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Soil contamination with radionuclides and potential remediation

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Abstract

Soils contaminated with radionuclides, particularly ^{137}Cs and ^{90}Sr, pose a long-term radiation hazard to human health through exposure via the foodchain and other pathways. Remediation of radionuclide-contaminated soils has become increasingly important. Removal of the contaminated surface soil (often up to 40 cm) or immobilization of radionuclides in soils by applying mineral and chemical amendments are physically difficult and not likely cost-effective in practicality. Reducing plant uptake of radionuclides, especially ^{137}CS and ^{90}Sr by competitive cations contained in chemical fertilizers has the general advantage in large scale, low-level contamination incidents on arable land, and has been widely practiced in central and Western Europe after the Chernobyl accident. Phytoextraction of radionuclides by specific plant species from contaminated sites has rapidly stimulated interest among industrialists as well as academics, and is considered to be a promising bio-remediation method. This paper examines the existing remediation approaches and discusses phytoextraction of radionuclides from contaminated soils in detail.

1. Introduction

Radionuclides exist in the environment either naturally or artificially. It has been estimated that, on average, 79% of the radiation to which humans are exposed is from natural sources, 19% from medical application and the remaining 2% from fallout of weapons testing and the nuclear power industry (Wild, 1993). However, most of the public concern over radiation from radionuclides has been due to the global fallout from atmospheric nuclear weapons testing and the operation of nuclear facilities. Both of these activities have introduced a substantial amount of man-made radionuclides into the environment and have caused radionuclide contamination of large areas of land worldwide. Contamination of soils with typical fission product radionuclides, such as ^{137}Cs and ^{90}Sr, has persisted for far longer than was originally expected (Kirk and Staunton, 1989). Radionuclides in soil are taken up by plants, thereby becoming available for further redistribution within foodchains. For example, two peaks of radiocaesium activity concentration were identified in milk in the UK in 1964 and 1986, corresponding to the most active period of nuclear weapons testing and the Chernobyl accident, respectively (Department of the Environment, 1994). Radionuclides in the environment can, therefore, eventually be passed on to human beings through food chains, and so may represent an environmental threat to the health of local populations (Howard et al., 1991; Robinson and Stone, 1992).

Although atmospheric testing of nuclear bombs has been banned globally, in the foreseeable future the nuclear power industry will continue to make an increasing contribution to power consumption, a trend which could help to reduce the global warming process due to the consumption of fossil fuel. As accidental and routine releases of radionuclides from the nuclear industry are inevitable, and can cause local, regional and even global environmental contamination, remediation of soils

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contaminated with radionuclides is becoming an increasingly important aspect of radiological protection.

2. Radionuclides in soils and plant uptake

Radionuclides which have been responsible for most environmental concern are listed in Table 1. A radionuclide undergoes the same reactions in soil as the non-radioactive isotope of the element in question (Wild, 1993), although their physical concentrations in soil are normally very low compared to non-radioactive isotopes. Once radionuclides enter the soil environment, their fate is determined by their own and the soil's physio-chemical properties, as well as by factors such as climate and vegetation. Understanding their geochemical behaviour in soil–plant systems is of critical importance for the modelling of their transport and retention in soils, transfer from soil to plants and hence into the food chain, and phytoremediation (Cremers et al., 1988; Agapkina and Tikhomirov, 1994; Ebbs et al., 1998). Ebbs et al. (1998) suggested that a soil $pH < 5.5$ would be required to convert U to its most phyto-available from in soil. For radiocaesium, 2:1 type clay minerals and organic matter within the soil have been shown to be the most important controls on its geochemical behaviour, and it is generally accepted that, following the Chernobyl accident, radiocaesium has been mainly retained in the surface horizons of soils mainly due to reaction with clay and humic components and/or the soil microflora (Thiry and Myttenaere, 1993). The migration velocities of ^{137}Cs and ^{90}Sr in typical soils of the Khoiniki district, Gomel region of Belarus, were shown to be 0.39–1.16 cm/yr and 0.71–1.54 cm/yr, respectively, and these were strongly influenced by soil type (Arapis et al., 1997). Detailed discussion of these results is beyond the scope of this paper.

