4 Effective Removal of Radioactive Waste from Environment Using Plants

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Abstract

Radionuclides released into the environment pose threat to human health and the environment, hence their remediation becomes mandatory. Phytoremediation technology assists in the removal of radioactive contaminants from the environment in an effective way. Plant surfaces take up radionuclides present in the atmosphere by leaves and those immersed in water or bound to soil through roots. Terrestrial and aquatic plants both remove radionuclides by mechanisms such as accumulation and rhizofiltration. The plasma membrane transporters present on the surface of cells facilitate the transfer of radioactive elements in plants. The potential of plants can be exploited for developing eco-friendly technologies for large scale remediation of harmful radioactive elements from the environment.

Keywords

Phytoremediation · Rhizofiltration · Radionuclides · Phytoextraction · Radioactive elements · Phytovolatilization

4.1 Introduction

Radionuclides get added to the environment through releases from various sources such as nuclear tests, nuclear weapons, accidental spills, and discharges from nuclear facilities or operations (Table 4.1). Radionuclide emissions in the atmosphere include fallout from atmospheric bomb, nuclear accidents, emissions from reprocessing plants, nuclear power stations (isotopes are ^{3}H, ^{14}C, ^{35}S), and waste disposal sites (^{14}C, ^{36}Cl, ^{99}Tc, ^{129}I, ^{135}Cs, ^{237}Np and ^{239}Pu, ^{240}Pu, ^{42}Pu) (Hattink

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Table 4.1 Major sources of radioactive contamination in the environment

Source	Radionuclides
Nuclear weapon testing	^{14}C, ^{137}Cs, ^{90}Sr, ^{95}Zr
Nuclear weapon production	^{137}Cs, ^{106}Ru, ^{95}Zr
Discharges in mining	^{226}Ra, ^{210}Pb, ^{210}Po, ^{232}Th, ^{222}Rn
Mining and milling sites	^{238}U
Nuclear accidents	^{137}Cs, ^{90}Sr, ^{131}I, ^{210}Po, ^{95}Zr, ^{144}Ce

et al. 2000; Ould-Dada et al. 2001; Peles et al. 2002). In addition, natural sources such as parent material, organic/mineral component, soil solution, leakage from unnatural sources such as buried radioactive materials in the soil also contribute to radioactive pollution. The presence of radionuclides in soil and water affects ecosystem stability and poses a threat to human health.

A variety of physicochemical methods such as washing, ion exchange, leaching with chelating agents, flocculation, and reverse osmosis have been used for the treatment of radionuclide contamination. These methods remove radioactive elements from soil and water but the efficiency of radionuclide removal varies depending upon the chemistry of the element, rate of deposition, and decay. Limitations of these methodologies generated the need for framing environmentally safe and affordable technologies. Phytoremediation proved to be an effective alternate to remove a variety of contaminants including radioactive elements from the environment (Zhu and Shaw 2000; Dhir 2013; Malhotra et al. 2014; Yan et al. 2021).

Phytoextraction, rhizofiltration, phytovolatilization, and phytostabilization are the major phytoremediation techniques involved in the remediation of radionuclides. Phytoextraction is a process in which plant biomass accumulates radionuclides and radionuclides get transported from soil into the aboveground parts of the plant which are harvested. It depends on the natural ability of vascular plants to take chemical elements through the roots, deliver them to the vascular tissue, transport and compartmentalize radioactive elements in the aboveground biomass. The phytoextraction technology has proven effective in treating areas having low-level of radionuclide contamination. Rhizofiltration utilizes plant roots to precipitate and concentrate radionuclides from polluted sites while in phytovolatilization plants remove radionuclides (such as ^{3}H) from the leaves/ foliage via volatilization (Prasad 2007). In phytostabilization process plants stabilize radionuclides in soils rendering them harmless. The features such as absorption, translocation, bioaccumulation, and contaminant degradation help plants to remove contaminants such as radionuclides from the environment (Yadav and Kumar 2019).

The present chapter highlights the role of plants in remediation/removal of the radioactive contamination present in the soil or water. The potential of the plants for removing radionuclides from the environment has been assessed.

4.2 Removal of Radionuclides by Plants

Radionuclides get deposited on the external plant surface directly from the atmosphere or by wet/dry deposition via resuspension from soil (Bell et al. 1988; Tagami 2012). High level of radionuclide accumulation has been reported in plants growing in soils having huge radioactive deposits and mine tailings.

