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Review of Phytoremediation Technologies for Radiological Contamination

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U.S. Environmental Protection Agency

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Foreword

The U.S. Environmental Protection Agency (EPA) is charged by Congress with protecting the nation's land, air, and water resources. Under a mandate of national environmental laws, the Agency strives to formulate and implement actions leading to a compatible balance between human activities and the ability of natural systems to support and nurture life. To meet this mandate, EPA's research program is providing data and technical support for solving environmental problems today and building a science knowledge base necessary to manage our ecological resources wisely, understand how pollutants affect our health, and prevent or reduce environmental risks in the future.

The EPA’s Center for Environmental Solutions and Emergency Response (CESER) within the Office of Research and Development (ORD) conducts applied, stakeholder-driven research and provides responsive technical support to help solve the nation’s environmental challenges. The Center’s research focuses on innovative approaches to address environmental challenges associated with the built environment. We develop technologies and decision-support tools to help safeguard public water systems and groundwater, guide sustainable materials management, remediate sites from traditional contamination sources and emerging environmental stressors, and address potential threats from terrorism and natural disasters. CESER collaborates with both public and private sector partners to foster technologies that improve the effectiveness and reduce the cost of compliance, while anticipating emerging problems. We provide technical support to EPA regions and programs, states, tribal nations, and federal partners, and serve as the interagency liaison for EPA in homeland security research and technology. The Center is a leader in providing scientific solutions to protect human health and the environment.

This study report identified key documents in regard to practical experiences in field-deployed phytoremediation efforts in the U.S., the former Soviet Union and Japan, with considerations to site preparation and maintenance, remediation effectiveness, and waste management. Recommendations are provided for candidate plant species based on the literature review, and technical gaps are identified.

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Executive Summary

Phytoremediation is a set of technologies that use various plants and microbes to degrade, extract, contain, or immobilize contaminants from soil and water. Phytoremediation is an integrated approach applied to the cleanup of contaminated soil that combines the disciplines of plant physiology, soil chemistry, and soil microbiology. It offers a viable method for stabilizing and removing contamination at significantly less cost than alternatives such as excavation or pump-and-treat methods. Initially deployed to address organic and heavy metal contamination of soil and groundwater, the application of phytoremediation has expanded over recent years to include mitigation of radionuclide contamination. A large volume of literature exists evaluating the feasibility of plant species to effectively remove actinide (e.g., uranium, plutonium, neptunium) and fission product radiological contamination in the environment. Key documents are identified here in regard to practical experiences in field-deployed phytoremediation efforts in the U.S., the former Soviet Union and Japan, with considerations to site preparation and maintenance, remediation effectiveness, and waste management. Recommendations are provided for candidate plant species based on the literature review, and technical gaps in the current knowledge base are identified.

Acronyms

Am-241	americium-241
ANOVA	analysis of variance
ANL	Argonne National Laboratory
BAF	bioaccumulation factor
CR	concentration ratio
Cs-134	cesium-134
Cs-137	cesium-137
DHS	U.S. Department of Homeland Security
DOE	U.S. Department of Energy
EPA	U.S. Environmental Protection Agency
H-3	tritium
IAEA	International Atomic Energy Agency
INL	Idaho National Laboratory
ITRC	Interstate Technology & Regulatory Council
JAEA	Japanese Atomic Energy Agency
LLRW	low-level waste
Np-237	neptunium-237
NPP	nuclear power plant
PPE	personal protective equipment
Pu-238	plutonium-238
RDD	radiological dispersal device
Sr-90	strontium-90
SRS	Savannah River Site
U-238	uranium-238
USDA	United States Department of Agriculture

Units

Bq	Becquerel(s) (one disintegration per second, equivalent to 2.7 × 10^{-11} Curies)
Ci	Curie(s) (equivalent to 3.7 × 10^{10} Becquerels)
g	gram(s)
ha	Hectare(s) (1 hectare = 2.471 acres)
m	meter(s)
L	liter(s)

Unit Prefixes

c	centi (10^{-2})
k	kilo (10^{3})
m	milli (10^{-3})
M	mega (10^{6})
p	pico (10^{-12})
P	Peta (10^{15})

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1. Introduction

Radiological contamination stemming from nuclear facility accidents or intentional releases such as a radiological dispersal device (RDD) can lead to large areas of urban and/or rural land requiring decontamination. A broad range of expertise and capabilities have been tried and tested for mitigating the effects of radiological contamination, particularly in the remediation and recovery phases following nuclear power plant accidents such as Chernobyl and Fukushima, but also including nuclear material handling and disposal sites where historical contamination existed. Various techniques and technologies may be employed to remove both surface deposited and entrained contamination. Decontamination can be non-destructive (e.g., washing) or destructive (e.g., surface removal and subsequent disposal) to surface, depending on the contaminant and surface. Contamination events can lead to atmospheric, surface and/or subsurface contamination, each of which can cause subsequent migration of contaminants into another medium. A variety of techniques can also be used to bind (or stabilize) contaminants in place, preventing both resuspension and further increase in impacted areas.

One remediation method that employs both stabilization and subsequent removal (decontamination) is *phytoremediation*. Specifically, phytoremediation is a set of technologies that use various plants and microbes to degrade, extract, contain, or immobilize contaminants from soil and water. It is an integrated approach applied to the cleanup of contaminated soil that combines the disciplines of plant physiology, soil chemistry, and soil microbiology (Hossner et al., 1998). Depending on the nature of the contaminant soil and plant pathways and properties, there are different approaches to using plants in the remediation of environmental contaminants. The United States Environmental Protection Agency's (EPA's) *Phytoremediation Resource Guide* (U.S. EPA, 1999) provides an overview of the various approaches:

> **Phytoextraction**
> Also called phytoaccumulation, phytoextraction refers to the uptake and translocation of metal contaminants in the soil by plant roots into the aboveground portions of the plants. Certain plants called *hyperaccumulators* absorb unusually large amounts of metals in comparison to other plants. One or a combination of these plants is selected and planted at a site based on the type of metals present and other site conditions. After the plants have been allowed to grow for several weeks or months, they are harvested and either incinerated or composted to recycle the collected contaminants such as metals. This procedure may be repeated as necessary to bring soil contaminant levels down to targeted limits.
>
> **Rhizofiltration**
> Rhizofiltration is the adsorption or precipitation onto plant roots, or absorption into the roots, of contaminants that are in solution surrounding the root zone. The plants to be used for cleanup are raised in greenhouses with their roots in water rather than in soil. To acclimate the plants once a large root system has been developed, contaminated water is collected from a waste site and brought to the plants where it is substituted for their water source. The plants are then planted in the contaminated area where the roots take up the water and the contaminants along with it. As the roots become saturated with contaminants, they are harvested and either incinerated or composted to recycle the contaminants.

Phytostabilization
Phytostabilization is the use of vegetation to contain soil contaminants in situ, through modification of the chemical, biological, and physical conditions in the soil. Contaminant transport in soil, sediments, or sludges can be reduced through absorption and accumulation by roots; adsorption onto roots; precipitation, complexation, or metal valence reduction in soil within the root zone; or binding into humic (organic) matter through the process of humification. This process reduces the mobility of the contaminant and inhibits migration to the ground water or air, and it reduces bioavailability for entry into the food chain. This technique can be used to reestablish a vegetative cover at sites where natural vegetation is lacking due to high metal concentrations in surface soil or physical disturbances to surficial materials. Metal-tolerant species can be used to restore vegetation to the sites, thereby decreasing the potential migration of contamination through wind erosion, transport of exposed surface soil, and leaching of soil contamination to ground water.

