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“Identification and Validation of Heavy Metal and radionuclide Hyperaccumulating  
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Enclosed is a copy of a manuscript we are submitting to the Journal of Environmental Quality on our research on the development of phytoremediation technologies for radiocesium contaminated soils. The acknowledgment of DOE support is on page 2 of the manuscript.

**Title: Potential for phytoextraction of  $^{137}\text{Cs}$  from a contaminated soil**

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1      **Abstract**

2      Potential for phytoremediation of an aged radiocesium-contaminated soil from Brookhaven  
3      National Laboratory was investigated in three phases: 1) hydroponic screening for plant species  
4      capable of accumulating elevated levels of cesium in shoots, 2) amending contaminated soil to  
5      enhance  $^{137}\text{Cs}$  bioavailability, and 3) phytoextracting radiocesium with plant roots and its removal  
6      in harvested shoots.

7              The bioaccumulation ratio of Cs in shoots of hydroponically grown plants ranged between  
8      38 and 165. From solution, dicot species accumulated 2- to 4-fold more cesium in shoots than  
9      grasses. The effect of several chemical compounds on  $^{137}\text{Cs}$  desorption from the contaminated  
10     soil was investigated. Ammonium salts were the most effective at desorbing Cs from  
11     contaminated soil, but only 25% of radiocesium could be desorbed. Although release of  
12     radiocesium from the soil was concentration-dependent, this effect appeared to level off above 0.2  
13     M ammonium in solution. In a pot study, from the soil contaminated with 400 pCi g<sup>-1</sup> soil, the  
14     greatest amount of  $^{137}\text{Cs}$ , 140 pCi, was removed in shoots of cabbage (*Brassica oleracea* var.  
15     *capitata*).  $^{137}\text{Cs}$  accumulation in shoots was significantly increased by the addition of 40 NH<sub>4</sub>NO<sub>3</sub>  
16     kg<sup>-1</sup> soil. Increasing NH<sub>4</sub>NO<sub>3</sub> application from 40 to 80 mmoles kg<sup>-1</sup> soil did not further increase  
17     radiocesium phytoextraction. The ability to accumulate radiocesium from soil in shoots was  
18     significantly different among species tested. This ability increased in order: reed Canary grass  
19     (*Phalaris arundinacea*) < Indian mustard (*Brassica juncea*) < tepary bean (*Phaseolus acutifolius*)  
20     < cabbage.

1      **Introduction**

2      Soils have become contaminated with radionuclides as a result of above ground nuclear  
3      testing, accidental release or nuclear energy generation.  $^{137}\text{Cs}$  is a long-lived ( $t_{1/2}$  30.2 years) by-  
4      product of nuclear fission. Although recently contamination of the environment with  $^{137}\text{Cs}$  from  
5      the testing of nuclear weapons has been drastically reduced, large areas are still polluted with  
6      radiocesium. For example, in the Republic of Belarus about 1 million hectares of arable land were  
7      contaminated with  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$  by Chernobyl nuclear accident (Kavkhuta et al., 1994).

8      Decontamination of these polluted-soils remains an intractable problem, in part because of the  
9      high cost. Recently, the U.S. Department of Energy Assistant Secretary for Environment  
10     Restoration and Waste Management was quoted as saying that the \$200 to \$300 billion cost for  
11     the cleanup of radionuclide-contaminated soils may be drastically underestimated (Watson et al.,  
12     1993).

13     Phytoremediation is emerging as an attractive alternative to energy-intensive, high-cost  
14     traditional cleaning methods. This new technology employs the use of higher plants capable of  
15     accumulating high levels of contaminants in shoots. Contaminants are removed by harvesting the  
16     above-ground plant tissues. In addition of being cost effective (Salt et al., 1995),  
17     phytoremediation decontaminates the soil in situ thus preserving both the environment and the  
18     ecosystems. Because of these advantages, phytoremediation has been cited as the method of  
19     choice for the clean up of large areas polluted with low to moderate levels of contaminants (Baker  
20     et al., 1994).

21     Numerous studies have reported on the potential of higher plants to accumulate

1       <sup>137</sup>Cs in shoots (Dahlman et al., 1975; Salt et al., 1992; Entry et al., 1993; Demirel et al., 1994).

2       These studies were conducted with soils of different physicochemical properties and  
3       contamination characteristics which both affect cesium uptake into plants. Therefore the available  
4       information is of limited use when planning to phytoremediate sites with different soil  
5       characteristics.

