29 Phytomanagement of Radioactively Contaminated Sites

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CONTENTS

29.1 INTRODUCTION

The application of nuclear energy and the use of radionuclides for industrial, medical, and research purposes have caused significant contamination of certain sites and their environments, which could result in health problems for several centuries if nothing is undertaken to remedy these situations. Except for the close environment of the facility, where decontamination activities may be feasible and affordable, the contamination often extends over a vast area and decontamination would result

in considerable amounts of waste. Therefore, a search should be conducted for more realistic, yet efficient, remediation options; phytomanagement is among the potential options.

Major sources of radioactive contamination and major radionuclides released with long-term impact are [1]:

- Nuclear weapon testing (release of mainly ^{14}C, ^{137}Cs, ^{90}Sr, and ^{95}Zr)
- Nuclear weapon production (release of mainly ^{137}Cs, ^{106}Ru, and ^{95}Zr)
- Nuclear power production
  - During the mining operation, the main radionuclide discharged is ^{222}Rn; the environment of the U mining and milling sites is contaminated through dispersion of ^{238}U (and daughters — e.g., ^{226}Ra, ^{210}Pb, and ^{210}Po) and ^{232}Th.
  - During the operational phase, small amounts of radionuclides are routinely released, mainly ^{14}C.
- Nuclear accidents can involve only small local contamination, such as in the Goiania accident involving a discarded ^{137}Cs medical source; other accidents have been of much greater significance (Chernobyl, Three Mile Island, Kyshtym, and Windscale). Although the cocktail of radionuclides released is case dependent, in general, elements released with a scale impact were ^{137}Cs, ^{90}Sr, and ^{131}I (^{210}Po, ^{95}Zr, and ^{144}Ce).
- Natural sources of contamination comprise those of terrestrial linked with ^{40}K, and the ^{238}U, ^{235}U, and ^{232}Th decay chains. World weight averaged concentrations are 420 Bq kg^{-1} for ^{40}K and, respectively, 33 and 45 Bq kg^{-1} for ^{238}U and ^{232}Th series radionuclides. In most minerals, natural levels of radionuclides are very low. In others, e.g., zircon and rare earths, the concentration of ^{238}U and ^{232}Th may be considerably elevated (e.g., for apatite, 2 kBq kg^{-1}; for zircon, 10 kBq kg^{-1}; for U ore, ~200 kBq kg^{-1}; and for monazite, 350 kBq kg^{-1}. During recent years, increasing interest has been paid to contamination linked with industries handling materials containing elevated levels of natural radionuclides (NORs). Ore extraction and processing may lead to enhanced levels of naturally occurring radionuclides (NORs) in products, by-products, and waste and in surroundings and installations of the facility. The most contaminating industries are uranium mining and milling, metal mining and smelting, and the phosphate industry [2,3].

In this chapter, the potential role of different phytomanagement options for the remediation of sites contaminated with radionuclides is discussed and illustrated with some examples. The phytomanagement options considered are: phytoextraction (including rhizofiltration), alternative land uses and phytostabilization. The radionuclides considered are the fission products ^{137}Cs and ^{90}Sr and the natural radionuclides uranium and, to some extent, radium and thorium.

29.2 POSSIBLE ROLE OF PHYTOMANAGEMENT

29.2.1 PHYTOEXTRACTION

Phytoextraction is an approach to the cleanup of contaminated soils using accumulator plants. This process requires that the target metal (radionuclide) be available for the plant root, absorbed by the roots, and translocated from the root to the shoot and biomass production should be substantial. The metal (radionuclide) is removed from the site by harvesting of the biomass; after that, it is processed to recover the metals or further concentrate the metal (thermal, microbial, chemical treatment) to facilitate disposal.

To maximize the metal content in the biomass, it is necessary to use a combination of improved soil management inputs (e.g., optimized soil pH and mineral nutrition, addition of agents that increase the availability of metals in soils), improved genotypes with optimized metal uptake,

translocation and tolerance, and improved biomass yield. The economics of the operation not only depend on the phytoextraction efficiency but also on costs associated with crop management (soil management, crop establishment costs [yearly returning for annual crops], crop harvest); postharvest biomass transport; and biomass treatment and potential disposal.

Contrary to the research on heavy metals, the phytoextraction approach for radionuclides (RN) is rather new. Furthermore, most experiments conducted to test the phytoremediation approach for RN have been done in hydroponics systems [4,5].

29.2.2 The Potential for Phytoextraction

The soil-to-plant transfer of metal contaminants is a major process that must be considered in the management of metal-contaminated sites. These fluxes should be minimized to reduce exposure of soil pollutants in the food chain or maximized in an attempt to remediate a soil by phytomining or phytoextraction. This section will give an overview of the soil–plant fluxes of metal contaminants. Examples will be given for three radiocontaminants that are considered important due to their toxicity and/or ubiquity. These elements are the natural radionuclide uranium, one of the predominant contaminants in the uranium mining and milling industry; the phosphate industry and other NORM industries; and the long-lived fission products radiocaesium (^{137}Cs) and radiostrontium ^{90}Sr.

The flux of an element from soil to plant is often referred to as the crop off-take or removal of that element, i.e., the removal of the element from soil with the harvested part of the crop. The removal of a contaminant from the soil with the harvested biomass (in Bq ha^{-1}) is the product of the concentration in the plant (C_{plant} in Bq kg^{-1}) and the yield of the harvested biomass (kg ha^{-1}):

$$\text{Crop removal} = \text{yield} \times C_{plant} \tag{29.1}$$

The concentration of an element in the plant depends on its concentration in the soil, type of soil, plant type, etc. It has been demonstrated that crop concentrations of nonessential trace elements, U, ^{137}Cs, and ^{90}Sr, are proportional to their concentration in soil, for the same crop, soil type, etc. The proportionality constant is defined as the transfer factor (*TF*, dimensionless)

$$\text{TF} = \text{C}_{\text{plant}}/\text{C}_{\text{soil}} \tag{29.2}$$

where C_{plant} is the concentration of the radiocontaminant in the plant (Bq/g) and C_{soil} is the concentration of the contaminant in the soil. The transfer factor is thus an important parameter determining the potential of phytoextraction. Table 29.1 gives a summary of transfer factors.

TABLE 29.1
TFs for Cs, Sr, U and Ra: Total Ranges, Ranges of Recommended Values, and Upper Limits (Several Sources)

	Total range	Range of recommended values	Upper limit	Comment on conditions for upper limit
Cs	0.00025–7.5	0.0038–0.29	7.5	Brassica, organic soil
Sr	0.0051–22	0.017–3.2	22	Green vegetables, sandy soil
U	0.000006–21.13	0.00075–0.02	21.13	Tubers, sandy soil
Ra	0.00048–0.66	0.00097–0.12	0.66	Ryegrass, organic soil
Th	0.000021–0.270	0.00004–0.058	0.27	Grass, U mining area

With Equation 29.1 and Equation 29.2, the soil-to-plant transfer factor (TF), the percentage yearly reduction in soil activity can be calculated as follows:

$$\text{Annual removal (\%)} = \frac{\text{TF} \times \text{Yield}}{\text{W}_{\text{soil}}} \times 100 \tag{29.3}$$

where W_{soil} is the weight of the contaminated soil layer (kg ha^{-1}). As made clear by Equation 29.3, the annual removal percentage increases with yield and TF. However, TF and yield values are not independent: high yield is often associated with lower TFs because of growth dilution effects.

Phytoextraction requires several years and the future trend in radionuclide concentration in the soil can be calculated according to:

$$\text{C}_{\text{soil,t}} = \text{C}_{\text{soil}_{\text{t=0}}} \exp\left\{-\left(\frac{\text{TF} \times \text{yield}}{\text{W}_{\text{soil}}} + \frac{0.69}{\text{t}_{1/2}} \right) \times \text{t}\right\} \tag{29.4}$$

The second term in the exponent of Equation 29.4 was included to account for radioactive decay ($t_{½}$ is the half-life of the radionuclide). Given the half life of ^{238}U (4.5 10^9 years), this component will not affect the phytoextraction potential. For ^{137}Cs and ^{90}Sr, with half-lives of 30 years, the phytoextraction potential will be affected (2.33% yearly loss in activity merely through radioactive decay). Equation 29.4 assumes a constant bioavailability of the contaminant, i.e., a constant TF.

For a soil depth of 10 cm and a soil density of 1.5 kg dm^{-3}, soil weight is 1,500,000 kg. Table 29.2 shows the percent of annual removal for different crop yields and TFs. Yields of more than 20 ton ha^{-1} and TFs higher than 0.1 (Table 29.1) may be regarded as average values or upper limits, except for Sr. This would result in an annual reduction percentage of 0.1% (excluding decay). In the case of a TF of 1, annual reduction is about 1%.

Rearranging Equation 29.4 allows calculation of the number of years needed to attain the required reduction factor as a function of annual removal percentage. Table 29.3 tabulates the years required to attain a reduction of the contaminant concentration up to a factor of 100, given an

TABLE 29.2
Percentage Yearly Reduction of Soil Contamination due to Phytoextraction (and Radioactive Decay [$t_{½}$ 30 years])

	Annual reduction % due to phytoextraction					Annual reduction % due to phytoextraction and radioactive decay				
Yield (t ha^{-1})	**5**	**10**	**15**	**20**	**30**	**5**	**10**	**15**	**20**	**30**
TF (g g^{-1})										
0.01	0.0033	0.0067	0.01	0.013	0.02	2.333	2.337	2.34	2.343	2.35
0.1	0.033	0.067	0.1	0.133	0.2	2.363	2.397	2.43	2.463	2.53
1	0.33	0.67	1.00	1.33	2.00	2.66	3	3.33	3.66	4.33
2	0.67	1.33	2.00	2.67	4.00	3	3.66	4.33	5	6.33
5	1.67	3.33	5.00	6.67	10.00	4	5.66	7.33	9	12.33
10	3.33	6.67	10.00	13.33	20.00	5.66	9	12.33	15.66	22.33

Notes: Soil depth 10 cm; soil density 1.5 kg dm^{-3}.