Consumption of agricultural produce contaminated with radionuclides represents the principal route of internal intake of radionuclides in humans (Shaw and Bell, 1994). It is for this reason that plant uptake of radionuclides has been widely studied from the early 1940s e.g., (Collander, 1941). Accumulation of radionuclides has been investigated on a wide array of plant species and has been shown to vary greatly among different habitats and plant species (Horrill et al., 1990; Entry et al., 1996; Broadley and Willey, 1997). To illustrate this, radiocaesium activity concentrations in species of frequent occurrence in upland habitats are listed in Table 2. Willey and Martin (1997) and Broadley and Willey (1997) investigated the differences in caesium uptake by 30 plant taxa and maximum differences in accumulation of caesium were found between *Chenopodium quinoa* and *Koeleria macrantha* (with 20-fold differences in Cs concentration and 100-fold differences in total Cs accumulation).

Information gathered on plant uptake of radionuclides has primarily been used for radiological assessment and radioecological research. This kind of information has now become more important for the purpose of phytoremediation of sites contaminated with high levels of radionuclides, such as nuclear facilities to be decommissioned and/or areas of land subject to accidental releases of radioactivity. A new EU coordinated data base of plant uptake of radionuclides from soil is being constructed at the moment, which will be useful both for devising strategies for phytoremediation as well as for more general radiological assessment (Nisbet, A.F., personal communication).

3. Physical approaches towards remediation of radionuclides contaminated soils

The ultimate remediation of radionuclide-contaminated soils probably would require soil to be removed from the affected site and to be treated with various dispersing and chelating chemicals (Entry et al., 1996). This method might be applicable to small scale sites with high activity concentrations of radionuclides. Removing and transporting soil is difficult in terms of equipment and manpower and is therefore very costly.

In addition, the cost of disposing of the vast amount of liquid dispersing and chelating chemicals may also be prohibitive (Entry et al., 1996). In the USA, the Department of Energy's Assistant Secretary for Environmental Restoration and Waste Management was quoted as stating that the \$200 to \$300 billion cost of radionuclide cleanup may be a dramatic underestimate (Entry et al., 1996). This method is even less feasible in

Table 1
Characteristics of major radionuclides that occur in soil

Isotope	Half-life (yr)	Principal radiation	Main occurrence
^{14}C	5.7×10^3	β^-	Natural and nuclear reactor
^{40}K	1.3×10^9	β^-	Natural
^{90}Sr	28	β^-	Nuclear reactor
^{134}Cs	2	β^-, γ	Nuclear reactor
^{137}Cs	30	β^-, γ	Nuclear reactor
^{239}Pu	2.4×10^4	α, X-rays	Nuclear reactor

Table 2
Range of activity concentrations of ^{137}Cs for species of frequent occurrence in upland habitats (Bq/kg dry weight), after Horrill et al. (1990)

Species	Activity concentration of ^{137}Cs				
	Number	Max	Min	Mean	SE
Juncus effusus	14	1341	14	378	116
Nardus stricta	13	868	10	237	86
Calluna vulgaris	7	4464	39	915	598
Pteridium aquilinum	7	700	11	236	96
Juncus squarrosus	6	1989	101	832	301
Erica cinerea	5	93	28	52	12
Erica tetralix	3	2324	179	1212	620
Vaccinium myrtillus	4	220	78	141	33
Polytrichum commune	10	4722	339	1589	410
Sphagnum spp.	10	5918	411	1992	551

the case of large scale contamination with low level concentrations of radionuclides. When such widespread contamination occurs in semi-natural ecosystems, which are often difficult to access using conventional machinery, then the problems of remediation becomes even more formidable. In the event of a nuclear accident contaminating forests, for instance, Guillitte et al. (1994) suggested a few possible physical countermeasures to reduce the consequences of contamination:

- spraying contaminated canopies with detergents or other cleaning agents;
- defoliation and removal of fallen leaves;
- clear-felling and removal of timber;
- ploughing after clear-felling and prior to planting;
- scraping and removal of the surface layer.