6       Earlier work indicated that upon entry into internal plant tissues radiocesium is fairly  
7       mobile (Resnik et al., 1969). However, phytoremediation of radiocesium-contaminated sites is  
8       impeded by the marked capacity of soils to tightly adsorb Cs (Nishita et al., 1968). In part, this  
9       results from the trapping of Cs in layer silicate minerals (Cremers et al., 1988). Desorption from  
10      soil particles is an important step toward increasing cesium bioavailability and consequently its  
11      uptake into roots. Although earlier reports indicate that radiocesium desorption can be chemically  
12      induced (Field et al., 1993; Gombert, 1993) the extent of this release was shown to be highly  
13      dependent on soil properties (Francis and Brinkley, 1976; Kirk and Staunton, 1989).

14       In this study, we analyzed the potential for phytoremediation of a <sup>137</sup>Cs-contaminated soil  
15      from Brookhaven National Laboratory (BNL). The work was conducted in three phases: 1)  
16      hydroponic screening for high-biomass-producing plants capable of accumulating elevated levels  
17      of cesium in shoots, 2) identification of chemical treatments capable of desorbing <sup>137</sup>Cs from the  
18      soil, and 3) evaluation of <sup>137</sup>Cs extraction in shoots of selected species grown in pots with  
19      contaminated soil.

1      **Material sand Methods**

2      *Hydroponic screening* was conducted in a greenhouse at ambient temperature (12°-20°C)  
3      with natural illumination supplemented daily with 10 h of artificial light. Two to three seeds of  
4      Indian mustard [*Brassica juncea* (L.) Czern], Arcadia, a commercial variety of broccoli (*B.*  
5      *oleracea* var *botrytis* L.), Storage #4, a commercial variety of cabbage (*B. oleracea* var *capitata*  
6      L.), Snow crown, a commercial variety of cauliflower (*B. oleracea* var *botrytis* L.), kochia  
7      [*Kochia scoparia* (L.) Schrad], tepary bean (*Phaseolus acutifolius* A. Gray), hairy vetch (*Vicia*  
8      *villosa* Roth.), colonial bent grass (*Agrostis capillaris* Sibth), red fescue (*Festuca rubra* L.), and  
9      reed Canary grass (*Phalaris arundinacea* L.) were germinated on discs cut of 1 mm nylon mesh.  
10     Discs were placed at the open bottom of polyethylene cups mounted in holes drilled in lids of 2-L  
11     containers (four cups/lid). The composition of the culture solution was; 1.2 mM KNO<sub>3</sub>; 0.4 mM  
12     Ca(NO<sub>3</sub>)<sub>2</sub>; 0.1 mM NH<sub>4</sub>H<sub>2</sub>PO<sub>4</sub>; 0.2 mM MgSO<sub>4</sub>; 50 µM KCl; 12.5 µM H<sub>3</sub>BO<sub>3</sub>; 0.1 µM NiSO<sub>4</sub>;  
13     1.0 µM ZnSO<sub>4</sub>; 0.5 µM CuSO<sub>4</sub>; 1.0 µM MnSO<sub>4</sub>; 0.1 µM H<sub>2</sub>MoO<sub>4</sub>; 10 µM Fe<sup>3+</sup>-EDDHA (for  
14     dicot) or Fe<sup>3+</sup>-HEDTA (for grasses); 1.0 mM Mes-Tris (pH 6.0) and 1.0 µM CsCl. After seed  
15     germination, discs were covered with a layer of polyethylene black beads to reduce illumination of  
16     the nutrient solution and provide support to emerging seedlings. Nutrient solution was replaced  
17     every week. Shoots of four-week-old plants were harvested and dried for 3 days at 65°C. Dry  
18     shoots were ground, weighed, and a 0.05-g aliquot was digested overnight in 1 mL of doubled-  
19     distilled HNO<sub>3</sub> in a heat block at 120°C. Subsequently, samples were dissolved in 0.75 mL of  
20     HNO<sub>3</sub>:HClO<sub>4</sub> (1:1, v/v) and incubated at 220°C until dry. Samples were then redissolved in 10  
21     mL of 5% HNO<sub>3</sub> and analyzed for cesium content with an Elan-Sciex-250 Analyzer via

1 inductively-coupled argon plasma mass spectroscopy.