Source: Vandenhove, H. et al., *Int. J. Phytorem.,* 3(3), 301, 2001.

TABLE 29.3
Years Required to Decontaminate a Soil Given a Required (Desired) Reduction Factor and a Given Annual Removal Percentage

Desired reduction factor	% activity remaining	Annual removal, % y^{-1}							
	$C_{soil,t}/C_{soil,t=0}$	20	15	10	5	3	2	1	0.1
1.5	66	2	2.5	4	8	13	20	40	400
2	50	3	4	7	14	23	34	69	693
3	33	5	7	11	22	36	55	110	1108
4	25	6	9	13	27	46	69	138	1609
5	20	7	10	15	31	53	80	160	1650
10	10	10	14	22	45	76	114	229	2301
20	5	13	18	28	58	98	148	298	2994
50	2	18	27	37	76	128	194	389	3910
100	1	21	28	44	90	151	228	458	4603

Notes: Soil depth 10 cm; soil density 1.5 kg dm^{-3}.

annual extraction percentage or percentage reduction in radionuclide activity varying between 0.1 and 20%. With an annual removal of 0.1%, it would take more than 2000 years to decontaminate a soil to 10% of its initial activity. With an annual removal of 1%, more than 200 years are required. Thus, it is clear that measures should be taken to increase the annual removal efficiency through crop selection, increasing the bioavailability with soil additives, and technical actions (e.g., decreased soil depth).

Decreasing the soil depth increases the removal percentage, according to Equation 29.3, and may intensify root–soil contact and result in an increased TF. The soil can be spread on geomembranes, which impede roots from penetrating to deeper layers. These membranes will also limit contamination of the underlying clean soil, but a substantial area may be needed for treatment. In most cases of soil contamination, control on the depth of the contamination is limited, though it may be feasible and advantageous to excavate and spread out the soil over the desired soil depth for phytodecontamination purposes. Therefore, considering a soil depth of 10 cm may be a realistic assumption.

Decreasing the soil depth is not specific to radionuclide and plant. The other factors influencing radionuclide bioavailability (crop selection and increasing the bioavailability of the radionuclide of concern) will be discussed per radioelement. For each element, a short description on the behavior of the element in soil will be given.

29.2.2.1 Uranium

29.2.2.1.1 Behavior of U in Soil

Natural uranium exists as three isotopes — ^{238}U, ^{235}U, and ^{234}U — with a relative abundance of 99.27, 0.720, and 0.0055%, respectively. The world average ^{238}U activity in soil is 40 Bq kg^{-1} [6]. Uranium behavior is similar to that of other heavy metals and its physiological toxicity mimics that of lead. Uranium is chemically toxic to kidneys and insoluble U-compounds are carcinogenic [7].

The complex, pH-dependent speciation of U in soils makes the study of U uptake by plants difficult. Uranium is present in soil primarily (80 to 90%) in the +VI oxidation state as the uranyl (UO_2^{+2}) cation [8]. Under acidic conditions, UO_2^{+2} is the predominant U species in the soil. Hydroxide complexes form under near-neutral conditions, and carbonate complexes predominate under alkaline conditions. Generally, negligible amounts of UO_2^{+2} are present in available forms, due to the high solid–liquid distribution coefficient of uranium (K_D = ratio of the radionuclide in

the solid soil fraction to the radionuclide concentration in the soil solution: Bq kg^{-1}/Bq dm^{-3}) (620 dm^3 kg^{-1}, [9]). Though extensive work has been done on U solubility in soils, comparatively little information is available regarding the uptake and translocation of U by plants as affected by soil properties. Work has focused primarily on the U content of native plants growing in U-contaminated environments [11,12] or U uptake by field and garden crops of importance to animals and humans [13–15].

29.2.2.1.2 Phytoextraction Potential of U

The U soil-to-plant TF varies with plant compartment and plant species. Roots incorporate much more uranium than stems, leaves, and seeds do [16–18].

Leafy vegetables generally show higher U TFs, followed by root, fruit, and grain crops (Table 29.4). TF values for the plants that have been studied rarely exceed a value of 0.1, except in plants grown on highly contaminated, often acidic U-mining sites. A study of U accumulation by mixed grass species, forbs, and big sagebrush (*Artemesia* spp.) growing near a U mine/mill complex indicated that plants growing on exposed tailings (low pH) had a TF value that approached unity; plants grown in proximity to the tailings pile had TF values ranging from <0.1 to approximately 0.4, depending on the distance from the tailings and the wind direction [12]. The plant with the highest TF for natural conditions is pH sensitive and does not grow at a pH below 4.

Ebbs et al. [18] showed that free UO_2^{+2} is the U species most readily taken up and translocated by plants. Because this species is only present at a pH of 5.5 or less, acidification of U-contaminated soils may be necessary for phytoextraction (Figure 29.1). The uranyl cation also binds to the soil solids and organic matter, reducing the extent of plant uptake [21]. Therefore, in addition to acidification, soil amendments that increase the availability of U by complexation may also be required.

Ebbs et al. [18] and Huang et al. [20] studied the role of acidification and chelating agents on the solubilization of uranium. Of the organic acids tested (acetic acid, citric acid, oxalic acid, malic acid), citric acid was the most effective for increasing U in the soil solution. The results also indicated that citric acid solubilized more U than simple acidification (HCl, S, HNO_3). Chelating agents like EDTA and DTPA did not increase uranium solubility significantly. Compared to potassium citrate, citric acid was much more effective for increasing U solubility and thus accumulation in plants. With the addition of 20 to 25 mmol citric acid per kilogram of soil, soil pH decreased 0.5 to 1.0 pH units, depending on initial soil pH.

Following citric acid treatment (20 mmol kg^{-1}) the U accumulation in Indian mustard (*Brassica juncea)* was increased 1000-fold [20]. Ebbs et al. [18] and Ebbs [8] observed an increase in U accumulation in beet (*Beta vulgaris)* of a factor of ten after citric acid treatment (25 or 20 mmol kg^{-1}). Citric acid was always applied 1 week before harvest by spraying a solution over the soil surface. The citric acid-triggered U accumulation in plants was observed within 24 h after application and a maximum was reached after 3 days. Thus, plants would maintain very low U concentrations for most of their growth and start accumulating rapidly following citric acid application, after which they could be quickly harvested. Citric acid is rapidly biodegradable, so no problems with residual levels of citric acid in soil should occur.

The potential to phytoextract uranium (U) from a sandy soil contaminated at low levels was tested in the greenhouse by Vandenhove et al. [22]. Two soils were tested: a control soil (317 Bq ^{238}U kg^{-1}) and the same soil washed with bicarbonate (69 Bq ^{238}U kg^{-1}). Ryegrass (*Lolium perenne* cv. *Melvina)*, Indian mustard (*Brassica juncea* cv. *Vitasso*) and redroot pigweed (*Amarathus retroflexus)* were used as test plants. Plants were selected on the basis of their reportedly high transfer factors and relative important dry-weight production. Citric acid addition resulted in a decreased dry weight production (all plants tested) and crop regrowth (ryegrass) (Figure 29.2).

The annual removal of the soil activity with the biomass was less than 0.1% for the control scenario. The addition of citric acid (25 mmol kg^{-1}) 1 week before the harvest increased U uptake up to 500-fold (Figure 29.2). With a ryegrass and mustard yield of 15,000 kg ha^{-1} and 10,000 kg

TABLE 29.4
Some Specific U-TF to Natural Vegetation and Garden Crops

Plant/crop/vegetation type		TF	Experimental conditions
Natural vegetation at U-milling site[a,b]			
Sagebrush		0.12	Background
		0.90	Edge of tailing pond
Mixed grasses		0.07	Background
		0.07	Edge of tailing pond
		0.69	Exposed tailings
Forbs		0.16	Background
		0.08	Edge of tailing pond
		1.1	Exposed tailings
Grasses[c]		0.003–0.18	Sandy and loamy soil
Vegetables			
Leaf vegetables	Lettuce[c]	0.025	Loam, pH 7
	Spinach[c]	0.033	Loam, pH 7
	Turnip green[a]	0.0058	Lake sediments, sandy, pH 4, 1.5% OM
	Indian mustard[e,f]	0.007	Loam, OM 4.2%, pH 7.7
Root crops	Carrots[c]	0.0057	Sandy loam, pH 7
	Potato flesh[c]	0.019	Loam, pH 7.3
	Potato flesh[c]	0.002	Sand, pH 4.9
	Potato[d]	0.009-0.0009	Sand, pH 8.1 (TF for fresh weight)
	Sugar beet[c]	0.01–0.06	Sand or sandy loam, pH 7
	Red beet[c]	0.0024	Loam, pH 5.1–7.0
	Turnip root[a]	0.00099	Lake sediments, sandy, pH 4, 1.5% OM
Fruit vegetables	Tomato[c]	0.0005	Sandy loam, pH 7
	Cucumber[c]	0.0009	Sandy loam, pH 7
Cereals	Bush bean[a]	0.0018	Lake sediments, sandy, pH 4, 1.5% OM
	Rice[c]	0.0005	Loam, pH 7.3
	Barley[c]	0.0021	Loam, pH 7.3
	Corn[c]	0.00021	Loam, pH 7.3
	Corn[a]	0.00021	Lake sediments, pH 4, 1.5% OM
	Corn[e]	0.006–0.01	Not specified
Amaranthus[g]		~0.007	