The practicality of such countermeasures, however, remains to be proven in practice.

4. Agriculture-based countermeasures

Processes involved in the transfer of radionuclides from soils to plant are affected by a number of factors, which are summarized in Fig. 1. Intervention to reduce human intake of radionuclides via the food chain can be implemented at several levels. It has been generally suggested that the application of agriculture-based countermeasures during crop production is one of the most effective ways to reduce the radiation dose to human population following a nuclear contamination event. Once radionuclides are deposited onto the soil, transfer from soil to plant is controlled at two interfaces, i.e. the soil–liquid interface within the soil and the soil solution–plant root interface.

4.1. Application of mineral and chemical amendments

The application of mineral and chemical adsorbents to soils is intended to reduce the phyto-availability of radionuclides within the soil. Valcke et al. (1996) suggested that if the radionuclide adsorption potential of the adsorbent exceeds that of the contaminated soil by a factor of 100, the material will be effective as an amendment. Natural and synthetic zeolites have been some of the best candidates for this purpose, and have been demonstrated to increase greatly the K_d (soil–liquid distribution coefficient) of radiocaesium in soils (Table 3). Plant growth experiments have shown that the addition of a natural zeolite to a podzol (1%, W/W) reduced the soil-to-plant transfer factor significantly from 0.75 to 0.164 (Valcke et al., 1996).

Ammonium-ferric-hexacyano-ferrate(II) (AFCF) is another chemical substance which is believed to be of great potential value in mitigating soil contamination with radiocaesium. It has been shown that, on a sandy

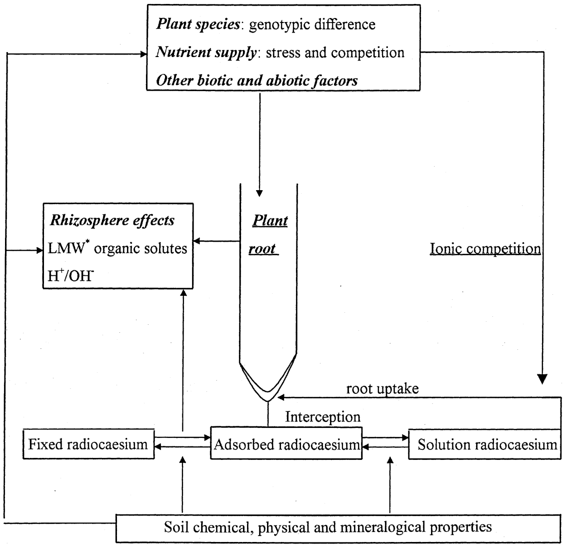

Fig. 1. Factors affecting radiocaesium uptake by plant roots, * = low molecular weight.

Table 3
Effect of adsorbent amendments on radiocaesium partitioning between soil solid and liquid phase (K_d value), data from Valcke et al. (1996)

Soil origin and type	Treatment	K_d (dm^3/kg)
Wyn-A, Belgium, Podzol	Na-Mordenite, 1%	2024
	No amendment	897
Vetka, Belorus, Sandy podzol	Natural mordenite, 1%	1832
	No amendment	896
Bragin, Belorus, Peat	Natural mordenite, 1%	1751
	No amendment	638
Novozibkov, Russi, Podzol	Natural mordenite, 1%	1383
	No amendment	562

soil, application of AFCF (10 and 100 g/m^2) can reduce radiocaesium transfer to ryegrass by a factor of 25–255, respectively, without causing plant growth problems (Vandenhove et al., 1996). Even a slight addition (1 g/m^2) caused a four-fold reduction of the transfer factor. However, the effectiveness of AFCF is also related to the inherent Cs-fixation capacity of the soil. In a loamy soil, with an apparently higher Cs-fixation capacity than sandy soil, 10 g AFCF/m^2 resulted in only a twofold reduction of the transfer factor (Vandenhove et al., 1996).