Phytodegradation
Also called phytotransformation, phytodegradation is the breakdown of contaminants taken up by plants through metabolic processes within the plant, or the breakdown of contaminants external to the plant through the effect of compounds (such as enzymes) produced by the plants. Pollutants are degraded, incorporated into the plant tissues, and used as nutrients.

Rhizodegradation
Also called enhanced rhizosphere biodegradation, phytostimulation, or plant-assisted bioremediation/degradation, rhizodegradation is the breakdown of contaminants in the soil through microbial activity that is enhanced by the presence of the rhizosphere and is a much slower process than phytodegradation. Microorganisms (yeast, fungi, or bacteria) consume and digest organic substances for nutrition and energy. Certain microorganisms can digest organic substances such as fuels or solvents that are hazardous to humans and break them down into harmless products through biodegradation. Natural substances released by the plant roots—sugars, alcohols, and acids—contain organic carbon that provides food for soil microorganisms, and the additional nutrients enhance microbial activity. Biodegradation is also aided by the way plants loosen the soil and transport water to the contaminated area.

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

Since radionuclide contamination cannot be *chemically* altered into a less radioactive material, techniques such as phytodegradation, rhizodegradation and phytovolatilization are not applicable to radionuclide remediation. However, phytoextraction, rhizofiltration and phytostabilization are methods that can be employed to stabilize and subsequently remove contamination from both soil and ground/surface water.

Once radioactive contamination is deposited on soil, its migration downward is governed by convective transport by flowing water, dispersion caused by spatial variations of convection velocities, diffusive movement within the fluid, and physicochemical interactions with the soil matrix (IAEA, 2010). The former three governing actions can be generally described by traditional convection and dispersion time-dependent models. The last action can be generally described by equilibrium processes occurring at the interface of the soil, soil water and plant. The contaminant must be soluble to interact with soil or sediment. Some contaminants are considered fixed in the soil strata and subjected to move with soil such as erosion or resuspension phenomena and to be removed mechanically for remediation. These fixed contaminants might not be directly treatable by phytoremediation depending on their binding status in soil. The interaction between contaminant and soil can be described by the equilibrium *distribution* (or adsorption) *coefficient* (K_d), which is dependent on the physical and chemical properties of the soil, liquid and contaminant, as shown by Equation 1.

$$K_d = \frac{Concentration\ of\ Contaminant\ in\ Soil}{Concentration\ of Contaminant\ in\ Soil\ Water} \qquad \text{Eq. 1}$$

The subsequent uptake by plant roots embedded in soil must occur through exchange of the contaminant from soil to water. This relationship is controlled by the selectivity of the plant root system, represented by the radionuclide plant to soil solution ratio (radionuclide activity, mass or concentration per kilogram of dry weight plant tissue versus the contaminant activity, mass or concentration per liter of soil solution), and referred to as the *bioaccumulation factor* (BAF), as depicted by Equation 2.

$$BAF = \frac{Concentration\ of\ Contaminant\ in\ Plant}{Concentration\ of\ Contaminant\ in\ Soil\ Water} \qquad \text{Eq. 2}$$

The reactions between contaminant, soil, water and plant tissue are in competition during equilibrium, characterized by the *concentration ratio* (CR) as defined in Equation 3.

$$CR = \frac{BAF}{K_d} \qquad \text{Eq. 3}$$

The reverse process can also occur, with desorption from either soil or plant material to the soil solution. Thus, in broad terms, phytoremediation effectiveness can be estimated based on known values for the physicochemical properties of the soil, plant and contaminant. In this work, we focus mostly on the equilibrium of radioactive contamination between soil, water and plant, with the equilibrium processes schematically represented in Figure 1.

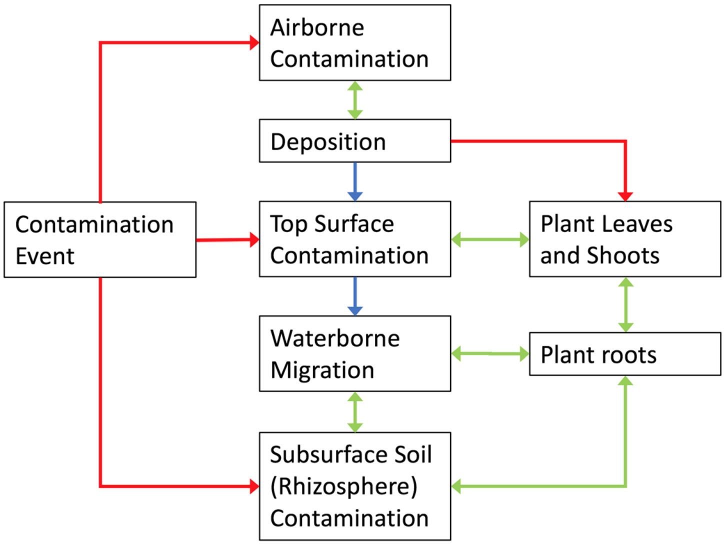

Figure 1. Schematic figure showing contaminant deposition and migration paths

A summary of the current state of radiological phytoremediation, providing general references and prior literature reviews, pilot- and field- scale studies performed at contaminated sites in the United States, former Soviet Union, and Japan and review of data relevant to the phytoremediation of radiocesium (e.g., Cs-134 and Cs-137) is given in Section 2. Similar datasets exist for plant uptake and phytoremediation of actinides (e.g., uranium, plutonium, neptunium) and other fission products (strontium, iodine, cerium, niobium, zirconium, etc.). An assessment of considerations and metrics for comparing available technologies with respect to cost, application time, labor/expertise and supporting equipment needs, waste generation and technical performance is given in Section 3, while recommendations and technical gaps in the knowledge base are presented in Section 4.

1.1 Quality Assurance

The data in the reviews and evaluations met the data quality objectives by collecting them from a combination of published, peer-reviewed journal articles, government reports (e.g., EPA, DOE, DOD, UK Government, EU, Japanese Atomic Energy Agency [JAEA], International Atomic Energy Agency [IAEA]) and industry/vendor information. By nature of their review by peers, journal articles and some conference abstracts are considered trusted sources of information. Similarly, reports published by government agencies such as EPA, U.S. Department of Energy (DOE), U.S. Department of Homeland Security, and the Interstate Technology Regulatory Council (ITRC) are considered highly trustworthy. International governmental reports were also utilized, including those from the UK and European Union (EU) as well as the JAEA and IAEA, particularly the reports relating to the response following the Fukushima and Chernobyl contamination events.

2. Radionuclide Phytoremediation Literature Survey

Laboratory and field studies involving radionuclide uptake in soil and plants have been performed for many years, beginning with understanding the impacts of nuclear testing on the environment and food (e.g., Nishita, Steen and Larson, 1958; Middleton, 1959), which included both actinide and fission product contamination. Plant uptake of radionuclide contamination gained additional interest after the 1986 Chernobyl nuclear power plant accident, in which radioactive contamination was spread across parts of the former Soviet Union and Europe. An IAEA (1991) assessment of radiological consequences and evaluation of protective measures following the release of radionuclides from the Chernobyl plant provides information on protective measures and restrictions on food (including plants). In 1994, IAEA published a handbook on parameters for the prediction of radionuclide transfer to plants, animals, marine estuaries and food in temperate environments (IAEA, 1994). Similar to previously published work, the IAEA document includes information on uptake of actinides and fission products, but there is no mention in the report of the data being used for phytoremediation. It was not until the mid 1990's that phytoremediation (which had previously been applied to cleanup of heavy metal and organic solvent spills) was applied to radionuclide remediation.