4.2.1 Accumulation and Uptake of Radionuclides by Plants

Uptake and accumulation of radionuclides by various crop plants and tree species has been well documented (Planinšek et al. 2018; Duong et al. 2021) (Tables 4.2 and 4.3). Phytoextraction technique has proven to be the major mechanism involved in the uptake of radionuclides such as ^{90}Sr (strontium), ^{95}Nb (niobium), ^{99}Tc (techtenium), ^{106}Ru (rubidium), ^{144}Ce (cesium), ^{226,228}Ra (radon), ^{239,240}Pu (plutonium), ^{241}Am (americium), ^{228,230,232}Th (thorium), ^{244}Cm (curium), ^{237}Np (neptunium) (Kabata-Pendias and Pendias 1996; Dushenkov 2003; Hattink et al. 2004). Phytoextraction of radionuclides depends on the bioavailability of radionuclides in soil, rate of uptake by plant roots and efficiency of radionuclide transport through the vascular system. The rate of transfer uptake of radionuclide from soil/water to plant is related to transfer factors (TF) which gives a measure of ratio of concentration of element in the plant to that present in the mine tailings or soil. TF can be used as an index for the growth of a target element in the plant and its transfer from the medium to the plant. This factor varies for each plant. The difference in TF values for the plants tissues may be due to change in metabolic rate.

Table 4.2 Radionuclide accumulation in crop plants

Element	Plant species
Co	Broccoli, tomato
Rb	Tomato, chard, sunflower, cucumber
Sr	Cucumber, sunflower, turnip
Cs	Tomato, chard, cucumber, sweet potato
U	Sunflower

Table 4.3 Radioactive elements taken up by tree species

Tree species	Radioactive elements
Paxistima myrsinites	^{210}Pb, ^{210}Po
Ribes lacustre	^{210}Pb, ^{210}Po
Abies lasiocarpa	^{226}Ra
Pinus contorta	^{226}Ra
Salix scouleriana *S. scouleriana*	^{226}Ra
Alnus incana	^{226}Ra
Larix occidentalis	^{226}Ra
Pseudotsuga menziesii	^{226}Ra
Acacia auriculiformis	^{226}Ra, ^{238}U, ^{137}Cs, ^{228}Ra, ^{40}K

Phytovolatilization uses the ability of plant to transpire enormous amounts of water. It is used for remediation of ^{3}H (Tritium), a radioactive isotope of hydrogen. A small portion of ^{3}H is absorbed by plant roots and most of it remains in the plant tissues in the form of easily exchangeable hydroxyl ions or is incorporated into organic molecules through photosynthesis. The roots of terrestrial plants efficiently remove radionuclides such as uranium (U) from aqueous streams. The process is referred as rhizofiltration. High concentrations of uranium get accumulated in the roots and bioaccumulation coefficients above 30,000 have been noted for the element.

Besides, phytostabilization, phytostimulation, phytotransformation, phytofiltration, and phytoextraction, combination of plant–microbe interaction also help in removal/remediation of radionuclides from soil. Microbes are known possess great potential to bio-transform, biosorb, and biomineralize radionuclides through their inherent catabolic process. Microbe assisted phytoremediation technique has proved beneficial in improving the mobility/removal of radionuclides from soil surfaces (Sarma and Prasad 2018; Thakare et al. 2021). The technique has played a role in restore balance of the soil (Lajayer et al. 2019). Transporters like NRAMP, ZIP families CDF, ATPases (HMAs) family like P1B-ATPases play an important role in phytoremediation of radioactive elements.

Radionuclide accumulation varies according to plant species and radioactive element. A high uptake rate has been found for many elements while low rate has been noted for others. The mobility and uptake of radionuclides in plants is controlled by external factors such as chemical composition of the soil, pH, and temperature and several plant physiological factors. The degree of radionuclide accumulation also depends on competition between the elements present in the soil, at the uptake site or within the plant tissue. Transfer of tritium into vegetation occurs mainly through stomata on the leaf surface and uptake of soil water. It gets incorporated in organic matter within plant tissues in both exchangeable and non-exchangeable form. Soluble forms of lithium (Li) present in soils get readily absorbed by the plants.

