metals, such as chromium, arsenic, cadmium, lead, mercury and others, without suffering serious damage. These plants referred as hyper-accumulators, capable of accumulating large quantities of pollutants without displaying sign of phytotoxicity in their aerial parts. There are numerous plants that fall into this group, includes Pteris vittata, Bidens pilosa, Jatropha curcas and Helianthus annuus, absorb at least 100 mg/kg of arsenic and cadmium and 1000 mg/kg of copper, cobalt, chro-mium, manganese, nickel and lead (Han et al.2017). Currently, over 400 species of such plants have been found globally and most of them belong to Cruciferae family, including the genus Alyssums, Brassica and Thlaspi (Xing et al.2003). Stress control mechanisms play a crucial role in main-taining homeostasis of metal in plants, leads to plants tolerant to metal contamination by forming less toxic metal complexes with active metabo-lites excreted through the root system. The root system of plants absorbs toxic elements, which are then decomposed or converted into safer forms (Ansari et al.2020). There are several factors that influence the results, in-cluding the plants' tolerance to pollutants, the biomass volume and the efficiency at which pollutant is transported from roots to shoots. There are different techniques of phytoremediation including phytoextraction, phy-tostabilization, phytofilteration and phytovolatilization depending upon the mechanism of remediation enumerated in table 3. A phytoremediation process is time consuming as it may take several years to clean up a heavy metal-contaminated site, compared to other conventional physical and chemical remediation methods. It is observed that a combination of As-resistant grapevine species plants and microorganism such as Bacillus licheniformis, Micrococcus luteus and P. fluorescens enhances phytore-mediation efficiency .This combination activated siderophore production, phosphate solubilisation and nitrogen fixation (Pinter et al.2016).

A study by Jiang et al. indicated that microorganisms found in the rhizosphere of plants growing near refineries and chemical plants helped plants adapt to the environment. In this study they isolated $Pb^{2+} > Zn^{2+} > Cu^{2+} > Cd^{2+}$ resistant *Pseudomonas, Cupriavidus* and *Bacillus* from the rhizosphere of Boehmeria nivea and consequently help plants survive in the soil with high concentrations of heavy metals (Jiang et al.2017).

**CONCLUSION:**

An anthropogenic activity like coal mining sites is responsible for the complete transformation of natural landscapes and the release of a variety of heavy metals into the environment. As a result, all living systems (plants, animals and microbes) are negatively affected, resulting in medical and social problems, as well as an increased incidence of all diseases, including the most severe ones. Even though a number of measures have been employed to remove even toxic levels of contamination, many degenerated areas like coal mines can still not be successfully treated. Thus, continuous efforts are being made to further improve the remedial techniques in order to make them more effective, flexible and to reduce the side effects and cost of the procedures. In this review, a variety of methods have been proposed for remediation and reclamation of soil. The most promising and accessible methods is bioremediation, which involves either plant communities or microorganism communities. A consortium of various microorganisms introduced into the rhizoplane of the plant is another promising strategy. A symbiotic relationship is the key to make this approach effective. Furthermore, biotechnological approaches might facilitate some advancement in bioremediation methods, which provides its successful application in the heavy metal remediation in all heavy metal polluted sector including coal mines.

**Conflict of interests:**

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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| Microbial remediation techniques | Mechanism process | Microbes involved | Reference |
|---|---|---|---|
| Bioleaching | Based on mobilization of metal cations from insoluble ores by biological oxidation and complexation, used in situ or ex situ to help to remove the pollutants from soils. | Acidithiobacillus (A.thiooxidans and A. ferrooxidans), Acidiphilium, Acidimicrobiu, Ferromicrobium | Ye F et al.2019 |
| Biostimulation | Uses growth rate stimulation nutrients, electron donors or acceptors to encourage the growth of site-specific indigenous microorganisms capable of degrading environmental pollutants | Escherichia coli, Pseudomonas maltophilia, Shewanella putrefaciens, Pseudomonas aeruginosa, Ent.cloacae | Zimmerman, 2010 |
| Bioaugmentation | Introduction of specific competent microorganisms to the local microbial population in order to increase the metabolic capacities needed for remediation | Bacillus spp. | Wiatrowski et al.2006 |
| Biosorption | Biosorption of metal ions on the cell surface, intracellular uptake of metal ions followed by transformation of metal ions | Aureobasidium pullulans, Cladosporium resinae, Aspergillus niger,Aspergillus versicolor or Rhizopus nigricans | Zhou et al.2004 |