2.1 General Published Guidance on Radionuclide Phytoremediation

Schnoor (1997) provided a technology evaluation report to the Ground-Water Remediation Technologies Analysis Center detailing information on the use of innovative technologies to clean up contaminated groundwater. In the 1997 report, Schnoor describes phytoremediation as an emerging technology for contaminated sites that is attractive due to its low cost and versatility, showing tremendous potential for treatment of shallow metal and organic contaminants. Only two applied examples were given at that time for radionuclide application of phytoremediation, specifically Cs-137 and Sr-90 pond water cleanup in the Ukraine using sunflowers resulting in a 90% reduction in two weeks, and a demonstration in Ohio also using sunflowers resulting in a 95% removal of uranium from waste water in 24 hours. Dushenkov (PNNL, 1998) further reported that the Ohio study using sunflower-based pilot-scale rhizofiltration system was successfully used at a former uranium processing facility, lowering uranium concentrations in the site source water to below the target limit of 20 micrograms per liter. In these applications, specific isotopes are not processed differently with phytoremediation, but as long as it is chemically the same (for example, be it Cs-134 or Cs-137), the isotopes are expected to be extracted with the same efficiency. Additional information on applied phytoremediation is given in Section 2.3 of this work.

Proceedings from a 1998 international workshop in Ukraine on Chernobyl phytoremediation and biomass energy conservation were published as a Pacific Northwest National Laboratory report (PNNL, 1998). The report summarizes that preliminary tests of phytoremediation systems had been established in highly contaminated areas of the Chernobyl exclusion zone, but that defining objectives, implementing the technology and evaluating the various options would mean optimum phytoremediation options would take at least an additional 5 years.

In addition to a significant number of peer-reviewed and published journal articles and early technology reviews on the subject of radionuclide phytoremediation over the last 20 years, several high-quality literature reviews have been performed that capture both the uptake of contamination in plants, and the state of phytoremediation research.

A prior literature review of phytoaccumulation of chromium, uranium and plutonium in plant systems was reported in 1998 (Hossner et al.), which summarized the state of phytoremediation science and provided examples of radioisotope phytoremediation applications. The EPA published a phytoremediation resource guide (U.S. EPA, 1999) providing references to journal articles, technical documents and websites that reported phytoremediation information for soil and ground water. The report largely focused on organic solvent and heavy metal contamination, with limited resources on radionuclide contamination beyond that of Hossner et al. (1998) and Broadley and Willey (1997) who focused on cesium uptake in 30 different plants. In 2000, EPA convened subject matter experts to discuss experimental and applied phytoremediation for solvents, heavy metals and radionuclides (U.S. EPA, 2001). In 2005, NATO hosted a conference on advanced science and technology for biological decontamination of sites affected by chemical and radiological nuclear agents. The proceedings were subsequently published in Marmiroli, Samotokin, and Marmiroli (2007), which included a paper authored by Soudek (2007) that discussed cesium, strontium, iodine and radium uptake in plants.

In 2009, the Interstate Technology & Regulatory Council published their third technical and regulatory guidance document on phytotechnology (ITRC, 2009). In addition to providing comments and phytoremediation effectiveness for a variety of plants and radionuclides (including cerium-144, cesium-134/137, cobalt-58, radium-224/226, ruthenium-106, strontium-90, technetium-99 and uranium-238), the ITRC report also includes valuable information on applied remediation project structure, site assessment, remediation strategy, plant selection, design and implementation, operation, maintenance, monitoring, and site closure. While the ITRC report was published in 2009, tabulated references related to radiological phytoremediation are limited to those before 1998, presumably since the 2009 third edition combines prior ITRC phytoremediation documents (ITRC, 1999; ITRC, 2001)[1]. However, ITRC (2009) remains one of the most concise and comprehensive sources for phytoremediation planning purposes.

IAEA (2009) supplemented their 1994 handbook with a report on the quantification of radionuclide transfer in terrestrial and freshwater environment for radiological assessment. With a much larger dataset, this report represented a significant improvement in peer reviewed literature data on the plant uptake of radionuclides. Also, in contrast to IAEA (1994), references cited in IAEA (2009) include those that specifically evaluated radionuclide phytoremediation (e.g., Vandenhove, Van Hees and Van Winckel, 2001; and Fuhrmann et al., 2002). The 1994 IAEA handbook was subsequently updated (IAEA, 2010) with the purpose of providing a very extensive dataset, which encompass data from studies evaluating radionuclide uptake in plants for both radiation protection and phytoremediation studies.

In 2012, IAEA provided guidelines for remediation strategies to reduce the radiological consequences of environmental contamination. On the subject of phytoremediation, IAEA (2012) stated that “until now, there has been no small- or large- scale adoption of this method at existing sites for radionuclides. There are three main reasons why this option has not been adopted: (i) the total amount of radionuclide removed from the soil is a very small fraction of the

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[1] ITRC https://www.itrcweb.org/Guidance/ListDocuments?TopicID=20&SubTopicID=30 (accessed July 15th 2019)

total radionuclide content present, even for those radionuclides with a comparatively high transfer from soil to plant; (ii) the process would need to be continued for decades before the soil became adequately decontaminated to be used for food production; and (iii) the option generates waste which would then have to be disposed of appropriately, generating additional costs." IAEA (2012) did not provide additional quantitative information on phytoremediation effectiveness and plant uptake compared to IAEA (2010), which remains one of the most comprehensive, quantitative and peer-reviewed broad data sets for radionuclide uptake in plants.

2.2 Literature Review of Radiocesium Phytoremediation

There are numerous examples of laboratory-scale cesium phytoremediation studies on a wide variety of plants and soils; several examples (not intended to be an exhaustive list) are provided in this section. Additional references and observations on cesium phytoremediation can be found in ITRC (2009). Published works by Entry studied the uptake of Cs-137 and Sr-90 in ponderosa pine and Monterey pine (Entry, Rygiewicz and Emmingham, 1993, Adriano et al., 1995), as well as in Alamo switchgrass (Entry and Watrud, 1998). Broadley and Willey (1997) studied Cs-137 uptake in the shoots of 30 plants, noting that there were maximum differences between quinoa and junegrass of 20-fold in cesium concentration and l00-fold in total cesium accumulated.

In the late 1990's, work was performed at the USDA-ARS [Agricultural Research Service] Plant, Soil and Nutrition Laboratory (Ithaca, New York) in support of a phytoremediation concept later to be deployed on contaminated land at the Brookhaven National Laboratory (Upton, New York). Lasat, Norvell and Kochian (1997) studied Cs-137 uptake on contaminated soil using Indian mustard, arcadia, a commercial variety of broccoli, cabbage and cauliflower, kochia, tepary bean, hairy vetch, colonial bentgrass, red fescue and reed canary grass. The results suggested that phytoremediation of Cs-137 contaminated soil was feasible. Later, Lasat et al. (1998) also studied uptake in root pigweed and continued studies with Indian mustard and tepary bean, finding that redroot pigweed is a plant with high potential for extraction of Cs-137 from contaminated soils. Lasat et al. (1998) also evaluated the impact of ammonium nitrate fertilizer on the uptake of cesium in Indian mustard, redroot pigweed and tepary bean, finding that the ammonium ion has the potential to desorb Cs-137 ions from the soil minerals, but did not enhance Cs-137 bioaccumulation perhaps because of competition for binding sites. Work on contaminated soil from the Brookhaven National Laboratory continued to include redroot pigweed (Fuhrmann et al., 2002) and Powell's amaranth (Fuhrmann, 2006).