4.2. Application of K- and Ca-containing fertilizers

Application of fertilizers to soils induces both chemical and biological changes which affect the overall transfer of radionuclides from soil to plant. It was realized in the 1960s that application of K- and Ca-based fertilizers to contaminated soils can depress the uptake of their respective radioactive analogues, radiocaesium and radiostrontium, by plants (Jackson et al., 1965; Evans and Dekker, 1968). However, results from recent and pre-Chernobyl studies have often been inconclusive, which is understandable given the wide range of soils investigated and the rates and forms of treatments applied (Nisbet, 1993).

The authors of this paper have conducted a series of experiments to demonstrate the suppressive effects of potassium on the uptake of radiocaesium by plants (Shaw and Bell, 1991; Shaw et al., 1992; Zhu, 1998). In large scale hydroponic experiments it was demonstrated unequivocally that increasing the external potassium concentration dramatically reduces the influx rates of radiocaesium into both spring wheat and broad bean plants (Fig. 2). The relationship between radiocaesium influx and external potassium concentration can be described empirically by a negative power function, which implies that when external potassium concentration exceeds a certain level (around 10 mg/l in our case) any further increase will only result in a slight augmentation of the inhibitory effect on radiocaesium uptake by plants. This inhibitory pattern of potassium on radiocaesium uptake by plant roots is strongly supported by recent findings by Sacchi et al. (1997), which provided evidence that there was low selectivity between Cs^+, $Rb^+$and K^+in a carrier-mediated transporter operating primarily at low external concentrations (which possibly uses a cation-H^+co-uptake mechanism) and higher selectivity for K^+ in a channel-mediated system operating at high external potassium concentrations.

We have also carried out lysimeter experiments to demonstrate the potential effect of potassium fertilization on the uptake of radiocaesium under field conditions. A weaker, although significant, relationship

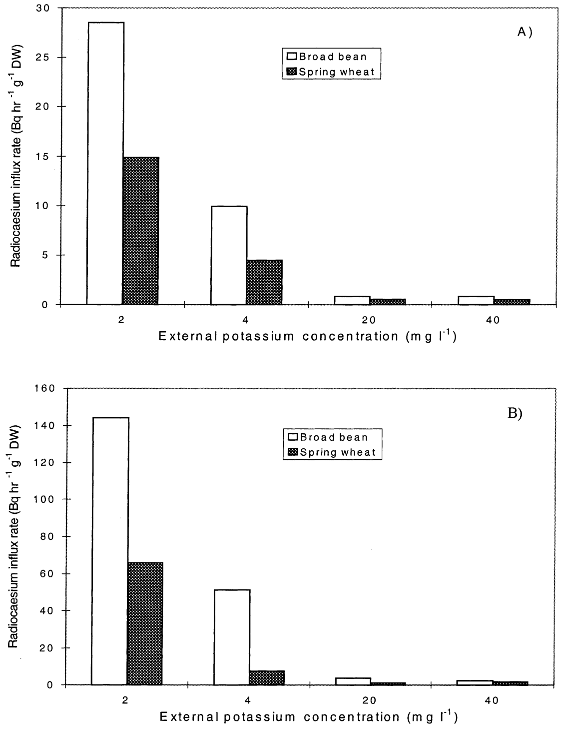

Fig. 2. The effect of external potassium concentrations on the influx rates of radiocaesium into plant roots: (A) 56 days after transplanting; (B) 21 days after transplanting.