2 *Study of the <sup>137</sup>Cs desorption from contaminated BNL soil*

3 The loamy sand soil obtained from Brookahven National Laboratory (BNL) was cleaned  
4 of gravel and organic debris before being passed through a 2-mm sieve. The < 2 mm soil fraction  
5 was air-dried and stored for further analyses. Desorption of the <sup>137</sup>Cs from the soil was  
6 investigated using 0.05 and 0.5 M solutions of the following extractants: 1) weak organic acids:  
7 acetic, citric, formic, malic, or oxalic, 2) strong acid: hydrochłoric and nitric acid, 3) reducing  
8 agents: Na-dithionite or hydroxylamine/hydrochloric acid, 4) neutral ammonium salts: chloride,  
9 sulfate, or nitrate, 5) buffered ammonium salts: modified Morgan's solution [0.7 M acetic acid  
10 plus 1.35 M ammonium acetate (Morgan, 1941)], or a modified Morgan's extractant containing  
11 0.02 M Na<sub>2</sub>-EDTA (sodium salt of ethylene diamine tetracetic acid) (Lakanen and Erviö, 1971),  
12 and 6) neutral potassium salts: chloride, nitrate, or sulfate. A 15-g aliquot of the <2 mm soil  
13 fraction and 45 mL of the chemical extractant were mixed in a 125-mL Erlenmeyer flask and  
14 agitated at room temperature in a rotary shaker at 150 rotations min<sup>-1</sup>. After 2 h, the slurry was  
15 filtered through a Whatman No. 42 filter paper. The resulting filtrate was vacuum-passed through  
16 a 0.45 µm Millipore membrane. The final filtrate was collected in a 20-mL counting vial and  
17 radioactivity was measured using a gamma counter (model 5530, Packard, Downers Grove, IL).

1      *Evaluation of <sup>137</sup>Cs phytoextraction from contaminated soil*

2              Before starting the pot study, the pH of the strongly acidic BNL soil was raised by adding  
3      3 g of Ca(OH)<sub>2</sub> to 20 kg of soil. Ten days later, soil pH had increased from 3.8 to 5.4. This pH  
4      was more suitable for plant growth, and because soil pH in the 3.9-8.4 range was reported to have  
5      little effect on cesium accumulation in plants (Fredericksson et al., 1966) we felt confident that pH  
6      correction would not be detrimental to <sup>137</sup>Cs transfer from soil to shoots. Twelve pots containing  
7      1 kg of soil were each seeded with two pregerminated seeds of cabbage, Indian mustard, reed  
8      Canary grass, and tepary bean and placed in a greenhouse at ambient temperature (18°- 27°C) and  
9      illuminated with natural light. Pots were watered with a nutrient solution with composition  
10     similar to that described above but containing only 300  $\mu$ M KNO<sub>3</sub>. We minimized the K<sup>+</sup>  
11     concentration because it is known that K<sup>+</sup> competes with Cs for uptake into roots (Shaw and Bell,  
12     1991) and because the differences in <sup>137</sup>Cs uptake rates between plant species were shown to be  
13     greater at low potassium concentrations (Buyssse et al., 1996). The twelve pots were divided into  
14     four sets of three pots. After 4 weeks of growth, one pot in each set received 13 mmoles of  
15     NH<sub>4</sub>NO<sub>3</sub> as 100 mL of a 130 mM NH<sub>4</sub>NO<sub>3</sub> solution. The second received 27 mmoles of NH<sub>4</sub>NO<sub>3</sub>  
16     as 100 mL of a 270 mM NH<sub>4</sub>NO<sub>3</sub> solution. The third pot from each set received only 100 mL of  
17     tap water. These treatments were repeated 3 and 6 days later. Thus, each of the four sets  
18     contained one pot which received: water (control), 40 mmoles of NH<sub>4</sub>NO<sub>3</sub>, and 80 mmoles of  
19     NH<sub>4</sub>NO<sub>3</sub>. Six days after the last treatment the shoots were harvested and plant material dried at  
20     65°C for 3 days. Dry shoots were weighed, ground and <sup>137</sup>Cs activity was measured via gamma  
21     activity detection.

1        *Statistical analysis*

2        The design of the pot experiment was a two-factorial (four plant species x three soil treatments)  
3        replicated in four completely randomized blocks. Data were subjected to ANOVA with soil  $\text{NH}_4^+$   
4        applications and plant species as the two experimental factors. The differences among treatments  
5        were compared with Fisher's least significant difference ( $p < 0.01$ ).