Sandeep and Manjaiah (2007) evaluated the transfer factors of Cs-134 in mustard, gram, spinach and wheat crops in semi-arid, tropical climate, finding that transfer factors were highest in spinach, with mustard and gram much higher than that of wheat. Sadhasivam, Pitchamuthu and Ayyavu (2010) studied Cs-137 phytoextraction in the presence of ammonium chloride fertilizer using amaranthus, maize, cowpea and sunflower, finding that amaranthus bioaccumulation was superior. Djedidi et al. (2014) studied both the uptake of stable cesium in komatsuna, amaranth, sorghum, common millet and buckwheat, together with the effect of inoculation with *Bacillus* and *Azospirillum* on komatsuna, which resulted in the greatest transfer factors, suggesting that the addition of bacteria can enhance bioaccumulation. Dan et al. (2015) studied phytoextraction of stable cesium using *Amaranthus mangostanus* L.*,* finding that the chlorophyll content in the amaranth initially increased and then decreased with the increasing cesium content in the soil. The results suggest overloading (leading to a decrease in uptake) can negatively impact plant photosynthesis and hinder plant growth. Fukuda et al. (2014) performed a search of 188 strains of algae and aquatic plants that could eliminate cesium, strontium and iodine from contaminated water, finding that the algae *Eustigmatophyceae, Florideophyceae and Chlorophyta* and the plant *Tracheophyta* yielded the greatest ability for removing Cs-137 contamination from freshwater within 8 days of contact.

The inclusion of fermented bark amendment was found to accelerate cesium uptake in rice plants (Sun et al., 2019), possibly due to a reduction in the oxidation potential in soil. The addition of ammonium sulfate fertilizer further increased the bioaccumulation factor in rice straw. Sun notes that increased cesium availability was due to exchange with ammonium ions within the soil. Thus, the application of both fermented bark and fertilizer should be considered in phytoremediation strategies. Statistical approaches have also been used to evaluate bioaccumulation of contaminants. Willey, Tang and Watt (2005) used analysis of variance (ANOVA) and residual maximum likelihood to predict inter-taxa differences in Cs-134 and Cs-137 plant uptake. In general, their findings were that Eudicots, and especially the Caryophyllales, Asterales, and Brassicales, had high cesium uptake concentrations, while the Fabales and Magnoliid*s*, in particular Poales, had low cesium uptake concentrations, noting that plant phylogeny and growth strategy might thus be used to predict a significant portion of inter-taxa differences in cesium plant uptake. Such an approach may be extremely helpful in determining plant species for phytoremediation studies when considering plants that will both thrive and behave as radionuclide hyperaccumulators.

2.3 Applied Radiological Phytoremediation: Pilot- and Full- Scale Studies

2.3.1 U.S. DOE Sites

In the mid 1970's, dredging of the contaminated Interceptor-Canal at the Argonne National Laboratory (ANL) West (Idaho Falls, Idaho) site resulted in a mound of soil contaminated with 30.53 pCi/g of Cs-137. DOE evaluated 5 options for remediation, including (i) no action, (ii) limited action, (iii) containment with institutional controls, (iv) excavation/disposal, and (v) phytoremediation. Subsequently, DOE submitted a proposal to utilize phytoremediation to reduce Cs-137 contamination in the Canal-Mound. A pilot/field-scale effort began in 1999 with the goal of reducing Cs-137 contamination to 23.3 pCi/g (U.S. EPA, 1998). Through planting of *Kochia scoparia* over 0.61 acres, the project was completed in 2002 after Cs-137 levels fell to 6.54 pCi/g (U.S. EPA, 2005). Irrigation, fertilization, pest control and harvesting were required during the project duration. Harvested plants were sampled, compacted and shipped for incineration to reduce waste volume before being sent to a permitted landfill (U.S. EPA, 1998). In 1998 USD, the phytoremediation cost estimate was $3M, compared to $9.0M for containment, $6.6M for excavation and INL disposal, and $13.4M for excavation and private facility disposal (U.S. EPA, 1998).

ANL East (Lemont, Illinois) began full-scale phytoremediation efforts on four acres of their Area 317/319 site in 1999 to treat soil and groundwater contaminated with a mixture of chlorinated hydrocarbons, heavy metals and tritium (U.S. EPA, 2005) using a combined approach of hydraulic control, phytoextraction, phytostabilization, rhizodegradation and phytodegradation with a variety of plants including hybrid poplar, eastern gamagrass, golden weeping willow, hybrid prairie cascade willow and laurel-leaved willow. The full-scale efforts required fertilization, replanting and significant health and safety aspects (due to hazardous and radioactive material concerns) increased cost and difficulty in application.

A phytoremediation investigation at DOE's Savannah River Site (SRS) published by Murphy and Tuckfield (1994) describes a study of transuranic (americium, neptunium and plutonium) contaminant uptake in native loblolly pine, sweet gum, and willow oak planted above the unlined Low-Level Burial Ground in 1978 with a goal of returning the site to general public access. The study, which evaluated contaminant uptake in the tree seedlings, grown trees, leaves and needles over the subsequent years, found that there was more Pu-238 uptake by pine tree seedlings compared to sweet gum and willow oak. Additionally, the study found that transuranic uptake in grown pine trees occurred to a greater extent than in seedlings, and that transuranic uptake in pines was higher on the Low-Level Burial Ground compared to a control plot. The study therefore indirectly demonstrated the feasibility of native plants as phytoremediation accumulators, with uptake activities of 0.088 Bq/kg for Am-241 (transport of 320 Bq/ha/year), 0.48 Bq/kg for Np-237 (transport of 1,900 Bq/ha/year), 0.79 Bq/kg for Pu-238 (transport of 2,800 Bq/ha/year), and 0.09 Bq/kg for Pu-239 and Pu-240 (transport of 240 Bq/ha/year). Murphy and Tuckfield (1994) also projected transuranic uptake in food grown in soil after 100 years of phytoremediation using barley, pea, bean, soybean, wheat and tomato.

In 2000, full-scale phytoremediation efforts began at the SRS Radioactive Waste Burial Ground Complex with tritium contaminated groundwater at approximately 500 pCi/mL. Naturally forested areas of sweet gum, loblolly pine, slash pine and laurel oak were utilized approximately 25 acres with a combination of hydraulic control and enhanced evapotranspiration. Blount et al. (2003) describes the process as one that dilutes tritium concentrations while absorbing approximately 60% of tritium (H-3) in biomass, exchanging the remaining approximately 40% in hydrogen atoms in water molecules, and evaporation of tritium-containing water in the plant system to the atmosphere. In addition, a dam was constructed to contain surface discharge of the tritium contaminated groundwater. Blount's calculations predicted a maximum fixed activity of 30 pCi/g occurs in less than 10 years, with fixed activity reaching the federal Primary Drinking Water Standard of 20 pCi/mL if the tree is harvested 15 years after irrigation began. As of 2004, approximately 133 million liters of irrigation has prevented 1,800 Ci H-3 from entering a nearby river, with levels between 2004 and 2014 typically below 100 pCi/mL (Hitchcock et al., 2005). No cost information was available.

In 2006, laboratory-, greenhouse- and pilot-scale phytoremediation studies were initiated to address groundwater at DOE's Hanford site (Benton County, Washington) with approximately 3,000 Ci Sr-90. Groundwater contamination of approximately 8 pCi/mL in the 100-N Area and along the 100-N Area Columbia River shoreline originated from two liquid waste disposal facilities operated from 1963 to 1991. This effort used native coyote willow stems that were subsequently harvested by cutting at 10 to 20 cm above the ground twice a year and monitoring was performed on a monthly basis. The studies indicate that coyote willow could function as a successful phytoextractant of Sr-90 (Ainsworth, 2006; Fellows, Fruchter and Driver, 2009; and Fellows et al., 2010). Fellows et al. (2010) estimated that a total of 7.7 metric tons of biomass would yield a removal rate of about 16 mCi of Sr-90 per year, with an initial specific activity of 2,100 pCi/g of biomass. The initial project costs were estimated at $433,000 (Ainsworth, 2006).