[a] Whicker, F.W. and Ibrahim, SA., Radioecological investigations of uranium mill tailings systems. Fort Collins, Colorado State University, 1984, 48.
[b] Ibrahim, S.A. and Whicker, F.W., *J. Radioanal. Nucl. Chem.,* 156(2), 253, 1992.
[c] Frissel, M.J. and van Bergeijck, K.E., Fifth Report of the IUR working group soil-to-plant transfer factors: report of the working group meeting in Guttannen Switzerland, Bilthoven, RIVM, 1989, 240.
[d] Lakshmanan, A.R. and Venkateswarlu, V.S., *Water, Air Soil Pollut.,* 38, 151, 1988.
[e] Mortvedt, J.J., *J. Environ. Qual,* 23, 643, 1994.
[f] Ebbs, S.D. et al., *J. Environ. Qual.,* 27, 1486, 1998.
[g] Huang, J.W. et al., *Environ. Sci. Technol.,* 32, 2004, 1998.

ha^{-1}, respectively, up to 3.5 and 4.6% of the soil activity could annually be removed with the biomass (Table 29.5). With the required activity reduction level of 1.5 and 5 for the bicarbonate-washed and control soil, respectively, it would take 10 to 50 years to attain the release limit.

A linear relationship was found between the plant ^{238}U concentration and the ^{238}U concentration in the soil solution of the control, bicarbonate-washed, or citric acid-treated soil. This points to the importance of the soil solution activity concentration in determining U uptake and thus to the importance of solubilizing agents to increase plant uptake.

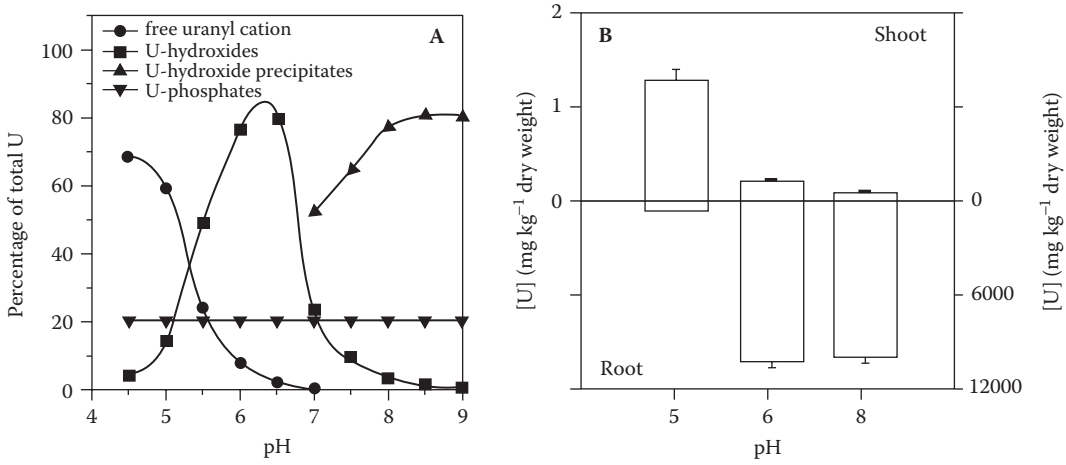

FIGURE 29.1 Dependence of U speciation and U uptake on pH. (After Ebbs, S.D. et al., *J. Environ. Qual.,* 27, 1486, 1998.)

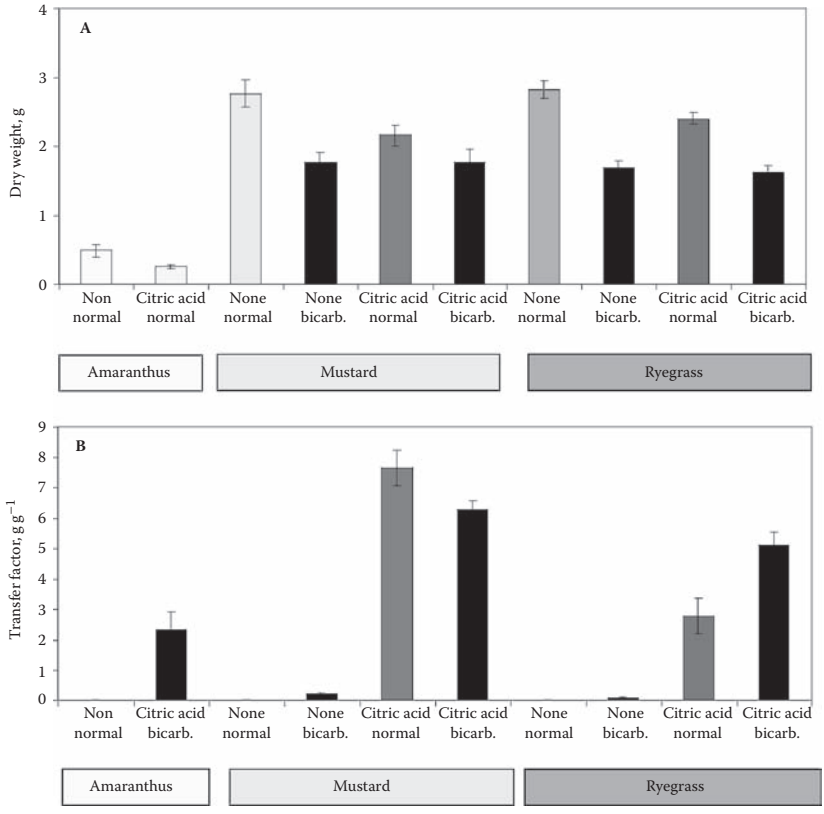

FIGURE 29.2 Effect of citric acid addition 1 week before harvest on the dry weight production and uranium transfer factor of ryegrass and mustard and redroot pigweed.

TABLE 29.5
Phytoextraction Potential[a] of Untreated and Citric Acid-Treated Soils (First Harvest)

Plant	Soil	Citric acid	Annual extraction %
Amaranthus	Control	No	0.0009 ± 0.0003
	Control	Yes	0.178 ± 0.058
Mustard	Control	No	0.010 ± 0.002
	Control	Yes	4.618 ± 0.384
	Bicarb.	No	0.103 ± 0.030
	Bicarb.	Yes	3.284 ± 0.250
Ryegrass	Control	No	0.007 ± 0.004
	Control	Yes	2.810 ± 0.689
	Bicarb.	No	0.052 ± 0.008
	Bicarb.	Yes	3.477 ± 0.474

[a] Annual removal, %.

Notes: Soil depth 10 cm; soil density 1.5 kg dm^{-3}.

Addition of citric acid is required to attain extraction levels that may make phytoextraction a feasible remediation option. Important uncertainties affect the phytoextraction potential:

- How does plant growth stage affect the efficiency of citric acid addition and what is the expected yield at the growth stage of maximal accumulation?
- What is the optimal level of citric acid addition to attain the highest increase in U-TF and the lowest impact on yield?
- What is the effect of continuous citric acid treatment on the soil?
- What is the global cost of phytoextraction compared to other remediation options?

Given the deleterious effect of citric acid on plant growth, one should possibly revert to other soil additives to increase the phytoavailability. However, several authors [18,20] have screened a number of additives, and only citric acid was found to be effective in increasing the TF. Though the experimental conditions of the latter authors were similar to the experimental conditions used by Vandenhove et al. [22], they did not mention a significant decrease in dry weight production or plants' dying off after citric acid treatment. When equivalent amounts of citric acid at pH 7 (adjusted with KOH) were added, ryegrass regrowth was also hampered (Vandenhove and Van Hees, results not shown). This implies that plants' dying off following citric acid addition is not predominantly due to a low pH, but rather to an increase in ion strength of the soil pore water when nutrients are solubilized under the action of citric acid.

Apart from application of soil additives to increase U export with the plant biomass, plant selection may also be important for improving the phytoextraction potential. However, the expected effects are small compared to the increase in uptake following soil amendments like citric acid addition. Huang et al. [20] recorded a difference of a factor of two in TF for four varieties of Indian mustard (*Brassica juncea).* They also demonstrated that plant species and cultivars differed significantly in response to citric acid treatment. Of the species tested, four showed significant potential in citric acid-triggered U accumulation: Indian mustard (*B. juncea),* Chinese mustard *(B. narinosa),* Chinese cabbage *(B. chinensis)* and redroot pigweed (*A. retroflexus)*. A twofold difference between cultivars was also observed.

29.2.2.2 Radium

29.2.2.2.1 Behavior of Ra in Soil and Soil Factors Affecting Ra Availability

Radium is one of the prominent potential contaminants linked with industries extracting or processing material containing naturally occurring radionuclides. It (^{226}Ra) is a natural decay product of ^{238}U, an alpha emitter, and is present in the soil at concentrations of 37 to 370 Bq kg^{-1}. The radium concentration may reach critical levels — orders of magnitudes higher — in tailings from uranium mills, phosphate mines, and processing sites, as well as in various waste products from the NORM-industry.