6        **Results and Discussion**

7        *Hydroponic screening*

8        Hydroponic screening allowed us to test a large number of plant species for their ability to  
9        accumulate cesium in shoots from a nutrient solution containing  $1 \mu\text{M}$   $\text{CsCl}$ . The greatest amount  
10      of cesium accumulated in shoots of cabbage followed by tepary bean (Table 1). Among grasses,  
11      reed Canary grass accumulated the most cesium. Bioaccumulation ratios ( $[\text{Cs}]_{\text{shoot}}/[\text{Cs}]_{\text{solution}}$ ) of  
12      the species tested ranged between 38 and 165. Compared to grasses, the values of  
13      bioaccumulation ratio were 3- to 4-fold greater in shoots of dicot species. Comparable values  
14      were reported for  $^{137}\text{Cs}$  accumulation in shoots of wheat (*Triticum aestivum* L.) (Smolders and  
15      Shaw, 1995). Values of bioaccumulation ratio significantly greater than 1 suggest that when  
16      available cesium is readily taken up plants.

17        Based on the results of hydroponic screening we selected cabbage, Indian mustard, reed  
18      Canary grass, and tepary bean to be tested in pots.

1      *Study of  $^{137}\text{Cs}$  desorption from BNL soil*

2      The major factor limiting  $^{137}\text{Cs}$  absorption by plants is likely to be its low bioavailability in  
3      soil. Cesium ions are selectively absorbed at the frayed edges or trapped into interlayer voids of  
4      micaceous minerals. Vermiculite (Sawhney, 1965) and illite (Tamura, 1964) are two common  
5      micaceous silicates found in soil that exhibit high affinity for cesium. Fixation of  $^{137}\text{Cs}$  by  
6      micaceous clays and by soils containing these clays has been demonstrated in desorption studies.  
7      For example, only 18% of the radiocesium could be displaced by 1 M sodium acetate from an  
8      illitic soil 11 days after the treatment (Dahlman et al., 1975). However, 87% of the  $^{137}\text{Cs}$  was  
9      desorbed from a soil containing predominantly non-expanding kaolinitic clays. The poor  
10     desorption from the illitic soil was interpreted as being caused by strong cesium fixation at  
11     selective binding sites.

12     In this study we investigated the effect of several chemical compounds on  $^{137}\text{Cs}$  desorption  
13     from the BNL soil. We used ammonium and potassium salts to desorb the exchangeable  
14     radiocesium soil fraction. Because Entry et al. (1996) suggested that synthetic chelates could  
15     enhance the release of  $^{137}\text{Cs}$  making it more accessible for plant uptake we also extracted the BNL  
16     soil with a  $\text{Na}_2\text{-EDTA}$  solution. Organic acids were tested because previous work indicated some  
17     success with these compounds (Field et al., 1993). Strong mineral acids hydrochloric and nitric  
18     acids were used to investigate the direct effect of  $\text{H}^+$  on  $^{137}\text{Cs}$  desorption. Reducing agents such as  
19      $\text{Na-dithionite}$  and hydroxylamine/hydrochloric acid were employed to release a  $^{137}\text{Cs}$  fraction  
20     potentially associated with iron or manganese oxides.

21     Finston and Kinsley (1961) noted that the ions  $\text{NH}_4^+$ ,  $\text{K}^+$ , and  $\text{Cs}^+$  form a homologous  
22     series with a great degree of physicochemical similarities. Therefore  $^{137}\text{Cs}$  displacement from the

1 soil is expected to increase following addition of monovalent cations. Jackson et al., (1965)  
2 reported that  $^{137}\text{Cs}$  mobility in soil increased with the addition of the monovalent cations in the  
3 following order:  $\text{Rb}^+ > \text{NH}_4^+ > \text{K}^+$ . Similar findings were reported by Tensho et al. (1961).  
4 However, Livens and Loveland (1988) reported greater radiocesium mobility under low  $\text{K}^+$  or  
5 high  $\text{NH}_4^+$  in soil. Although it is generally agreed that  $\text{NH}_4^+$ , and  $\text{K}^+$  are important for  $^{137}\text{Cs}$   
6 mobility in soil, the effect of these ions on radiocesium uptake into roots is less obvious . For  
7 example, as a result of enhanced bioavailability, greater cesium uptake into plants was reported  
8 following ammonium and potassium addition (Schultz et al., 1965). Shaw and Bell, (1991),  
9 however, described a competitive inhibition of Cs uptake into roots by any ion of the homologous  
10 series.. Clearly, there is some confusion as to the effect of  $\text{NH}_4^+$  and  $\text{K}^+$  ions on radiocesium  
11 mobility in soil and its uptake into roots. Furthermore, because some of these earlier studies were  
12 conducted with experimentally contaminated soils (binding to soil particles not completed), it is  
13 difficult to apply their findings to a soil contaminated with aged radiocesium (binding to soil  
14 particles completed) such as BNL soil.