2.3.2 Post-Chernobyl Applied Phytoremediation

The Chernobyl nuclear power plant (NPP) accident in the Ukraine released radionuclides with a total activity estimated at 1,900 PBq (50 million Curies), including approximately 19 PBq Cs-134 and 38 PBq Cs-137 (IAEA, 1991) in 1986. Several phytoremediation studies have been performed, ranging from the contaminated exclusion zone to other countries that experienced land contamination as a result of the plume. Sorochinsky (PNNL, 1998) reported results from laboratory and field studies performed in 1995 and 1996 using sunflower, Indian mustard and pea plants within the Chernobyl exclusion zone, finding that all three plants were successful in rhizofiltration studies to remove both Cs-137 and Sr-90. Sorochinsky also suggested that the technology of short-rotation forestry may be successfully introduced in the Chernobyl exclusion zone, for example fast-growth clones of poplar and willow, which typically have a rotation every 6 to 7 years and have a high density of 1,000 plants/ha, producing between 10,000 to 20,000 kg of dry biomass/ha/year. Dushenkov et al. (1999) studied phytoremediation of Cs-137 in 20 plant species installed on a loam/sand soil experimental plot of heavily contaminated land at the northwest boarder of Chernobyl, finding that several species of amaranth possessed promising bioaccumulation coefficients, with bioaccumulation coefficients ranging from 0.53 to 2.03. Victorova et al. (2000) performed Cs-137 and Sr-90 phytoremediation studies on the west bank of the river Pripyat and in Yanov (Ukraine), in sandy contaminated soil using natural willow, resulting in radiocesium transfer factors ranged from 10^{-4} and 10^{-3} m^2/kg. Highest uptake was observed in plants that were 7 to 8 years old as compared to younger 1 to 2-year-old plants.

IAEA (2012) noted that until recently, no small- or large-scale adoption of phytoremediation had been successful at existing contaminated sites in the former Soviet Union. IAEA cited three reasons for a lack of phytoremediation adoption to address contaminated land, including the relatively small fraction of total radionuclides removed from soil in comparison to the high values in soil, the long-term nature of phytoremediation given the scale of contaminated land, and the biomass disposal options and costs. Paramonova et al. (2015) studied the root uptake of spring barley, maize, summer rape, galega, potatoes and amaranth in ecosystems of dry and wet meadows in the Tula region, Russia. The findings showed that galega and amaranth could be considered for phytoremediation, since 87-93% of Cs-137 inventory is located in shoots. However, meadow grasses and cereals appeared to not be feasible phytoremediation species due to 86-97% of the contamination being associated with roots remaining in soil after removal of shoots.

2.3.3 Post-Fukushima Applied Phytoremediation

The March 2011 Fukushima Daiichi NPP accident resulted in radionuclide releases on the order of 10,000 PBq (270 MCi), with between 8 and 50 PBq (~200 kCi to 1,300 kCi) of Cs-134 and 7 to 20 PBq (~190 kCi to 540 kCi) of Cs-137 (IAEA 2015). For two seasons in 2011 and 2012, Terashima, Shiyomi and Fukuda (2014) examined Cs-134 and Cs-137 levels in grassland 32 km northwest of the Fukushima Daiichi NPP, including timothy, orchard grass, perennial ryegrass and clovers as well as soil. The results showed that the level of radiocesium in the biomass increased over the two years, with 97% of the contamination present in the top 5 cm of soil, but that concentration ratios were lower in 2012 compared to the initial spike in 2011. Yamashita et al. (2014) estimated the soil-to-plant transfer factors of radiocesium in 99 wild plant species grown in contaminated arable lands in the Fukushima Prefecture one year after the Fukushima NPP accident. Some species (e.g., *Athyrium yokoscense*, *Dryopteris tokyoensis*, and *Cyperus brevifolius*) exhibited relatively high concentration ratios, while others (e.g., *Salix miyabeana*, *Humulus scandens*, and *Elymus tsukushiensis*) exhibited almost negligible values. Yamashita concluded that the weed community is not a practical candidate for phytoremediation techniques.

Sugiura et al. (2016) evaluated Cs-134 and Cs-137 in new leaves of wild plants two years after the Fukushima Daiichi NPP accident. The results showed that woody plants exhibited high concentration ratios resulting from deposited Cs-137 on above-ground portions of plants transferred to new plant tissue. Additionally, Sugiura found that concentration ratios in 2012 decreased compared to those measured in 2011, with concentration ratios ranging from 1.3 to 54 for herbaceous species and 3.6 to 30 for woody species during the first year. Interestingly, Sugiura also notes that species previously identified in other studies as being cesium hyperaccumulators in this case showed no clean Cs-137 accumulation ability. Rather, the perennial plant *Houttuynia cordata* (chameleon plant) and deciduous trees *Chengiopanax sciadophylloides* and *Acer crataegifolium* displayed high concentration ratio (CR) values, which may be considered better options for phytoremediation. In laboratory experiments, Tamaoki et al. (2016) evaluated amaranthus, chamomile, cherry sage, cockscomb, hollyhock, ice plant, Indian spinach, kochia, okra, reed, rumex, salvia, scarlet rose mallow, sunchoke, sunflower and tomato cultivated in soil removed from contaminated land as a result of the Fukushima Daiichi NPP accident. The results showed that the plant biomass is a significant contributor to the uptake of Cs-137 and that kochia showed the most favorable results for phytoremediation applications.

Fifty-six local Japanese cultivars of field mustard*,* Indian mustard and rapeseed were assessed by Djedidi et al. (2016) for variability in growth and Cs-137 uptake and accumulation in association with a *Bacillus pumilus* strain, applying research from Djedidi et al. (2014) to field studies on contaminated farmland in Nihonmatsu city, in Fukushima prefecture. *B. pumilus* induced a significant increase in shoot dry weight in 12 cultivars that reached up to 40% in one field mustard and three Indian mustard cultivars. Soil to plant Cs-137 transfer ratios varied by a factor of 5 depending on species and inoculation. Aung et al. (2016) measured Cs-137 uptake plants and soils from contaminated areas in Japan, also showing that inoculation with *Bacillus pumilus* led to an increase in root volume and a subsequent increase in the transfer of Cs-137 from soil to plant.

3. Technical Considerations for Radiological Phytoremediation

The use of phytoremediation in the removal of radiological contamination requires consideration from subject matter experts and stakeholders at each stage, from species selection, planting and ground preparation to maintenance, removal and waste disposal. Dushenkov (PNNL, 1998) identified the key factors determining the effectiveness of phytoextraction:

- Plant characteristics
- Planting density
- Fertilizer and amendment effectiveness
- Soil characteristics including type, pH, electrical conductivity and mineralogy
- Climate
- Total soil radionuclide concentration
- Metal solubility and species distribution

For rhizofiltration, Dushenkov (PNNL, 1998) suggests the factors determining effectiveness are:

- Root production
- Metal concentration in solution
- pH
- Temperature
- Root density and plant age

Soil characteristics and rhizosphere chemistry will impact the diffusion of contamination through the soil and the interaction between the contaminant and the soil. Clay soils are more likely to show a higher tendency to bind cesium compared to sandy or loam soil due to a high affinity for cesium in clay mica layers, in which the reaction is almost irreversible and making phytoremediation difficult even for hyperaccumulators. Similarly, the chemical environment imparted by the soil on the contaminant can (in most cases) change its behavior. The pH and chemical composition of rhizosphere water, which is governed by the soil chemistry, fertilizer and amendments. In the case of amphoteric ions of elements such as uranium and plutonium whose chemical speciation, ionic charge and solubility changes drastically depending on local chemical conditions and causes significant differences in bioaccumulation. In the case of cesium contamination, local changes in pH or soil chemistry impact the soil's ability to bind contamination, and the plant biochemistry far more than the chemistry of cesium itself. As already discussed, the application of potassium fertilizers can enhance plant health and growth but can also compete with cesium for binding sites within the plant, lowering cesium bioaccumulation.