Radium is the last member of the alkaline earth metals, a group of metals whose lighter members (Ca and Mg) play a very important role in plant growth and nutrition. Because radium is highly electropositive, it reacts readily with many agents, most of which are insoluble. It also coprecipitates with barium and strontium to form insoluble sulphates. Due to its basic character, it is not easily complexed. The most stable complexes are formed with EDTA [23].

Radium has a high affinity for the regular exchange sites of the soil. According to Simon and Ibrahim [24], organic matter adsorbs about ten times as much radium as clay, which is more adsorptive than other soil minerals. Increased exchangeable Ca [25], increasing pH [19], and high soil-sulphate content [12] are reported to decrease the radium transfer factor. Vasconcellos et al. [26], however, did not find a relation between soil Ca concentration and Ra TF.

29.2.2.2.2 Phytoextraction Potential of Ra

The transfer factor depends on soil characteristics, plant type, the part of the plant concerned, climate conditions, and the physicochemical form of radium. Compared to many other radionuclides, little information is available on the uptake of radium from soil. Among reported values, large discrepancies are noticeable for the Ra-transfer factors (TF: concentration in plants to concentration in soil). Conducting a field study in a high natural background area testing seven field and garden crops, Linsalata et al. [34] reported values ranging from 0.3 10^{-4} to 0.02 g g^{-1} (lower value for corn; higher value for carrot). TFs on loamy, sandy, and organic soils ranged from 0.05 to 0.44 g g^{-1} for clover and from 0.14 to 0.62 g g^{-1} for ryegrass (lowest value on loam and highest on organic soil) [27]. IUR [28] reported 95% confidence limits ranging from 2.9 10^{-4} to 0.12 g g^{-1} considering a large series of crops and soil types (Table 29.6).

Thus far, no information was found by the author on studies dedicated to the phytoextraction of radium or methods to promote radium availability in the soil–plant environment. Addition of EDTA or decrease in pH could be expected to decrease the radium transfer factor. Highest TFs for radium are around 0.1 g g^{-1}. Considering a crop yield of 20 t ha^{-1}, only 0.13% (see Table 29.2) of the radium could be annually extracted with plants.

29.2.2.3 Thorium

29.2.2.3.1 Behavior of Th in Soil and Soil Factors Affecting Th Availability

More than 99% of natural thorium exists in the form of ^{232}Th ($t_{½}$ 4.5 × 10^9 a). Thorium occurs naturally in the Earth's crust at an average background concentration of 8 to 12 mg kg^{-1}. The typical concentration range of Th in soils is 2 to 12 mg kg^{-1} with an average value of 6 mg kg^{-1} [29]. More than 120 minerals contain thorium and thorium compounds [30]. The major source of thorium is monazite, which is a component of different granites and other igneous rocks. The geochemical behavior of Th is very similar to the behavior of rare earth elements (especially cerium), zirconium, and uranium.

The geochemistry of thorium is simplified by the existence of just one valent state: +4. It has been shown [31] that the Th ion is largely hydrolyzed at pH above 3.2, and the hydroxyl complexes are involved in the sorption process. The adsorption of Th on clays, oxides, and organic matter increases with increasing pH and is completed at pH 6.5. On the other hand, it has been suggested

TABLE 29.6
Some IUR Recommended Values for Ra-TF for Different Crops and Soil Types and 95% Confidence Intervals[a]

Crop group	Soil Type	Recommended TF value	95% Confidence interval[b]
Cereals (maize)	Sand	0.00104	0.00048–0.00223
Tubers (potatoes)	Sand	0.00097	0.00077–0.00123
	Clay	0.00358	0.00029–0.04362
Brassicas (beans)	Sand	0.00458	0.00223–0.01057
	Clay	0.0210	0.0092–0.0479
Root vegetables (carrots)	Sand	0.00676	0.00321–0.0142
	Clay	0.0379	0.0123–0.1161
	Organic		
Grass	Sand	0.12	0.0692–0.2083
	Loam	0.0617	0.0410–0.0928
	Clay	0.0330	0.0243–0.0449

[a] TF: Bq kg^{-1} dry mass plant per Bq kg^{-1} dry mass soil.
[b] 95% confidence intervals of the mean (i.e., geometric mean ± t_n standard errors) (t_n ~2 if $n > 10$).

Source: IUR, Sixth report of the IUR working group soil-to-plant transfer factors: report of the working group meeting in Guttannen, Switzerland, 198924-26, Bilthoven, RIVM: 240, 1989.

[32] that mobility of Th in soil may be less affected by soil pH than by soil organic matter. Tetravalent thorium may be strongly complexed with soil organic matter, thus increasing the mobility of Th in soil.

29.2.2.3.2 Phytoextraction Potential of Th

Information on Th-TF is rather scant. Shtangeeva et al. [33] report TFs for Couch grass ranging from ~0.01 to 0.05 and for plantain from 0.008 to 0.03 g g^{-1} — the higher values for the shoots and the lower values for the roots. For garden crops cultivated on high background areas, Linsalata et al. [34] found values ranging from 10^{-5} to 10^{-3} g g^{-1}. Whicker et al. [35] measured values ranging from 2×10^{-5} to 2×10^{-3} g g^{-1} for crops grown on a contaminated lake bed. Only Vera Tome et al. [36] reported rather high Th-TF, ranging from 0.013 to 0.270 g $g^{-1}$for grass pasture samples collected in granitic and alluvial soils around a disused uranium mine.

As was said for radium, no information was found by the author on studies dedicated to the phytoextraction of thorium or methods to promote radium availability in the soil–plant environment. Because the highest TF for thorium observed is around 0.1 g g^{-1} (yet most TFs observed are a factor of 100 to 10,000 lower), the maximal amount annually extracted by plants is of the order of 0.1%.

29.2.2.4 Caesium

29.2.2.4.1 Behavior of Cs in Soil and Soil Factors Affecting Cs Availability

After the Chernobyl accident, the study of the fate of radiocaesium in the environment was of particular importance, given its relatively long half-life, its widespread contamination, and its similarity with K favoring its uptake by plants [37–40].

The soil K status affects Cs availability: in general, the higher the soil K is, the lower is the TF. This K effect is explained by the increased solution concentration of a cation, which competes with radiocaesium for uptake. The consensus now is that the solid/liquid partitioning of radiocaesium in the soils, and thus its soil bioavailability, is governed by the presence of micaceous, potassium-bearing clay minerals [41–43]. The process relates to the action of a small number of

TABLE 29.7
Annual Removal of ^{137}Cs for Some Agricultural Crops[a]

Crop	Yield (ton dm ha^{-1})	TF (g g^{-1})	Crop off-take (% of total in soil)
Cereals (grain)	5–7	0.0004–0.25	0.0005–0.06
Potato tuber	6–10	0.003–0.89	0.0006–0.3
Leafy vegetables	5–10	0.008–1.7	0.001–0.6
Grassland	10–15	0.01–1.0	0.007–1.0

[a] Expressed as fraction of total content in the plough layer (arable crops) or in the 0- to 12.5-cm layer (grassland).

Note: The range in dry weight-based transfer factors (TF) represents a typical range and is derived from Nisbet A.F. and Woodman R.F.M., *Health Phys.,* 78(3), 279, 2000.

Source: IUR, Eighth report of the working group soil-to-plant transfer factors, report of the Working Group Meeting in Madrid, Spain, IUR Pub R-9212-02, IUR Technical Secretariat, Balen, Belgium, June 1–3, 1992.

very selective sites — the so-called frayed edge sites (FES) — located at the edges of illite particles. In general, the higher the soil clay content is, the lower is the caesium availability.

In studies following the Chernobyl accident, lime was applied to reduce the radiocaesium TF through an increase in pH (extent of increase not reported) and transfer reductions of a factor of two were obtained [38,44]. Plant growth may, however, be reduced at lower pH and in reducing conditions (high water content). For example, the Cs-TF may be much higher on an organic soil, but yield may also be substantially reduced.

Enhanced uptake of caesium by plants is reported in the presence of increased amounts of NH_4 [7,45,46] or when K is depleted [47–49]. NH_4 additions increased the TF up to twofold and also enhanced biomass production; both factors favored the phytoextraction potential [50]. Extremely low soil fertility with regard to potassium may increase the radiocaesium TF 10- to 100-fold, but will also decrease plant growth. The rather high TF for ryegrass recorded by IUR [28], 3.3 g g^{-1}, may have been obtained for soil with low fertility.

Given the number of parameters that may possibly enhance the radiocaesium TF (low pH, high ammonium, low K), it may be concluded that increasing the TFs reported in literature two- to threefold is a possibility worthy of testing.

29.2.2.4.2 Phytoextraction Potential of Cs

In normal agricultural systems, the annual Cs flux is small compared to that present in soil (Table 29.7). The ^{137}Cs off-take values are all less than 1%; the highest off-take is found for grassland. The high sorption of ^{137}Cs in soil and the typical K levels in soil required for optimal plant growth limit high off-take values.

Table 29.8 presents radiocaesium TFs for sandy soils (generally higher TF) for some crops and natural vegetation. Most crops have a TF << 1 g/g, resulting in annual removal percentages of around 3%, decay included (Table 29.3).