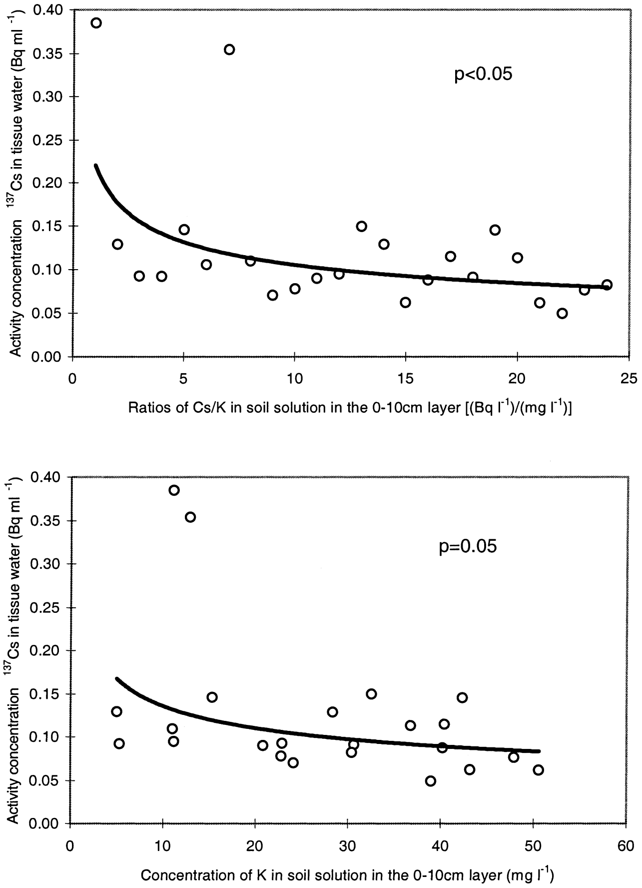

Fig. 3. Correlation of activity concentrations of ^{137}Cs in shoots of spring wheat with potassium and radiocaesium in soil solutions (0–10 cm).

between radiocaesium uptake and potassium concentrations in the soil solution was observed (Fig. 3). The effect of potassium on the uptake of radiocaesium by crops under field conditions was much less obvious than in the hydroponic system, which reflects the combined effects of the complex of factors controlling ion uptake from soils. Nevertheless, as potassium fertilization is practiced in most agricultural production systems to sustain the soil fertility, the inhibitory effect of potassium on the uptake of radiocaesium by crops is still viable in terms of radiological protection and assessment, especially in permanent grassland ecosystems.

5. Bioremediation

Soils contaminated with significant quantities of radionuclides in the vicinity of nuclear facilities (reactors, weapons research establishments, nuclear fuel production and waste reprocessing facilities, etc.) represent one of the major sources of radioactive contamination for both food chains and groundwaters (e.g., Ebbs et al., 1998). Remediation of these sites is an increasingly pressing problem in many places of the world and bioremediation is emerging as an alternative to some of the energy-intensive and high-cost soil cleaning methods discussed above.

5.1. Soil fungi and mycorrhizae

Fungi are often the major component of the soil microflora, particularly in acid soils, and play a critical role in the soil's ecological functions. High radiocaesium activities have been reported in the fruiting bodies of a number of fungal species both before and since the Chernobyl accident (e.g., Dighton and Horrill, 1988). Clint et al. (1991) have measured the influx of stable (non-radioactive) caesium into hyphae of 18 fungal species. The mean observed influx rates were between 85 and 276 nmol/g dry wt/h, which explained the large variation of radiocaesium activities in fruiting bodies of different fungal species collected in field surveys after the

Table 4
Phytoextraction of radiocaesium from contaminated soils, data derived from Lasat et al. (1998)

Plant species	Indian mustard	Red root pigweed	Tepary bean
Total ^{137}Cs in soil (kBq/m^2)[a]	2640 ± 480	2400 ± 960	1680 ± 720
^{137}Cs removed in shoots (kBq/m^2)	1.2 ± 0.12	52 ± 26.4	0.8 ± 0.4

[a] Assuming the soil mass over 20 cm depth is 240 kg.

Chernobyl accident. Dighton et al. (1991) further demonstrated that upland grassland soil fungi were a potential pool of radiocaesium immobilization. Based on these findings on bioaccumulation of radionuclides by soil fungi, it might be viable to explore the possibility of using the fungi to ‘lock up’ the radionuclides (radiocaesium in particular) in surface soil layers so as to reduce soil migration and a groundwater pollution.