Regarding the limitations of phytoremediation, Schnoor (1997) notes difficulty accessing contaminants deeper than 3 meters, uptake of contaminants into leaves, inability to meet decontamination action levels in a short period of time, safety implications for accessing contaminated areas and vegetation at the site, and possible migration of contaminants off-site. Schnoor (1997) suggests that phytoremediation may serve as a final "polishing step" to close sites after other clean-up technologies have been used to treat the hot spots, and that winter operations may pose problems for phytoremediation when deciduous vegetation loses its leaves, transformation and uptake cease, and soil water is no longer transpired.

3.1 Plant Selection and Technical Performance

The most important factor in phytoremediation is the selection of the plant species. However, as highlighted in this work, the ability of a given plant to extract contaminants from contaminated soil or water is not the only factor in plant selection. Biomass health and production is also an important factor in plant selection. According to Soudek et al. (2007), the most critical plant property for remediation of radionuclide contamination is a high growth rate and the ability to generate large amounts of biomass in a given environment. Additionally, Soudek notes that for soil cleaning purposes, the solubility of the contaminant and its mobility in soil are the most limiting factors along with the extent of the soil volume exploited by the roots of the remediating plant species.

Substantial data exist (IAEA, 2010) on K_d values for radionuclide/soil interaction, migration rates for Cs-137 and Sr-90 for a variety of case studies, and soil-to-plant transfer factors for a significant number of elements and plant groups divided between soil types. The information can be used to calculate the contaminant transfer vertically from surface deposition to soil interaction and subsequent plant uptake, ultimately providing an estimate of bioaccumulation and thus the viability of phytoremediation for broad plant species in a range of soils for elements and radionuclides of interest to this work. The purpose of the IAEA report was to provide data for use in the radiological assessment of routine discharges of radionuclides to the environment and primarily for the use in risk assessment. However, the broad grouping of soil types and plant species in IAEA (2010) prove of limited use when considering phytoremediation plant selection. For example, when considering cesium binding in clay and loam soil, IAEA (2010) provides a mean K_d value of 3.7×10^2 with a range that differs by 4 orders of magnitude. Similarly, for cesium in sandy soil, a mean K_d value of 5.3×10^2 is provided with a range of values that differ by more than 3 orders of magnitude. Soil-to-plant transfer factors are also given in IAEA (2010). For cesium uptake in the grain of cereals, transfer factors of 1.1×10^{-2} and 3.9×10^{-2} are provided for clay and sandy soil, respectively, together with a range that differs by 2 orders of magnitude. It is important to note that IAEA (2010) information is extremely useful for risk assessment and offers boundary conditions for phytoremediation, but plant screening and selection needs to include published literature studies and values, particularly since soil conditions, chemistry, experimental duration and field versus laboratory conditions vary from study to study.

When selecting plant species for phytoremediation, the plant's ability to grow and survive in the environment and climate in which the contamination is present is important. Plants that do not typically grow or flourish in non-native environments will create a challenge for phytoremediation applications without amendments such as fertilization and irrigation. ITRC (2009) provides the following five options in regard to plant selection during the initial planning stages of phytoremediation applications, assuming the site has been first characterized:

1. species found in phytotechnology databases and growing at the site
2. species found in phytotechnology databases and suitable to the region, but not currently growing on the site
3. hybrid or species related to a plant identified as a candidate in above bullets
4. species not found in the databases but currently growing at the site or in the region
5. genetically modified organism species designed specifically to conduct the desired phytotechnology

Furthermore, ITRC (2009) suggests that general operability factors such as growth rate, habit (perennial, annual, biennial, deciduous, evergreen), form (grass, herbaceous, shrub, tree, etc.), ability to reach desired depths, water usage, disease/pest resistances, and tolerances should also be considered when selecting phytoremediation species. ITRC recommends generating a list of native plants or obtaining input from local agricultural specialists. Hybrid plants offer additional benefits over already in-situ plant species, such as increased biomass production, disease-, pest- and climate-resistance. For these reasons, ITRC (2009) states hybrids such as from poplar and willow have been extensively and successfully used in phytoremediation applications. Several decision-trees on phytoremediation application are provided by ITRC (2009), allowing the determination of techniques best suited to both the contaminant and contaminated media. While seasonal variations should be considered when selecting plant species, ITRC (2009) notes that a mix of warm and cool seasons can be considered in the initial phytoremediation planning, if phytoremediation will span multiple seasons.

When phytoremediation plant species are not already present at the contaminated site, or not enough plant mass is present, new or additional plants must be added. This represents challenges to cost, application time and safety, which not only depend on the number of plants needed, but the area to be cultivated and whether plants are added as seed or rooted plant stock. For seed application, ITRC (2009) notes that advantages include lower cost of stock, easy installation by general labor and simple storage and transport, while disadvantages include lower success of establishing root and coverage at the site, and the risk of predatory removal from the site. Similarly, for root stock, ITRC (2009) notes advantages of higher survivability, quicker remediation effects, while having higher installation cost, time and labor, requiring more complex storage, transport and planting expertise. Comparatively, ITRC (2009) notes that the cost of bare root stock is generally less than that of potted stock by approximately 25%, but that both are more costly than seed.

3.2 Labor, Expertise and Supporting Equipment

In the event of wide area contamination and the need for phytoremediation, labor, expertise and supporting equipment will come from a variety of sources, including local, state and federal governments, industry, academic and community volunteers at each stage of the process, from planning and land preparation, to maintenance, harvesting and waste management. Guidance in ITRC (2009) suggests creating a project team to plan and implement phytoremediation, generally including the following positions:

- Project Manager
- Radiochemist
- Soil Scientist/Agronomist
- Hydrologist/Geologist
- Plant Biologist/Botanist
- Environmental Scientist
- Risk Assessor/Toxicologist
- Regulatory Specialist
- Environmental Engineer
- Field Manager/Health, Safety and Environmental Officer
- Cost Engineer/Analyst

For radiological phytoremediation, substantial emphasis should be placed on hydrology and environmental science to prevent the migration of contamination, on health physics to maintain worker safety, and on waste strategies to minimize environmental and financial impacts of resultant waste. Staffing levels will depend on the scale of the radiological phytoremediation efforts, as well as the desired schedule to complete preparation, planting, removal and waste management.

Contamination covering a wide area will require a substantial amount of planting if phytoremediation is to be used effectively in removing radionuclides. Land may need to be tilled before planting occurs, and contaminated vegetation will need to be removed after phytoremediation has been completed or after a plant die-off. In these cases, given the scale of phytoremediation likely necessary in a wide area event, mechanical methods will be preferred. Traditional farming equipment to till land can be utilized. Resources could be provided by local farmers, machine rental or supply companies or manufacturers. Farming equipment will require decontamination after working in radiologically contaminated environments. In the phytoremediation pilot study at ANL-W, $10,000 of farming equipment was purchased and procedures were considered both labor and irrigation intensive (U.S. EPA, 2001). Planting mechanisms for phytoremediation purposes will depend on type and maturity of species, topography and area to be covered. Some plant species would be more amenable to mechanical planting, with others may require hand-planting. Similarly, plant maturity will dramatically impact the planting method, with seeds being easier to mechanically disperse and plant compared to already-established plants with existing root structures. Topography will also impact the ability to use mechanical methods for planting, with relatively flat surfaces easier than hillside, depending on gradient. Similarly, mechanical removal of vegetation at the end of the phytoremediation lifespan of plants will be dependent on type and maturity of species, topography and land area covered.