Accumulation of radionuclides by plants is determined by the content of exchangeable and mobile forms of radionuclide. The availability of radionuclides in soil depends on several factors including pH (Ebbs et al. 1998; Echevarria et al. 2001). It is suggested that pH regulates the solubility of Rb^{+} in soils. Addition of chelators (Huang et al. 1998) and soil supplements stimulates radionuclide removal from soil (Dushenkov et al. 1999). The presence of organic matter, inorganic colloids (clay), and competing elements strongly affect the uptake of radionuclides. Radionuclides such as ^{137}Cs and ^{134}Cs get firmly bound to clay fraction of the soil. Clay strongly binds Cs and restricts the uptake by root. Addition of monovalent cations similar to Cs causes removal of ^{137}Cs from the soil (Dushenkov et al. 1999). Addition of organic matter increases the uptake of Cs by plants. Nutrient elements such as Ca^{+}, Mg^{2+} depress the uptake of Cs. The availability of Cs decreases with increasing soil moisture and percentage of fine sand and silt.

High U concentration has been noted in roots of plants raised in citric acid-treated soil (@5000 mg kg^{-1}) (Huang et al. 1998). Addition of fertilizers increases the retention of radioactive elements such as ^{137}Cs in roots. Fertilizer (potassium sulfate) treatment lowers the concentrations of ^{241}Am, ^{244}Cm, ^{232}Th, and ^{238}U. Some radioactive elements act similar to nutrients. Cesium (Cs) and rubidium (Rb) act similar to K. Similarly Sr act similar to Ca and Se is similar to S. They follow the same path as the nutrient. Uptake of radionuclide is related to essential elements such as K. The presence of potassium in the medium strongly disturbs the uptake of Cs (Bystrzejewska-Piotrowska and Urban 2003). Cesium uptake capacity decreased when 1.3 mM potassium was present in the medium (Marčiulionienė et al. 2015). The potassium concentrations of 0.5–3 mg l^{-1} cause inhibition of Cs uptake. These elements are interrelated in a complex, concentration-dependent, manner. Excessive addition of monovalent cations results in strong competition between the ions for uptake by plants. This affects the levels of radionuclide (such as ^{137}Cs) accumulation in plants (Lasat et al. 1998). The presence of potassium depresses the uptake of Cs. Rubidium and cesium follow the similar uptake route as for potassium. The size of the hydrated Rb^+ molecule is similar to that of hydrated K^+, thus the binding site at the plasma membrane of root cells cannot distinguish between the two cations. Hence, studies established that addition of potassium fertilizers decreases uptake of Cs uptake while addition of nitrogen increases uptake of ^{137}Cs by plants (Seel et al. 1995). The influx of Cs and Li into cells occurs via potassium transporters.

4.2.2 Mechanism of Radionuclide Accumulation in Plants

Radionuclides enter the plant by two ways. First is through direct deposition on the leaf surface, i.e. through foliar absorption and second is through root uptake from the soil (Fig. 4.1). The interaction of radioactive elements in the plant occurs either in the aerial portion of the plant, i.e., or in the soil-root zone of the plant (rhizosphere).

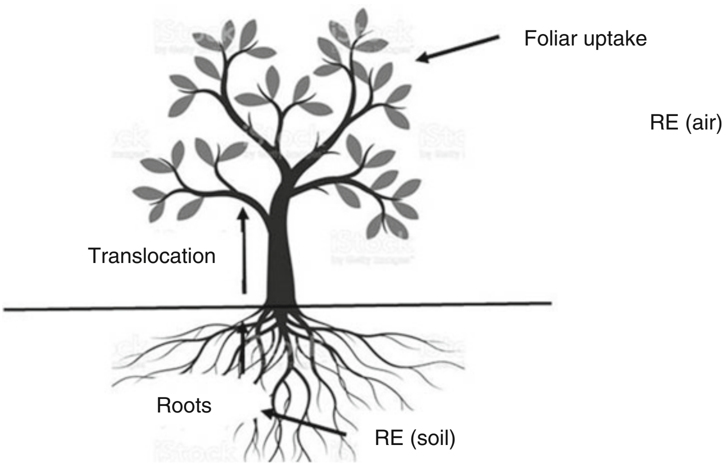

Fig. 4.1 Major routes of uptake of radioactive elements (RE)/radionuclides by plants

The root uptake has been considered as the main radionuclide transfer pathway in plants. The adsorption of radionuclides such ^{210}Pb by leaves and uptake by roots has been reported in many plant species (Chandrashekara et al. 2015). The absorption of radionuclides is followed by translocation to other plant parts particularly leaf or other organs such as stems or storage organs within the plant. Radionuclides also get distributed throughout the reproductive structures such as seeds and fruit.