Many crops may have a wide range of TFs, even for a specific soil type. For example, the range of TFs observed for ryegrass grown on sandy soil extends from 0.05 to 3.3 [52]; this means that TFs substantially higher than 1 are within reach. Potential ryegrass yield is between 15 and 20 t/ha as long as fertilization is adequate. This would mean a phytoextraction ranging between 5.3 and 6.3%, decay included (Table 29.2).

Using sandy Cs-contaminated soil (5 Bq g^{-1}) at a waste processing facility, Vandenhove [50] tested the effect of ammonium addition on the phytoextraction potential of two crops with reportedly high TF and DW production in a greenhouse experiment. Target was 1 Bq g^{-1} Cs. Ammonium

TABLE 29.8
Average Radiocaesium Soil-to-Plant Transfer Factors

Crop	Transfer factor (gg^{-1})
Winter wheat (leaves + grain)[a]	0.03
Summer wheat (leaves)[a]	0.05
Ryegrass[a]	0.03
Potato (tuber)[a]	0.09
Lettuce[a]	0.24
Yellow lupine (seeds)[b]	1.64
Yellow lupine (straw)[b]	1.02
Sunflower (seeds)[b]	0.15
Sunflower (straw)[b]	0.59
Sunflower (roots)[b]	2.88
Ryegrass[c]	1.2 (0.05–3.3)
Ryegrass[d]	1.87 (0.92–2.82)
Flax (leaves)[e]	0.66 (0.57–0.84)
Indian mustard[f]	0.4–0.5
Red root pigweed (amaranthus species)[f]	2.2–3.2
Tepary bean[f]	0.2–0.3
Different amaranthus species[g]	0.53–2.03

[a] Lembrechts, J., *Sci. Tot. Environ.*, 137, 81, 1993.
[b] Gopa, Belarus: study on alternative biodiesel sources in relation with soil decontamination. Project N° TACIS/REG93, 1996.
[c] IUR, Fifth report of the IUR working group soil-to-plant transfer factors: report of the working group meeting in Guttannen, Switzerland, 198924-26, Bilthoven, RIVM: 240, 1989.
[d] Vandenhove, H. et al., *Sci. Tot. Environ.,* 187, 237, 1996.
[e] Van Hees, M. and Vandenhove, H., Unpublished data, 1998.
[f] Lasat, M.M. et al., *J. Environ. Qual.,* 27, 165, 1998.
[g] Sorochinsky, B., in *Proc. Chernobyl Phytoremediation Biomass Energy Conversion Workshop.*, Slavutych, Ukraine, 1998, 229.

addition increased DW production with 20% and the TF with 80%, resulting in a TF of 0.8 g g^{-1}. With a realistic yield of 20 t ha^{-1} under field conditions, this would result in an annual reduction of 3.3% (decay included). This would imply 50 years of continued phytoextraction to reach the target of 1 Bq g^{-1}. Lasat et al. [55] observed that adding ammonium did not significantly affect the Cs-TF for three species (amaranthus, tepary bean, Indian mustard) (Figure 29.3).

Amaranthus species screened by Lasat et al. [55] and Sorochinsky [56] have TFs as high as 3.2 g g^{-1} with a yield potential projected at 30 t ha^{-1} a^{-1} (based on two harvests per year, which is very improbable). The TFs reported by Lasat et al.[55] were derived for a sandy loam from Hanford with contamination levels of 15 Bq g^{-1} soil; thus, the annual removal, including decay, would be 8.3% (Table 29.3). Their target was a fourfold reduction in soil activity (to 4 Bq g^{-1}), which would require 16 years. In the more likely event of a yield of 15 t ha^{-1}, the time required would be 28 years.

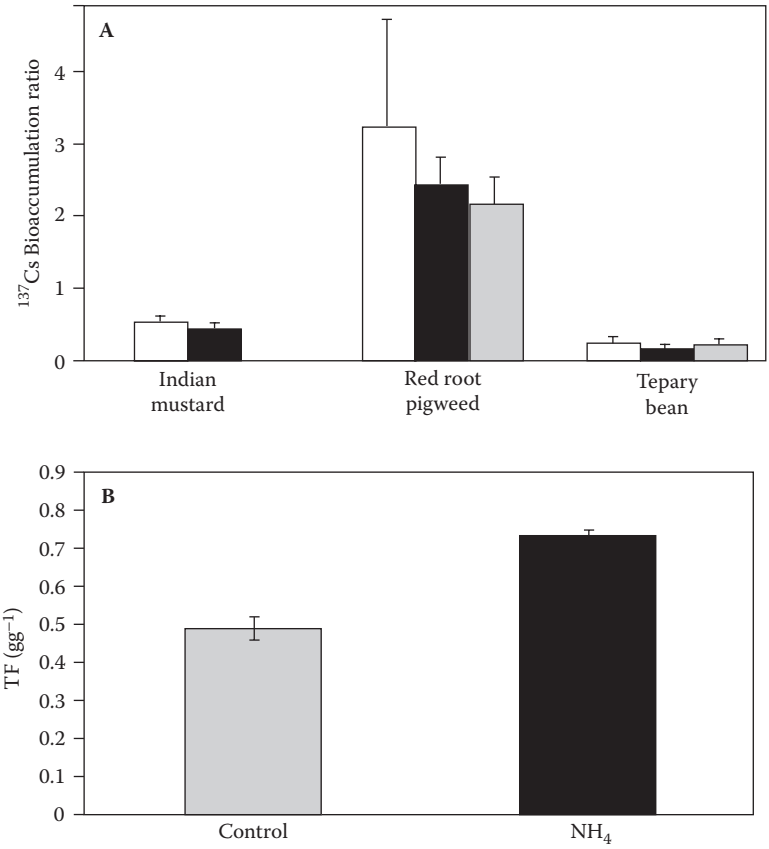

FIGURE 29.3 Effect of ammonium addition on the Cs-TF of Indian mustard, redroot pigweed, and tepary bean. (Sorochinsky, B., in *Proc. Chernobyl Phytoremediation Biomass Energy Conversion Workshop*, Slavutych, Ukraine, 1998, 229) and ryegrass (Vandenhove, H., Internal SCK•CEN report R-3407, 1999).

29.2.2.5 Strontium

29.2.2.5.1 Factors Affecting the Strontium TF

As radiocaesium, radiostrontium is among the most abundant radionuclides in the suite of nuclear fission products that are routinely or accidentally released. Its relatively long half-life (~30 years) and its metabolic similarity with Ca favor its uptake in plants. For the Sr-TFs, the TFs for each crop tend to be highest for sand and lowest for clay and organic soil types; the maximal differences between soil types for any one crop tend to be around one order of magnitude. The soil-exchangeable Ca content seems to affect the Sr-TF, but not significantly. Sauras et al. [58] found that lower Sr-TF was observed with increased soil solution Ca + Mg and increased CEC.

29.2.2.5.2 Phytoextraction Potential of Sr

The off-take of ^{90}Sr is higher than that of ^{137}Cs because the Sr availability is typically tenfold above that of Cs. The TFs of ^{90}Sr in green vegetables and *Brassica* are typically around unity and upper levels are around 10 (Table 29.9). With a yield of 10 ton/ha for leafy vegetables and a TF of 10, 9% could be annually removed, considering that contamination is restricted to 10 cm and accounting for decay. Under those conditions, a fivefold reduction of the soil contamination would require 15 years. Phytoextraction of ^{90}Sr has not yet been investigated at field scale. The high off-take values in agricultural crops (Table 29.9) suggest that this phytoextraction should be explored for this element.

TABLE 29.9
Annual Crop Off-Take of ^{90}Sr for Some Agricultural Crops[a]

Crop	Yield (ton dm/ha)	TF (g/g)	Crop off-take (% of total in soil)
Cereals (grain)	5–7	0.02–0.94	0.0037–0.22
Potato tuber	6-10	0.03–1.4	0.006–0.5
Leafy vegetables	5–10	0.45–9.1	0.07–3.0

[a] Expressed as a fraction of total content in the plough layer (arable crops) or in the 0- to 12.5-cm layer (grassland).

Note: The range in dry weight based transfer factors (TF) represents a typical range. The TFs of ^{90}Sr have been derived from Nisbet A.F. and Woodman R.F.M., *Health Phys.,* 78(3), 279, 2000.

Source: IUR, Eighth report of the working group soil-to-plant transfer factors, report of the Working Group Meeting in Madrid, Spain, IUR Pub R-9212-02, IUR Technical Secretariat, Balen, Belgium, June 1–3, 1992.

29.2.3 Conclusions for the Potential of Phytoextraction

Except for radiostrontium, TFs are generally too low to allow phytoextraction to be efficient without soil additives that increase bioavailability. Following citric acid addition, the uranium TF may increase with a factor of 500 resulting in annual activity reductions up to 3%; however, even then, it would still require more than 50 years to reduce the soil contamination level with only a factor of 5. Moreover, the effect of continuous citric acid treatment on the soil and plant growth and TF is not clear. For Cs, the same remark can be made. TFs are higher than the U-TF by a factor of 10 to 100. However, additives will only increase the availability with maximally a factor of two, without really affecting the DW production.

Given the low TFs observed for Ra and Th and the actual absence of an adequate method to increase the TFs, the feasibility of phytoextraction of Ra- and Th-contaminated soils is strongly questioned. The high off-take values in agricultural crops for Sr suggest that phytoextraction should be explored for this element. The question about long-term effectiveness still remains: will TF remain constant or will it decrease while radionuclide concentration decreases and when ageing processes occur?