Schnoor (1997) states that planting density depends on the specific application, providing specific information on poplar trees, hardwood trees and grasses. Poplar hybrids can be accommodated at 1000 to 2000 trees per acre and are typically planted with a conventional tree planter at 12 to 18 inches depth or in trenched rows one to six feet deep. Hardwood trees and evergreens may require a lower planting density initially. A high initial planting density assures a significant amount of evapotranspiration in the first year, which is normally desirable. Grasses are usually drilled or broadcast for planting at waste sites. Biomass densities (above ground) of 200 to 600 g/m^2 are achieved by the second crop, with 1 to 3 crops per year depending on climate and water availability. The initial planting density of aquatic species in a created or natural wetland is normally three plants to a pod, located on three-foot centers. Schnoor (1997) also recommends the inclusion of replanting and maintenance costs in the estimated budget, with at least 30 percent of the plants potentially needing to be replanted in the second or third year, as a contingency. A general rule of thumb for a preliminary phytoremediation design, according to ITRC (2009), should include a planting density of 75 square feet per tree, staggered with an average of 10 feet on-center with a 5-foot radius of fill to create a full canopy with adjacent trees.

In some cases, irrigation may be needed to maintain plant growth and health, particularly in the case of non-native plants, or heavy planting of native plants on the contaminated site. Schnoor (1997) states that for terrestrial phytoremediation applications, it is often desirable to include irrigation costs in the design, on the order of 10 to 20 inches of water per year, noting that irrigation of the plants ensures a vigorous start to the system even in a drought. However, irrigation can lead to potential for migration of contaminants from the original site, requiring hydrologic modeling to maintain the source-term boundary. Schnoor (1997) recommends withdrawal of irrigation from the site over time provided local precipitation is sufficient to maintain plant health and growth.

In addition to sunk costs such as site assessment, literature review and feasibility studies, ITRC (2009) provides optional cost items specific to phytotechnologies including capital, engineering and design, labor and operation, and maintenance considerations for both groundcover and tree systems. Capital costs include earthwork, soil amendments, seed or root stock, site and plant protection systems, and irrigation systems, among others. Engineering and design costs should include those related to site planning, planting density and irrigation systems. Labor costs (in addition to planting) should also consider plant litter collection, spot reseeding or replanting, and expertise in determining plant health. Operating and maintenance costs should include irrigation water and electrical supply if needed, fertilizers and other amendments, invasive plant or pest control, local meteorological station, soil, plant and water sampling supplies and subsequent analysis, and transportation and disposal costs. Clearly, the cost of each item is very much site-specific.

Radiological contamination of soil, plants and equipment must be considered in all phases of the process, from land preparation, planting and irrigation, to vegetation removal, transport, and disposal. Workers will need to be trained and/or qualified on mechanical operations. At a minimum, training on the properties of radiological contamination and personal protection equipment (PPE) should be provided. Radiological field technicians can provide support for monitoring personal contamination. In reality, labor and expertise might be provided by both those experienced in farming and those with professional radiological worker qualifications. This will likely increase costs relative to other types of contamination, and potentially slow the process of evaluating and removing contamination. Additionally, mechanical equipment will likely require decontamination, generating additional waste. According to EPA (U.S. EPA, 2019), technologies and methods that are employed in other situations/applications (e.g., U.S. DOD, USDA) should serve as a starting point, and important insights can be gained by examining technologies and procedures developed by USDA to clean non-radiological contaminated farm vehicles and by DOD to decontaminate military vehicles/planes, in addition to recent experiences in Japan.

Schnoor (1997) provides 3 examples from other authors regarding remediation cost, showing that the 5-year cost for phytoremediation using hybrid poplar trees ($250k) compared favorably to that using pump and treat methods ($660k). In another example, while phytoextraction requires a longer duration compared to fixation, landfill or soil extraction/leaching methods demonstrated for metal contaminants, the costs are approximately $15 to $40 per cubic meter of soil, an order of magnitude less compared to other techniques.

A detailed cost estimate for phytoremediation of DOE's Argonne West site (Idaho) was presented in EPA (U.S. EPA, 1998), summarized below. The assessment shows significant cost savings through the utilization of phytoremediation, mostly in the management, construction and operation/maintenance costs.

Estimated Cost of Remediation Options for ANL-W Site (U.S. EPA, 1998)

	Containment	**Excavation with on-site disposal**	**Phytoremediation**
Management costs	$1,527k	$1,232k	$528k
Documentation package	$126k	$128k	$98k
Construction	$4,963k	$4,438k to $4,593k	$1,623k
Operation and maintenance	$2,347k	$780k	$780k
Total	**$8,963k**	**$6,578k to $6,733k**	**$3,029k**

3.3 Generation and Management of Waste

Management of wastes resulting from a disaster or incident (including those from NPP accidents and RDD releases) is a significant part of EPA's Emergency Support Functions responsibilities as outlined in the National Response Framework (U.S. DHS, 2016). The workshop (Jablonowski et al., 2011) sought stakeholder input on their questions and concerns regarding waste management following a hypothetical wide area RDD release, with broad reoccurring areas or themes (all of which apply to the management and final disposition of waste generated from phytoremediation activities, including:

- Regulatory restrictions/agreements/exceptions
    - Are necessary agreements in place to expedite waste handling?
    - Can disposal in non-radiological waste facilities be considered?
    - What is the response framework?
    - Who is in charge and what do they expect from the private sector?
    - Are potential waste accepting facilities willing to accept the wastes if permission can be granted by the relevant regulatory agency(ies)?
- Scientific/technological
    - What is an acceptable cleanup level for free release of sites or material?
    - How will appropriate decontamination technologies be deployed?
    - What is an acceptable level of contamination for alternate disposal in solid or hazardous waste landfills?
    - Waste disposition jurisdiction
    - How will regional low-level waste compacts be involved?
    - Can DOE disposal sites be considered?
    - What agency is ultimately responsible for decision-making and paying for disposal?
    - Is it feasible to build a landfill using RCRA Subtitle C technological specifications, potentially on government-owned land, for managing waste specifically from the incident?
- Ultimate disposition capacity
    - Is there sufficient disposal capacity to manage a large radiological incident?
    - What is necessary to provide adequate temporary storage or staging?
    - Is there local disposal capacity for the low activity waste so that only the waste with the higher activities would need to be sent to the low-level radioactive waste (LLRW) repositories?

- Communications
    - How can the public's concerns regarding storage, transportation, and disposal be addressed?

The results of the 2011 workshop (U.S. EPA, 2012) found that landfill operators that expressed unwillingness to accept incident-related waste in their own facilities might be willing to operate a government-owned landfill. In regard to waste generated from phytoremediation throughout the lifetime of the project, the following stages will generate waste:

- Test beds to select plants and methods
- Land preparation
- Planting
- Irrigation and maintenance
- Vegetation removal

Waste generation and associated costs for laboratory scoping studies are well established by routine experiments performed at universities, government agencies, national laboratories and remediation contractors. In these cases, waste generally can be categorized as LLRW. Some segregation into municipal waste can be performed to reduce the volume of LLRW generated. Similarly, PPE generated at all stages of the process may be segregated into municipal and LLRW. However, the waste generated from wide-area contamination (and subsequent large-area phytoremediation efforts) will be on a scale far greater than that experienced by industry, academia or government agencies, as demonstrated by efforts following both the Chernobyl NPP and Fukushima Daiichi NPP releases.