Foliar uptake of radioactive particles has been commonly reported in plants. Leaf axils, leaf sheaths, grasses, and vegetation have shown accumulation of radioactive elements in the range of 0.5–5.0. After uptake, the radionuclides get transported across the cuticle and epidermis of plant leaves. The absorption of radionuclides by leaves may take place through cuticle and epidermis. The epidermal features help in retention of radioactive particles. The absorbed element get in the cuticle or cell wall of the outer cell or translocated via phloem and further to other plant parts. The phenomenon of uptake of radioactive elements is different for upper and lower leaf surfaces. The cuticle is a selectively permeable membrane, cationic, negatively charged, and hydrophobic. The cuticle is made up of waxy substances or cutin. The cuticular wax is a non-cellular and insoluble substance composed of long-chain fatty acids, alcohols, and esters. Being hydrophobic, it reduces the contact of the surface contaminant with the leaf. Fine, dense pubescence entraps radioactive particles. Hence the structural components of the cuticle and the epidermis of the leaf act as barriers to ionic movement toward the interior of the leaf. The radioactive ion cross the plasmalemma of the epidermal cell and reach the protoplasmic pool. Uptake of radionuclides through the epidermis and across the plasmalemma of the epidermal cell occurs by active and passive transport mechanism (Tagami 2012). Active transport moves the elements into the protoplasm while in passive diffusion elements move into the apparent free space (AFS) of the cell walls (Greger 2004). Active transport is associated with biosynthetic processes such as oxidative phosphorylation. Passive transport of radionuclides occurs by leaf and is affected by other cations that compete for sites on exchange compounds (Ambe et al. 1999). The radionuclide moves into the biochemical pools of the leaf.

Some radioactive elements such as ^{234}U, ^{238}U, ^{238}Pu show less mobility, hence remain adsorbed to outer layer of roots while other radioactive elements such as ^{85}Sr, ^{90}Sr, ^{137}Cs show high mobility and hence are able to enter the plant (Baeza et al. 1999). Most of the ^{137}Cs gets retained in the roots and some part (only 25%) of it gets translocated to shoots. Some elements such as^{137}Cs, Rb show accumulation in fruit and seeds. The translocation of elements into the edible part of plants depends on the element, the plant and the time between deposition and harvest. The rate of translocation varies for each species. *Calluna vulgaris* (heather) have shown high rate of translocation of ^{134}Cs from leaves to other plant parts. In contrast members of Ericaceae such as *Erica tetralix* (bell heather) and *Vaccinium myrtillus* (bilberry) showed low translocation rate. Elements such as technetium (Tc), tellurium (Te), iodine (I), and cesium (Cs) show only 10% of translocation to the grain of cereal crops (Echevarria et al. 1997). Plants differ in their ability to accumulate radionuclides. Elements such as Cs are absorbed by leaves primarily via metabolic processes linked to developmental of the plants (Carvalho et al. 2006). About 5–30%

of the Cs is absorbed by plant leaves and a substantial portion is translocated to other plant parts. Apart from this, Cs is also absorbed by roots. The ability of plant species to accumulate ^{137}Cs in the aboveground parts differs (Zhu and Smolders 2000). High level of ^{137}Cs level has been found in the wood xylem of tree trunks as well as storage roots and leaves. The accumulation of ^{137}Cs varied from 2- to 4-fold within cereals and about 27-fold in field crops (Sanzharova et al. 1997). *Amaranthus* species *A. cruentus, A. retro-flexus,* and *A. caudatus* have reported high level of ^{137}Cs accumulation in the aboveground parts (Dushenkov et al. 1999). In *Picea abies* high accumulation of ^{134}Cs has been found in roots. The sunflower has been reported to absorb 150 μg Cs in 100 h whereas a vetiver (*Vetiveria zizanioides*) absorbed 61% of ^{137}Cs in 168 h (Singh et al. 2009). Cs is adsorbed onto the cell surface. The sunflower plants showed potential to absorb radionuclides ^{134}Cs and ^{60}Co from hydroponic media. Most of the ^{134}Cs accumulated on the leaves, then inside the stem and the lowest at the root (Achmad and Hadiyanto 2018).