There is the question of cost of waste treatment and site monitoring. With respect to this last issue, phytoextraction involves costs at different stages in the process. The soil must be prepared for crop establishment and the crop must be well maintained. During crop establishment or before the harvest, the soil may need to be treated with radionuclide-specific amendments to improve crop yield. The treatment of 1 m^3 of contaminated soil (10 m^2 if 1 dm soil layer) will annually result in about 10 to 20 kg biomass (~2 to 4 kg ashes), which must be harvested, transported, and treated; this entails considerable costs. This scheme of action must be repeated on a yearly basis for several years.

29.2.4 Rhizofiltration

Rhizofiltration is the use of plants to accumulate compounds from aqueous solutions through adsorption on the roots or assimilation through the roots and eventual translocation to the aerial

biomass (phytoextraction). Rhizofiltration is being investigated for the removal of radionuclides from aqueous waste streams, including groundwater and wastewater.

The removal of a radionuclide from an aqueous waste stream is governed by the plant dry weight production and the concentration factor, CF (ratio of Bq/g plant to Bq/ml water). Because absorption in (waste) water per volume is lower than in soil, the CF is higher than the TF. This becomes clear when considering the relationship between TF and CF:

$$TF = CF/K_D \tag{29.5}$$

where CF is the concentration factor (i.e., the ratio of the radionuclide activity concentration in plant shoots to that in soil solution) and K_D is the solid–liquid distribution coefficient of the radionuclide ($dm^3\ kg^{-1}$) (i.e., the ratio of radionuclide activity concentration in the solid phase to that in the soil solution).

The K_D for Cs in soils ranges from 10 to 10^5 ml g^{-1} [59]; for Sr, from 1 to 100; for U, the geometric mean K_D per soil group ranges from 35 to 1600 (full range from 0.03 to 395,100) [60]; and, for Ra, the average K_D per soil group ranges from 490 to 36,000 ml g^{-1} (full range from 1 to 1.810^7 [61]). The K_D for all radionuclides studied is generally substantially higher than 1, so it is clear that the CF exceeds the TF with the same factor and that rhizofiltration is generally more effective than soil phytoextraction.

Vasudev et al. [4] commented upon the removal of radionuclides from contaminated waste water and ground water. They found a 95% reduction in the U concentration in water through U absorption by sunflowers (*Helianthus annuus*) after a 24-h contact time. In a comparable experiment using Chernobyl-contaminated pond water, the Cs and Sr levels decreased 90 and 80%, respectively, after a 12-day contact time with 8-week-old sunflower plants replaced every 48 h. Sunflowers showed higher removal rates than timothy, meadow foxtail, Indian mustard, and peas (Cs-CF to shoots ranging from 400 to 2300 L kg^{-1}, Cs–CF to shoot from 860 to 8600 L kg^{-1}; for Sr, respectively, 650 to 2000 and 110 to 980 L kg^{-1}).

Dushenkov et al. [62] tested a number of species for the removal of uranium from water in a greenhouse experiment. Beans and mustard were less effective than sunflower in U removal. The latter removed 95% of the U in the water in less than 24 h. Practically all U was concentrated in the roots, with almost none in the shoots (Figure 29.4).

Concentration of uranium in the water did not affect the CF, but pH did: an average bioaccumulation coefficient for U in sunflower roots was 6624 and 3370 L/kg, respectively, at pH 5 and pH 7. As mentioned before, U accumulation is affected by U speciation. The U species most readily taken up by the plant is the uranyl form. This form predominates at pH below 5.5 [18]. Uranium was removed much faster from the contaminated water compared to Cs and Sr.

In natural waters, uranium is usually complexed with carbonate, hydroxide, sulphate, and phosphate. These complexes increase the solubility of U and make uranium precipitation more challenging. For this reason, it seems that U is one of the best candidates for a biological removal process.

However, Dushenkov et al. [62] demonstrated that the rhizofiltration method has its limits. Setting up an experiment with rather highly contaminated waste water (1000 µg L^{-1} U; 20% above the upper limit of local processing waste water) and high flow rate (1.05 L min^{-1}), 95% of the U was removed by 6-week-old sunflowers grown for 2 weeks in the waste water. This resulted in effluent concentrations of 40 to 70 µg/L^{-1}, above the 20 µg L^{-1} drinking water limit.

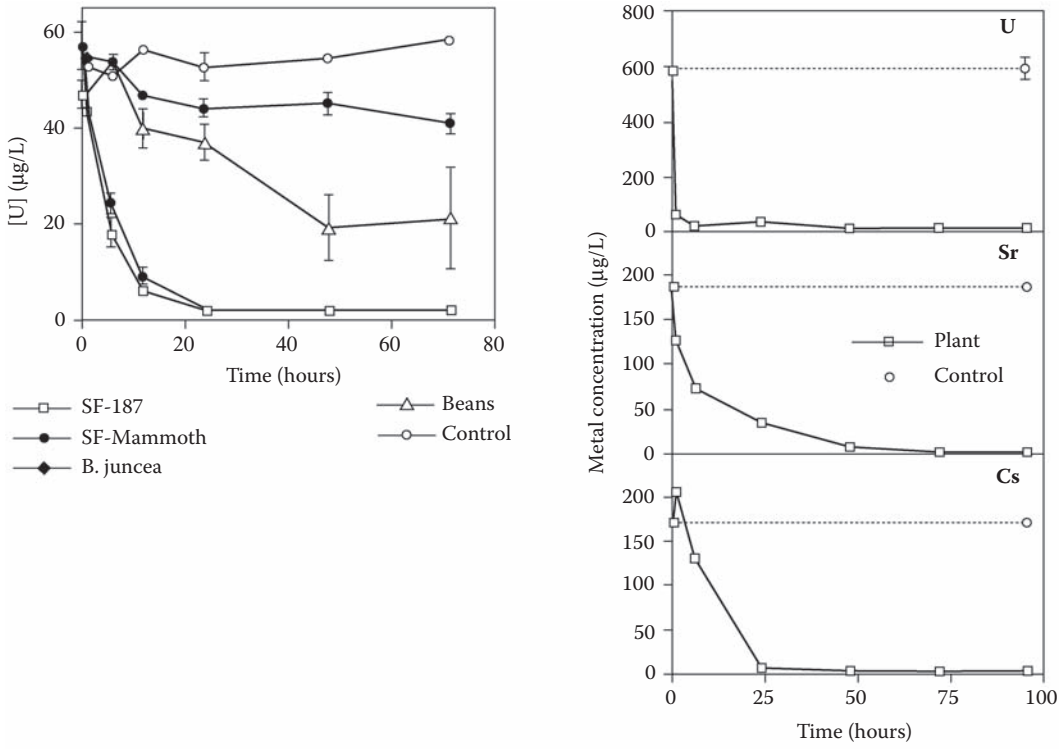

FIGURE 29.4 Removal of uranium by different sunflowers cultivars (left) and removal of Cs, Sr, and U by sunflowers (right) in a hydroponics system. (Dushenkov, S. et al., *Environ. Sci. Technol.,* 31, 3468, 1997.)

(*Disclaimer*: This section is certainly not exhaustive. No attempt has been made to perform a literature search for CFs for the radionuclides considered. Rather, a few figures are highlighted and the potential of rhizofiltration is discussed to some extent.)

29.3 ALTERNATIVE LAND USE: NONFOOD CROP PRODUCTION IN CONTAMINATED AREAS

29.3.1 Introduction

The question of how to manage large territories contaminated with a broad spectrum of radionuclides still remains 15 years after the Chernobyl accident. An area of 29,200 km^2 in Belarus, Ukraine, and Russia, was contaminated with levels exceeding 185 kBq ^{137}Cs m^{-2} [63] and as much as 4300 km^2 of agricultural land had to be excluded from use. The Sr contamination occurs mainly within a 70-km radius from the reactor, although some significant contamination (37 to 74 kBq m^{-2}) can be found in the area northeast of Gomel [64]. Belli and Tikhomirov [65] reported that, 9 years after the accident, the radiocaesium concentrations in plants grown in forests and on meadows did not significantly decline. In the more contaminated territories of Belarus, Ukraine, and Russia, no lifting of the restrictions on land use is likely in the foreseeable future.

When an appropriate countermeasure for a specific area is selected, apart from radiological criteria, the optimal solution to a given problem will depend as much on economic, social, and political factors as on sound scientific considerations [66]. Many studies have targeted possible agricultural countermeasures in response to concentration levels in food and agricultural crops that

are too high. Most studies have been conducted to test the effect of different physical and chemical countermeasures.

In contrast to this, information on long-term effects of countermeasures and, especially, the change to nonfood crops is still limited. Countermeasures can also be based on the selection of crops that exhibit smaller radionuclide uptake, on food processing, or choosing for nonfood crops such that the products from the land are radiologically acceptable [65,67,68]. Impact on dose to people and on the ecology and economy of the affected area may vary enormously: change in crop variety will have a much smaller impact than more radical changes such as substitution of vegetables by cereals or changing from an arable or cattle system to forestry.

- When an alternative crop or land use is advocated, the principle questions to be asked are:
- What is the fate of the radionuclide in the cultivation system and conversion routes and what is the expected radionuclide concentration in the end-products?
- How does the radionuclide behave during the biomass processing?
- What is the exposure during biomass cultivation and processing?
- How well are the crops adapted to the climate and soil conditions prevailing in the contaminated area?
- What are the conclusions with regard to economic feasibility for the production and use of these alternative crops?
- What prospective land use for large contaminated surfaces do these various alternative crops offer?