Mycorrhizal fungi are an important group of soil fungi which form close symbiotic relationships with most terrestrial plants and, in doing so, can alter their nutrient acquisition patterns (Smith and Read, 1997). Investigations into the role of mycorrhizae in the context of soil radionuclide contamination have been inconclusive. For example, Riesen and Brunner (1996) reported that ectomycorrhizae reduced the uptake of radiocaesium and radiostrontium by Norway spruce seedlings, but Entry et al. (1994) reported that ectomycorrhizal infection lead to the enhancement of radionuclide uptake. The same situation applies to the VA mycorrhizae: Haselwandter et al. (1994) observed an increase in radiocaesium uptake by VA mycorrhizal *Paspalum notatum* while Haselwandter and Berreck (1994) found that VA mycorrhizae could decrease the uptake of radiocaesium by *Festuca ovina*. While there is increasing interest in the role of mycorrhizae in soil heavy metal pollution generally, the authors suggest that it would be worthwhile conducting further investigations into the role of mycorrhizae (or soil fungi in general) in designing better management strategies for land contaminated with radionuclides. Such investigation could eventually influence substantially the design of agricultural countermeasures and, in particular, bioremediation strategies.

5.2. Phytoextraction

As discussed above, different plant species have different abilities to accumulate radionuclides from soil. While this variation has particular relevance in terms of being able to reduce the transfer of radionuclides from soil to food chains, it can also be exploited for the purpose of phytoremediation. Willey and his colleagues (Broadley and Willey, 1997; Willey and Martin, 1997; Willey, personal communication) have obtained relative radiocaesium uptake values in about 200 species and found that the highest values are all in the Chenopodiaceae or closely related families. Selection of such taxa could be used to increase the phytoextraction of radiocaesium from contaminated soils.

Lasat et al. (1997, 1998) have also conducted hydroponic and field experiments to select potential plant candidates for phytoremediation. They identified that red root pigweed (*Amaranthus retroflexus*, which is closely related to the Chenopodiaceae) is an effective accumulator of radiocaesium which is capable of combining a high degree of uptake of ^{137}Cs with high shoot biomass production (Table 4).

According to this set of data, it might be feasible to decontaminate moderately radiocaesium-contaminated soil in less than 20 yr with two croppings of *Amaranthus retroflexus* per year. Phytoremediation could be optimised by applying soil amendments to release the fixed-form of radionuclides. Lasat et al. (1998) suggested that application of NH_4NO_3 might be beneficial by mobilizing fixed radiocaesium in the soil so as to speed up phytoextraction. Ebbs et al. (1998) found that application of citric acid could greatly improve the shoot U concentration of beet grown five weeks in U-contaminated soil.

However, with the current knowledge of plant uptake of radionuclides from soils, phytoremediation of soils contaminated with radionuclides is still likely to take an excessively long time. To speed up the process of selection of suitable plant taxa, a special plant breeding program assisted by molecular biotechnology may be useful. Recent developments in molecular aspects of plant mineral nutrition, such as identifying and cloning the genes responsible for Zn^{2+}, $K^+$and other transporters in plant root cells, have provided some promising evidence for the potential acceleration of phytoremediation technology. A problem arising from current phytoremediation strategies is also related to the sustainability of the fertility of contaminated soils. After a sustained period of phytoextraction the soil may well be exhausted for subsequent use. Therefore, soil nutrient management should be an integral part of phytoremediation and needs equal attention.

6. Conclusions

Contamination of soil with radionuclides is a worldwide problem which has developed in line with the development of nuclear technology itself over the last few decades. Physical approaches to eliminate or clean-up polluted sites have often proved to be prohibitively expensive. Agriculture-based countermeasures, such as application of minerals or chemical fertilisers, have been proven to be effective in reducing the transfer of radionuclides from soil to plants in agricultural production systems with low levels of contamination. However, further investigation is still needed both to optimize these countermeasures and to understand any side effects in which non-standard additions of agrochemicals may result. Bioremediation is emerging as an alternative approach. Using soil fungi (or the soil microflora in general) to retain the radionuclides in the surface soil may be a new strategy to reduce soil migration of radionuclides and to avoid ground water pollution. Phytoextraction is a near-ideal method for clean-up of polluted sites. However, development of comprehensive and integrated technologies by soil chemists, plant physiologists and molecular biologists is required for efficient and sustainable remediation.

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