Strontium (Sr), barium (Ba), and radium (Ra) are the elements considered to be analogous to calcium (Ca). Calcium and Sr exist largely as immobile complexes with glutauronic acids and pectate in the plant tissue. The root uptake of Sr from soil includes mass-flow and exchange diffusion. ^{90}Sr is found to be more concentrated in leaves than in storage roots. In *Picea abies*, ^{85}Sr is predominantly accumulated in fine-roots. Accumulation of Sr in the range of 10–1500 μg DW^{-1} has been noted in plants. After foliar deposition Sr becomes moderately mobile in plants. *Vaccinium myrtillus* has been found to be a hyperaccumulator of beryllium (Be). High concentrations of Ba (barium) (up to 10,000 μg g DW^{-1}) have been reported for different trees and shrubs (Carini et al. 2016). Radium (^{226}Ra) accumulation has been found to be more in fruit than that of leaves and roots. Selective uptake of Gallium (Ga) by plants has been reported. Plants take up indium (I) and concentrations up to 100 μg g DW^{-1} has been reported from pine trees. Around 17,000 μg g DW^{-1} has been reported from the flowers of *Galium* sp.

Tellurium (Te) is rare radioactive element. Bacteria methylate Te and reduction of tellurite to Te is also influenced by micro-organisms. In onion and garlic high concentrations of Te (300 μg g DW^{-1}) have been reported. Woody seed plants can accumulate high levels of yttrium(Y) up to 700 μg g DW^{-1}. The translocation of Cerium (Ce) is very low, i.e. after foliar application and after root uptake. Higher concentration of ^{144}Ce has been found in the shoots than in roots. Ce applied to foliage is also absorbed to a lesser extent, which is probably the reason for the low translocation. Niobium (Nb) (^{95}Nb) is generally complexed with organic agents and is relative mobile and therefore available for uptake by plants. Accumulation ranging from 1 to 10 μg g DW^{-1} has been reported in plants such as *Rubus arcticus*. Vanadium (V) is easily taken up by plant roots and absorption is passive. Uptake of vanadium depends upon pH. A high pH decreases the uptake. It is more rapidly absorbed by roots as VO^{3-} and $HVO_4{}^{2-}$ species under neutral and alkaline soil solutions. Biotransformation of V from vanadate ($VO_3{}^{-}$) to vanadyl (VO^{2+}) occurs during plant uptake. Rhenium is found in anionic form as $ReO_4{}^{-}$. It is taken up by plants and concentrations up to 70–300 μg g DW^{-1} have been reported. After foliar deposition ^{183}Re shows medium mobility in the plant body. Radioactive cobalt

(^{58}Co) is easily taken up through the cuticle. Concentrations reported in terrestrial plants range from 0.4 to 200 ng g DW^{-1}. Iridium (Ir) uptake by plants has also been reported. Terrestrial plants contain concentration of 20 ng g DW^{-1} and accumulate it in the leaf margins. After foliar deposition the element has been found to immobile in the plant.

Uptake of technetium (Tc) in plants occurs in the form of TcO^{4-}. It is transported as TcO_4^- across plasma membrane into the cytosol. The Tc uptake occurs via active mechanism involving transport of TcO_4^- across the cell membrane and the reduction of Tc^{7+}. Reduction from $TcO_4^-(Tc^{7+})$ to Tc^{5+} is probably mediated by ferredoxin within the chloroplast, and up to 10 bioorganic complexes with Tc were found in leaves.