With respect to the fate of radionuclides in the cultivation system and conversion routes and the concentration to be expected in end- and waste products, one should have an idea of the entry radionuclide flux. This depends on the deposition level, the crop accumulation factor (which depends on plant and soil characteristics), and the radionuclide accumulation in the source material (e.g., wood, rape seeds, root beet, etc.). Whether the source material is safe for conversion (e.g., burning of wood or straw), the products are acceptable for use, and the waste products to be disposed of as radwaste or not depends on the radionuclide concentration and the exemption limits prevailing in each country.

29.3.2 Liquid Biofuels

Crops used for liquid biofuel production, e.g., rapeseed, wheat, sugar beets, barley, potatoes, and winter rye, may also be considered as suitable alternative crops. TFs to the useable product (rapeseed, wheat grains, beet root, barley, potatoes, and winter rye) are low (Table 29.10) and the liquid biofuels are almost free from activity. Also, radiocaesium levels in the waste products are generally of no concern.

Caesium levels in oil cake from oil seed rape (~2000 t ha^{-1}) and the pulp and vines from sugar beet (~4000 t ha^{-1}) may be too high for use as animal fodder and for incineration; they may need to be disposed of as radwaste. Yet, this is only in case of high contamination levels because only about 3% of the total radiocaesium and 6% of the total radiostrontium taken up will be potentially involved in the soil–plant–fodder–animal–man chain. Moreover, usually, fodder constitutes only 10% of oil cake and pulp and thus animal fodder can generally be used without restriction. The cost of liquid biofuels is actually (in general) about a few times to several hundred times the cost of fossil fuels, so a price subsidy is needed [53,71].

On the other hand, the production of rapeseed and the processing to edible rapeseed oil are profitable technologies. Furthermore, levels of caesium and strontium in the rapeseed oil after three filtrations and bleaching were below detection limit.

TABLE 29.10
Cs Transfer Factors[a] to Different Plant Parts of Some Potential Biofuel Crops

Crop	Plant component	Cs-TF, 10^{-3} m^2 kg^{-1}
Spring wheat	Leaves	0.23–0.42
	Straw	0.23–0.36
	Grain	0.08–0.16
Cereals	Grain	0.0048/26 (2.6–260)[b]
	Grain (peaty)	83(8.3–830)
Winter wheat	Straw	0.27–0.44
	Seeds	0.08–0.18
Spring rape seed	Stems and leaves	0.12 (0.46)[c]
	Straw	0.017–1.4[c]
	Seeds	0.03–0.04 (0.0006–0.5)[c]
Brassicaceae	Seeds	0.037–3.4[a]
Legumes	Seeds	94(12–750)[a]
Sugar beet	Root	0.43[d]
Root crops	Root	0.025/11 (1.1–110)[a]
Green vegetables	Leaves	0.07–4.86[a]
	Leaves (peaty)	260 (25–2700)[a]

[a] TF, g g^{-1}.
[b] Nisbet, A.F. et al., National Protection Board, NRPB-R304, Didcot, UK, 1999, 50.
[c] Gopa, Belarus: study on alternative biodiesel sources in relation with soil decontamination. Project no.°TACIS/REG93, 1996.
[d] IAEA (International Atomic Energy Authority), *Handbook of Parameter Values for the Prediction of Radionuclide Transfer in Temperate Environments*, Technical Report series no.°364, IAEA, Vienna, 1994.

Source: Data for Belarus from Grebenkov, A., Internal deliverables for RECOVER, 1997.

29.3.3 Willow Short-Rotation Coppice for Energy Production

The feasibility of willow short-rotation coppice (SRC) for energy production for revaluation of contaminated land was studied by Vandenhove et al. [71–74]. Coppicing is a method of vegetative forest regeneration by cutting trees at the base of the trunk at regular time intervals. Fast-growing species of the *Salix* genus (willows) are frequently used in a coppice system because of the ease of the vegetative reproduction and the large biomass production. The harvested biomass is converted into heat or power. As such, this nonfood industrial crop is a potential candidate for the valorization of contaminated land with restricted use.

In the case of reuse of contaminated land, SRC may be preferred to traditional forestry because revenues come sooner and more regularly (every 3 to 5 years) after establishment. SRC yields are also high on good agricultural soils, and it is not a drastic change in land use (easy to apply and easy return to food crops). SRC may also be considered as complementary to forestry, given the different culture requirements of both vegetation systems (forests perform well on sandy soils whereas SRC requires soils with sufficient water retention capacity). SRC has other potential advantages in a contamination scenario. Because it is a perennial crop, dispersion of radionuclides may be reduced. Harvest can be in winter when the soil may be covered by snow, resulting in protection for the people. Finally, SRC cultivation is not labor intensive, which is also advantageous with relation to exposure.

Willow SRC may be a suitable rehabilitation tool for highly contaminated land, but only if

- The radionuclide levels in the wood are below the exemption limits for fuel wood
- The average yearly dose received during coppice cultivation and coppice wood conversion is acceptable
- SRC can be grown successfully in the contaminated territories (soils, climate)
- The cultivation of SRC is technically feasible
- SRC production and conversion are economically profitable

It has been shown [71–74] that, for soils with a medium to high fixation (finer textured soils) and sufficient K, the TF (ratio of concentration in plant biomass to concentration in soil, $m^2\ kg^{-1}$) is $<10^{-5}\ m^2\ kg^{-1}$, and wood can be safely burned and the ashes disposed of without concern. Only in the case of light-textured soils with a low radiocaesium fixation and low soil K, the TF to wood is around $10^{23}\ m^2\ kg^{-1}$ and concentrations in wood may be elevated so that prevailing exemption limits are reached. Given that TFs for common forestry and for straw of winter wheat and oil seed rape are comparable, the same applies for burning wood or straw for energy.

Regarding the agrotechnical aspects affecting SRC production and feasibility, this crop has generally a high annual yield of about 12 t ha^{-1}. Sandy soils are only suitable for SRC production if well fertilized and irrigated. Regarding radiation exposure, doses in the vicinity of ash collectors may exceed the acceptable level of 1 mSv a^{-1} for a member of the general public only during the conversion phase and when highly contaminated wood is burned (3000 Bq kg^{-1}). Contributions from other possible exposure pathways are negligible (external exposure during culturing and transport, inhalation dose in the combustion plant and to the public following wood burning).

The economic sustainability of cultivation and conversion of SRC was evaluated by previously mentioned authors [71-74]. Crop yield and capital cost of the conversion units are among the most important parameters affecting system profitability. At the production site, a minimal yield of 6 t $ha^{-1}\ y^{-1}$ is required for the Belarus production conditions and 12 t $ha^{-1}\ y^{-1}$ for West European conditions, if all other parameters are optimal. Heat schemes may be a viable option for wood conversion in Belarus; electricity schemes are not. In Europe, subsidies are required to make wood conversion economically feasible. From this study it was also concluded that the existence of a contamination scenario does not necessarily hamper the economic viability of the energy production schemes studied. The cost associated with disposal of contaminated ashes was estimated as less than 1% of the biofuel cost [70,72,73] and would not affect the economic feasibility.

29.3.4 Forestry

Forestry can also be considered as an adequate alternative land use. Soil-to-wood TFs to coniferous and deciduous wood are around $10^{-3}\ m^2\ kg^{-1}$ [75] and thus comparable with the TFs to willow wood observed for soils with low fertility and limited Cs fixation. They are high compared with the TFs observed for willow in finer textured soils and soils with an adequate potassium status. Moreover, annual biomass increase is only 6 t ha^{-1} for forests and may attain 12 t ha^{-1} for SRC grown on soils with an adequate water reserve and fertility status. On these types of soil, SRC may thus be a more promising land-use option than traditional forestry. On soils with low water reserves (e.g., sandy soil), willow yield without irrigation is too low to be economically feasible, so forestry may be the preferred option here [71,72].

29.3.5 Fiber Crops

Fiber crops are also potentially alternative crops for agricultural land with restricted use. Potentially suitable crops are the annual fiber crops hemp (*Cannabis sativa* L.) and flax *(Linum usitatissimum* L.). Hemp and flax are well known arable crops that have been cultivated for centuries. Ukraine

has a legacy of flax and fiber hemp cultivation. In Belarus, there is only some flax production because, in the early 1990s, the acreage for production of flax and hemp declined dramatically in Ukraine. Establishment of fiber crops on contaminated arable land is generally of no radiological concern [76]. The TFs observed in the hemp fibers are higher than the TFs observed in flax by a factor of 4 to 50; cultivation should generally be restricted to areas that are not too contaminated (<1000 kBq m^{-2}). For both crops, contamination levels in the waste products (oil seed cake, chaff, ash after burning of straw) may, however, be high enough so that they should be considered as radioactive waste. The economics of this land use was not investigated.

29.3.6 Conclusions

Energy production from SRC in a contaminated area is a feasible remediation option on radiological, technicoagricultural, and economic grounds under certain conditions. On sandy soils, it is advisable to install forests or drought-resistant grasses instead of SRC or to apply irrigation because SRC is clearly not adapted to dry sandy soil conditions. For the other energy crops considered, generally no concern existed on radioecological grounds, but conversion to liquid biofuels is not profitable; therefore, the cultivation of crops for liquid fuel production cannot be recommended.