Table 2: Microbial remediation approach

| Heavy metals | Sources | Permissible limit | | Toxicity |
|---|---|---|---|---|
| | | USEPA (mg/L) | Indian Standards (mg/L) | |
| Cr | Solid waste, tanneries Mining; Disposal of industrial wastes; Fertilisers; Flying ash; | 0.05 | 0.05 | point mutations in DNA, chromium poisoning, ulcer, allergic dermatitis, lung cancer, liver necrosis, brain damage, kidney problems, birth defects |
| As | Electroplating industry, coal Mining; volcanoes, petroleum refining, Disposal of industrial wastes; Pesticides ,Fossil fuel combustion | 0.05 | 0.05 | chromosomal abnormalities, neoplastic changes of the skin, Bowen's disease |
| Cd | Electroplating; Geogenic sources, metal smelting and refining, Industrial Wastes; Paints and pigments; Fertilizers | 0.005 | 0.01 | lungs, kidneys, skeletal and cardiovascular systems are all affected adversely, premature birth |
| Pb | Mining and smelting of metalliferous ores, burning of leaded gasoline,paints, Batteries waste; Pesticides | 0.05 | 0.1 | mental retardation, birth defects, allergies, dyslexia, hyperactivity, paralysis, muscular weakness, brain damage, kidney damage |
| Cu | Smelting and refining, mining, biosolids, Electroplating industry | 0.25 | 0.25 | Wilson's diseas, liver damage, |
| Hg | Volcano eruptions, emissions from industries producing caustic soda, coal Mining,Fossil Fuel Combustion; | 0.002 | 0.001 | brain damage, lung damage, vomiting, diarrhea, nausea, skin rashes, or increased blood pressure or heart rate |
| Se | Oil refining, Coal mining, combustion of fossil fuels, chemical synthesis (e.g., pigment formulation), | 0.05 | 0.05 | nausea, vomiting and diarrhea, Loss of hair and nails, bad teeth etc |
| Ni | Land fill, Volcanic eruptions, bubble bursting and gas exchange in ocean, weathering of soils | 0.1 | 0.05 | chronic asthma,nausea, dermatitis,human carcinogen |
| Zn | Mining; smelting and refining, Fossil fuel combustion; Industrial Wastewater | 0.80 | 0.80 | Depression, lethargy, neurological signs and increased thirst |

| Techniques of phytoremediation | Transformation process | Reference |
|---|---|---|
| Phytoextraction | Plants absorb, translocate and store toxic contaminants into their root and shoot tissue which then enter its aerial parts | Ansari,2020 |
| Phytostabilisation | Contaminant immobilized in the soil and groundwater and prevented their entry into the food chain. Stabilization is also enabled by precipitation within the root zone of plants (rhizosphere) | Yu,D.M, 2021 |
| Phytovolatilization | Plants convert pollutants into volatile forms that enter the atmosphere | Yu,D.M., 2021 |
| Phytodegradation | Plant biodegraded Organic pollutant under the action of various enzymes such as peroxidase, dehalogenase, nitroreductase and others | Ansari,2020 |
| Rhizofiltration | Plants secrete special organic compounds in order to attract microbial communities followed by pollutant are adsorbed by plant roots with a developed fibrous system. | Yu,D.M., 2021 |
| Rhizodegradation | Microorganisms, including bacteria, fungi and yeast, whichlive in the plant's root system in symbiotic manner ,removes such contaminants as pesticides, oil and PCBs | Yu,D.M., 2021 |

Table 3: Pollutant transformation processes in plants.