In greenhouse experiments sunflower (*Helianthus annuus*) showed U removal capacity of more than 95% from contaminated water in 24 h (Dushenkov et al. 1997; Sorochinsky et al. 1998). In pilot rhizofiltration system sunflower plants showed U accumulation of more than 1% in roots (Dushenkov et al. 1997). The sunflower, vetiver, and purple guinea grass showed ability to absorb U from water. All three plants could accumulate U in their roots. Sunflower showed the best capacity for removal of U. The non-addition of plant nutrients to the culture solution prevent competition between U and nutrient absorption. The amount of U in the plants increased with the length of the exposure period (Roongtanakiat et al. 2010). Sunflower plants also showed capacity to accumulate ^{134}Cs and ^{60}Co mostly in the leaves and roots (Achmad and Hadiyanto 2018). Dushenkov et al. (1997) reported similar results where the plants grown in a radioactive solution for long showed more radionuclides were absorption. Pilot scale studies also proved that aquatic plants possess high capacity to treat radionuclide-contaminated water. Greenhouse experiments demonstrated successful removal of Cs, Sr, and U from contaminated water (Dushenkov et al. 1997). Aquatic plants accumulates significant amounts of radionuclides depicting a high bioconcentration factor for ^{90}Sr, ^{137}Cs in case of *Cladophora glomerata,* and ^{90}Sr and ^{137}Cs for *Elodea canadensis*. *Lemna aoukikusa*, a floating vascular plant showed successful elimination of Cs and I from contaminated water. Cyanobacterium *Stigonema ocellatum* (NIES-2131) show high capacity to remove elements like Sr and I. A large number of radionuclide such as ^{3}H, U, Pu, ^{137}Cs and ^{90}Sr has been treated using plants (Negri and Hinchman 2000).

Hydroponic studies demonstrated that U uptake occurs at pH 5. At this pH, uranium occurs as uranyl (UO_2^{2+}) cation which is readily taken up and translocated by plants (Ebbs et al. 1998). Uranium forms stable uranium-phosphate complexes in roots and this prevents translocation of uranium to aboveground plant parts. In contrast, elements such as ^{90}Sr show about 80% of localization in the shoots. The cultivars of the same species show variation in accumulation of radiocesium (Cs). Rubidium (Rb) generally concentrates in flowers and young leaves. Studies have demonstrated accumulation of ^{86}Rb within reproductive structures and young tissues. *Phaseolus vulgaris* (bush bean) showed ten times higher absorption of ^{89}Sr than *Zea mays* (maize) after 72 h of treatment and two times higher than *Raphanus sativus* (radish) and *Lactuca sativa* (lettuce). Pine and aster exhibit ability to accumulate U. Grouseberry, larch, fireweed, and grass accumulate high levels of ^{226}Ra from the soil.

4.2.3 Effect of Radionuclide Accumulation on Plant Growth

The growth of plants gets affected after accumulation at high concentration of radionuclides in the plant tissues (Markovic and Stevovic 2019). Morphological changes, reduction of stem growth and root biomass are some of the changes noted in plants in response to high radionuclide accumulation. The decrease in growth rate in plants depends upon the rate of translocation from root to shoot. Uranium accumulation reduced plant biomass (ash weight) in plants. This may be due to detrimental effects of U on plant growth.

Cesium accumulation in cress, i.e. *Lepidium sativum* plants grown in hydroponic culture lead to high accumulation in leaves after both root and foliar treatments. Exposure to high concentration of cesium (3 mM) affected water uptake, tissue hydration (FW/DW), and production of biomass (DW). The gas exchange parameters such as stomatal conductance (C) and transpiration rate (E) also showed strong inhibition while the rate of photosynthesis did not get altered significantly. Changes in photochemistry of photosystem II (PSII) related to the alteration in photosynthetic potential. Decreased stomatal opening affected the rate of transpiration and uptake of water. The decrease in tissue hydration decreases photosynthetic CO_2 assimilation, synthesis of organic matter, and affects light reactions of photosynthesis (Bystrzejewska-Piotrowska and Urban 2003).

A greenhouse experiment showed that growth attributes such as relative growth rate, net assimilation rate, leaf are index, specific leaf area, dry matter allocation and production of reproductive organs showed a decrease as the radionuclide content in the plant increased. The decrease has been noted in plants such as *Cakile maritima, Senecio glaucus* and *Rumex pictus* at different stages of growth (seedling, juvenile, flowering, fruiting and senescing). High level of radionuclide accumulation in these plants affected dry matter allocation and root to shoot weight ratio (Hegazy et al. 2011).

4.3 Conclusions

Plants possess capacity to treat radioactive particles. The removal of radionuclides in plants occurs via adsorption by leaves or absorption by roots. The uptake and removal of radionuclides from the environment is regulated by various physical and environmental factors. The radionuclide removal capacity also varies for each plant species depending upon its affinity for taking up radioactive elements followed by translocation and/or accumulation in the plant tissues. The radionuclide accumulation capacity of the plants can be exploited for developing large scale technologies for treatment of water and soil contaminated with radioactive waste provided that we have a better understanding of the physiological and molecular mechanisms related to radionuclide accumulation in plants.

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