29.3.7 Phytostabilization

Phytostabilization reduces the risk presented by un- or sparsely vegetated contaminated soil by the use of plants and/or soil amendments to establish a stable vegetation cover that may progressively reduce the soil labile metal pool. This technology does not achieve a cleanup of the soil, but changes the mobility of potentially toxic elements by reducing concentrations in the soil water and other freely exchangeable sites within the soil matrix or by reducing re-entrainment of toxic particulates following the development of a stable and permanent vegetation cover. Both processes alter the speciation of soil metals, reducing potential environmental impact. These technologies draw upon fundamental plant and soil chemical processes as well as established agricultural practices.

The development of a stable and self-perpetuating ecosystem as a result of this type of treatment may be a further beneficial process because, in some circumstances, plant root activity may change metal speciation (changes in redox potential, secretion of protons, and chelating agents); the microflora associated with their root systems may produce similar effects. The rainwater infiltration rate can be reduced by increasing the plant-induced evapotranspiration, thus reducing the potential for leaching and acid drainage generating [77–80]. Plants will also physically stabilize topsoils [81]. Physical stabilization will reduce erodability and dust generation, thus reducing the mobility of radionuclides and respective exposure pathways. Some examples of phytostabilization approaches are presented next.

29.3.8 Phytomanagement with Willow Vegetation Systems in the Chernobyl Exclusion Zone

Remedial actions to control the radionuclide efflux from the Chernobyl Exclusion zone are still being investigated because most radioactive fall-out following the Chernobyl accident was deposited within the Dnieper catchment system, which adjoins the site of the Chernobyl nuclear power plant (ChNPP). This area and adjacent drainage basins form an extensive area from which contaminated water and sediments flow downstream through the Pripyat and Dnieper Rivers across the Ukraine to the Black Sea. Phytostabilization technologies could in this context also be considered as remedial options. Three phytorehabilitation approaches with willow vegetation systems for the Dnieper catchment system in the vicinity of the Chernobyl nuclear power plant were studied [72] (Figure 29.5): the effectiveness of willow vegetation on vertical migration of radionuclides; on the stabilization of the Chernobyl cooling pond sediments; and on horizontal erosion control.

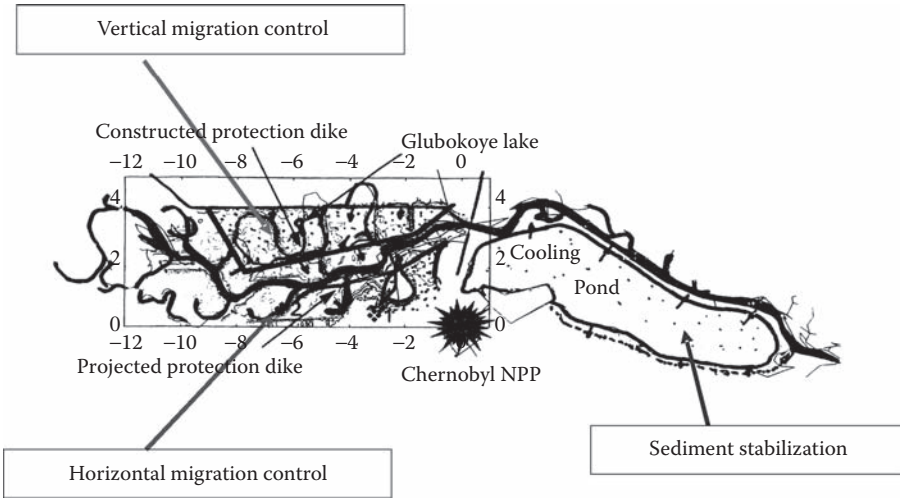

FIGURE 29.5 Phytostabilization approaches at the contaminated area of the Dnieper close to the Chernobyl NPPP.

The area of interest to study the vertical migration control by willow was an extremely contaminated zone of 16 km^2 at the left bank of the Pripyat (between 3.7 and 18.5 TBq km^{-2} ^{90}Sr and ^{137}Cs and 0.37 TBq km^{-2} Pu), which is partly protected from the spring floods by a dam. Through modeling exercises, it was shown that, due to their high evapotranspiration rate, willow SRC stands are expected to decrease the groundwater table level by 100 to 200 cm in fertilized stands. Without fertilization, only a decrease of groundwater table level of less than 50 cm was predicted. Because the immobilization potential of ^{137}Cs and ^{90}Sr in the willow wood is limited, the influence of plant uptake on migration remains low.

Following the effective closure of the ChNPP, the water level of the cooling pond (22.5 km^2; depth between 1.5 and 15 m, with about 111 TBq ^{137}Cs and 37 TBq ^{90}Sr) will drop with 4 to 7 m, and 15 km^2 of the sediments will come to surface and may be in need for stabilization. In this respect, the SALIMAT option was investigated. SALIMATs consist of a roll of willow rods (stems) rolled around a central disposable tube; these are unrolled by dragging them across the lagoon. Small tests have demonstrated that SALIMATs establish well on contaminated pond sediments and produce a full vegetation cover during the second year following establishment. The approximate cost of the phytostabilization option ranges from 0.8 to 1.9 10^6 EUR for the reclamation of 15 km^2 of sediments. This is a low cost compared with the prospective cost of removal of the sediments ($6,000,000, transport and disposal costs not included) or the maintenance of the present water level ($200,000 per year).

The projected area for the horizontal erosion control was the right bank of the Pripyat River, which was significantly less contaminated than the left bank, yet not protected with a dam. After inundation, part of the activity is eroded and transported to the Pripyat with the withdrawing water. It was calculated that, even in case of extremely high flooding, dense willow plantings will effectively decrease the horizontal soil erosion and concomitant transport of radionuclides into the Dnieper River system.

29.3.9 Uranium Mining Tailings and Debris Heaps

(Re)vegetation is a commonly employed measure on the capping of engineered waste disposal facilities and on mining residues such as spoil heaps [82,83] or tailings ponds. The final step in closing out an impoundment for uranium mill tailings is the design and placement of a cover that will give long-term stability and control to acceptable levels of radon emanation, gamma radiation,

erosion of the cover and tailings, and infiltration and precipitation into the tailings and heaps. Surface vegetation can be effective in protecting tailings or a tailing cover from water and wind erosion.

Factors affecting the effectiveness of surface revegetation on impoundments can be broadly classed into climatological and agrobiological factors. Plants should be chosen to match the local climate conditions. Concerning agrobiological factors, the nature of the ore and the mill processes will largely determine the tailing characteristics from the point of view of sustaining growth. Considerable efforts to correct adverse characteristics, such as low or high pH values and low plant nutrient content, will usually be required before tailings can sustain growth. Depending on the substrate, revegetation requires preparation and amelioration of the topsoil to remove, for instance, acid-generating minerals [84,85]. Techniques and strategies to overcome such difficulties have been developed [86] — for instance, hydroseeding or the use of compost from organic household refuse [87]. The method may be limited to low contaminant concentrations owing to the (root) toxicity of higher concentrations. An adequate soil cover may need to be established.

Water and wind erosion are the primary causes of erosion of tailings or tailing cover material. A vegetation cover may decrease the erosion hazard. However, vegetation surfaces may raise concerns: the vegetation can promote radon emanation by drying out the tailing or tree roots may penetrate the contaminated material and break the cover integrity. Given the increased evapotranspiration rate and interception of precipitation following vegetation establishment, the vegetation cover alters the water household of the tailings and may decrease seepage. The effect of a vegetation cover on the radionuclide dispersion through an alteration of the water balance and also potentially because of the effect of plant roots on the physicochemical characteristics of the tailing material (biologically driven acidification of the tailing material) has not been studied intensively thus far.

For a 35-year-old reclaimed site on a uranium mining dump near Schlema (Saxony, Germany), it was concluded that the biomass could reduce infiltration by 40 to 60% due to interception by the canopy (25 to 40%) and increased transpiration [88]. Of the 165,000 g ha^{-1} U in soil (30 cm depth), only 4 g ha^{-1} was in the above-ground plant parts and 510 g ha^{-1} in the below-ground plant parts. Of the uranium taken up during the growing season, 90% is recycled (returned) with the needles. U-dispersion by uptake through vegetation is thus minimal. It may be concluded from these preliminary results that forest vegetation may reduce infiltration rate and will not favor radionuclide dispersion.

Kistinger et al. [89] evaluated some design criteria for tailing coverage regarding vegetation aspects. Because the plant roots can penetrate the compacted sealing layer (trees have roots up to 3 to 4 m) and because the trees should also have a certain degree of mechanical support in order to minimize probability of uprooting, a vegetation substrate depth of at least 1.5 m is required. The vegetation substrate layer must be such that the critical suction is not exceeded at the top of the clay seal. It must be thick enough for plants to find sufficient water and nutrients not to generate high suction at the seal. Cracks resulting from such suctions become accessible to roots and can be widened as further water is extracted.

29.4 CONCLUSIONS

Different phytomanagement options may be applied for the remediation of radioactive contaminated sites. For vast contaminated surfaces, only the phytostabilization option and alternative land use options seem feasible options. The effect of the vegetation cover on the radionuclide dispersion and the subsequent dose should be thoroughly studied. Similarly, the effectiveness of alternative land use should not be studied on mere radiological grounds only. Socioeconomic factors are equally important.

Phytoextraction for soil cleanup seems only reasonable for limited surfaces and will only be effective under very specific conditions. Side effects (costs, treatment of contaminated biomass, and potential for ground water contamination) should be evaluated in depth.

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