Waste generated from planting of vegetation for radionuclide phytoremediation will depend on the method of planting, either mechanical or manual. In both cases, contaminated PPE will be generated. For mechanical planting, equipment will need to be surveyed to determine the extent of contamination and whether disposal of contaminated equipment will occur or whether decontamination will result in additional waste. By far the largest fraction of waste generated outside the Fukushima NPP fence-line in Japan is from contaminated soil and vegetation (U.S. EPA, 2016), where the top six inches of soil was removed, and vegetation was trimmed to remove contamination deposited on plant surfaces such as leaves and branches. Selective vegetation removal was performed by hand in Japan, but larger-scale complete removal was also performed using mechanical methods such as excavators. Manual removal did not require expensive tools or skilled labor. Larger-scale removal required personnel experienced in mechanical operations and also resulted in soil removal as well as vegetation, creating larger waste volume. Vegetation may be washed, or a suppression spray may be used, to prevent airborne resuspension of contaminants (U.S. EPA, 2013).

Contaminated vegetation may undergo three different general paths for disposal. The most basic path is to bury untreated contaminated vegetation in large underground pits. Pits should be lined with impermeable material to prevent water ingress and subsequent migration of contamination outside of the pit boundary. This option results in a large volume of waste relative to other paths, in which costs associated with excavation, pit lining, waste containerization, transport and pit maintenance are highest. Shredding and compaction will help reduce volume of vegetation slightly. The remaining two paths involve treatment to minimize waste volume by separating the contaminant from the plant material by either chemical or physical means. Leaching of radioactive contamination from vegetation can reduce the volume of vegetation destined for solid LLRW disposal. However, this leaching requires the use of chemicals that can add additional waste streams including liquid waste and potentially mixed hazard waste, which are both problematic and expensive to manage.

According to Efremenkov (1989) and EPA (U.S. EPA, 2014), incineration has become a largely effective and efficient process at nuclear power plants for waste streams that have a combustible component, but further improvements are needed such as control of ultra-fine particles. Incineration can allow 50-80% or more of a solid radioactive waste to be burned efficiently, greatly reducing the volume of waste. Incineration of biomass also significantly reduces waste volume but must be administered carefully to prevent further atmospheric release and spread of contamination.

Remediation efforts in Dushenkov (PNNL, 1998) suggest that compared to excavation of 10 acres of contaminated land (resulting in 30,000 tons of waste), phytoremediation would produce approximately 1,200 tons of biomass that could be further reduced to around 120 tons of ash after incineration. Kalb and Grebenkov (PNNL, 1998) reported that in excess of 100,000 tons of ash (including soil and vegetation) with an activity up to 50 kBq/kg had been accumulated in the two years following the Chernobyl NPP accident, reporting that the safe collection, treatment and disposal of contaminated hearth ash is therefore a serious health issue in Belarus. According to Marmiroli, Samotokin, and Marmiroli (2007), a reasonable cost estimate to site incineration is about $28 per metric ton for large sites of several hundred to several thousand tons of waste generated.

In collaboration with DOE, Kalb and Grebenkov (PNNL, 1998) reported the feasibility of reduction in the volume of radioactively contaminated ash and the use of thermoplastic encapsulation technologies to stabilize contaminated ash. Waste loadings of 40-50 wt% were demonstrated without reaching maximum processing limits, and a pilot-scale feasibility test using low-density polyethylene encapsulation demonstrated waste loadings up to 70 wt%.

By 2015, an estimated 7.8 million cubic meters of combustible waste (mostly in the form of vegetation) had been generated following the Fukushima Daiichi NPP accident (Osako, 2015). A review of remediation technologies is presented by EPA (U.S. EPA, 2016) and includes a variety of incineration technologies that have been demonstrated in Japan. These include advanced off-gas treatment, transportable carbonization, superheated steam carbonization, mobile air-cooled furnace, and low temperature incineration. An approximate volume reduction rate of 95% was achieved by incineration and thermal decomposition. In Japan, fly ash with an activity less than 100 kBq/kg resulting from incineration was sent to a controlled landfill site monitored by the government. Ash greater than 100 kBq/kg is stored in the interim storage facility before being moved to a long-term permanent disposal facility (U.S. EPA, 2016).

Emissions of particulate-bound radioactive isotopes such as Cs-137 from combustion systems can be undesirable (Parajuli et al., 2013). The resulting fly ash from incineration can undergo subsequent treatment to further separate radionuclide contamination from the ash or stabilize the cesium to prevent volatilization during the incineration process. Such treatments have also been demonstrated in Japan (U.S. EPA, 2016), including the use of Prussian blue to bind cesium, magnetic nanoparticle coated absorbent, solidification, washing and absorption on resin, and melting of slag. A study by EPA (U.S. EPA, 2018) highlighted the potential for cesium vaporization during incineration and the use of kaolinite sorbent to enhance cesium capture, successfully increasing particulate diameters into the super-micron (non-respirable) range, achieving capture efficiency of 91%, and making the sorbent-bound cesium much easier to catch in particulate control devices.

In the case of wide area radiological contamination, waste generation costs are likely to be the largest fraction of the response and remediation phases. However, when adequately planned and correctly executed, phytoremediation offers the flexibility to mitigate soil and water contamination, while providing options for waste minimization.

4. Recommendations and Gaps in Phytoremediation Use in Wide Area Radiological Contamination Events

Phytoremediation offers a viable method for stabilizing and removing contamination with significantly less cost than alternatives such as excavation or pump-and-treat methods. Selection of appropriate plant species requires understanding of local soil and climate conditions and assessment of hyperaccumulator feasibility in the local environment. Subject matter experts should be convened early in the planning process, in conjunction with stakeholder input to determine the holistic approach to phytoremediation, from site preparation and plant selection through waste generation and disposal. The extent to which plant uptake has been researched for a wide variety of radionuclides (and stable isotopes) is significant, largely performed to better understand the effects of contamination on the environment, on the food chain and in relation to human health risk assessment. Significant experience has been gained through the application of phytoremediation technologies applied to wide areas in the former Soviet Union and Japan following nuclear power plant accidents. This experience has driven both scientific data and technical innovations to improve phytoremediation understanding and minimize waste generation. Plant species such as amaranthus, Indian mustard, kochia and hybrid varieties of poplar and willow have been shown repeatedly in published literature to serve as hyperaccumulators, but this should be considered together with soil type and contaminant geochemistry. However, according to Schnoor (1997), “Phytoremediation systems are like any other treatment scheme; one cannot simply walk away from them and expect success. There are events that can cause failure that should be realistically assessed at the outset. These include killing frosts, wind, storms, animals, disease or infestation, and latent toxicity.”

Nevertheless, technical gaps in our knowledge base still exist, particularly when phytoremediation is applied to soils with a high clay content. In such cases, the use of amendments and the addition of bacteria have shown to be somewhat beneficial in increasing bioaccumulation effectiveness. However, research in this area is still lacking. Recent work by Varazi et al. (2015) screened plant species, bacterial strains, and natural sorbent materials (e.g., native clays and rock formations) to create enhanced bioaccumulation systems. Work by Djedidi et al. (2014 and 2016) and Aung (2016) have also demonstrated the enhancements achieved when microbial strains are added to a phytoremediation regimen. More experience can be learned from continued management of vegetation waste in Japan.

The next step will be to identify under what conditions phytoremediation methods would be beneficial compared to other remediation options and which of the methods would be most applicable. The comparison should be comprehensive to include factors such as radiation reduction, cost, difficulty, time, labor, stakeholder perception, etc. The beneficial phytoremediation methods should be further developed with their operating procedures to prepare for responses to the potential contamination scenarios.

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