15 Desorption of  $^{137}\text{Cs}$  from the soil following chemical treatments is shown in Table 2.  
16 Ammonium salts of strong acids at 0.5 M desorbed about 25% of the soil  $^{137}\text{Cs}$ . Somewhat less  
17  $^{137}\text{Cs}$  (17%) was desorbed from the soil with 1 M ammonium acetate with or without EDTA.  
18 These results indicate that EDTA had little effect on radiocesium desorption from soil.. Compared  
19 to  $\text{NH}_4^+$ , approximately 40% less radiocesium was desorbed with  $\text{K}^+$  salts (Table 2, Fig. 1).  
20 Despite successes reported in the literature (Field et al. 1993), extraction of  $^{137}\text{Cs}$  with organic  
21 acids was totally ineffective. Only small reductions in soil  $^{137}\text{Cs}$  activity were obtained with strong  
22 mineral acids (2-3%) or reducing agents (1-7%). Our results show that most of the  $^{137}\text{Cs}$  was

1 desorbed from the BNL soil by exchange (25% of total radiocesium in the soil).

2 Desorption of  $^{137}\text{Cs}$  by monovalent cations was concentration-dependent (Fig. 1); it  
3 increased with cation concentration up to 0.2 M. Further increases in concentration did not  
4 significantly increase  $^{137}\text{Cs}$  desorption. It is possible that after a 2-h incubation in a 0.2 M solution,  
5 equilibrium was reached between monovalent cations and a rapidly exchanging  $^{137}\text{Cs}$  soil fraction.

6 *Phytoextraction of  $^{137}\text{Cs}$  from contaminated soil*

7 We investigated the potential for radiocesium removal in shoots of four plant species  
8 grown in pots with untreated (control) or  $\text{NH}_4^+$ -treated BNL soil. The effect of soil treatment on  
9  $^{137}\text{Cs}$  shoot concentration and shoot biomass is shown in Fig. 2. Concentration of  $^{137}\text{Cs}$  was  
10 greater in shoots of the plants grown in ammonium nitrate-treated soil. Compared to control (no  
11  $\text{NH}_4\text{NO}_3$ ), addition of 80 mmoles increased  $^{137}\text{Cs}$  concentration in shoots from 135% (Indian  
12 mustard) to 1200% (reed Canary grass).

13 Although ammonium addition increases radiocesium desorption from soil particles, other  
14 mechanisms have also been proposed to account for the ammonium-induced increase in  $^{137}\text{Cs}$   
15 mobility and subsequently its uptake in roots. Thus, Kavkhuta et al. (1994), proposed that  $^{137}\text{Cs}$  is  
16 retained in soil via two distinct processes: 1) specific sorption to soil minerals and 2) radionuclide  
17 incorporation in insoluble particles of fuel origin where  $^{137}\text{Cs}$  is trapped by a cover of uranium  
18 dioxide. These authors proposed that  $\text{NH}_4\text{NO}_3$  addition stimulates the activity of soil  
19 microorganisms involved in the breakdown of the uranium cover increasing bioavailability of this  
20 radiocesium fraction. However because of the short period of time which elapsed between  
21 ammonium application and shoot harvesting, it is unlikely that the latter mechanism may have

1 played a significant effect on  $^{137}\text{Cs}$  uptake into roots.

2 Regardless of the soil treatment, the greatest level of radiocesium was concentrated in  
3 shoots of cabbage. From the soil treated with 80 mmoles  $\text{NH}_4\text{NO}_3$ , cabbage concentrated  
4 approximately 1100 pCi  $\text{g}^{-1}$  dry shoot (Fig. 2A). This represents a 3.2-fold increase compared to  
5 shoots of cabbage grown in soil without added  $\text{NH}_4\text{NO}_3$ .

6 In all treatments, including control, the greatest amount of biomass was produced by  
7 tepary bean followed by cabbage. Addition of ammonium nitrate at either 40 or 80 mmoles  $\text{kg}^{-1}$   
8 soil depressed biomass production in all four species (Fig. 2B); undoubtedly as a result of  
9 ammonium toxicity. High ammonium generally inhibits cation uptake, which in turn suppresses  
10 growth primarily by inducing deficiencies of magnesium (Manolakis and Lüdders, 1977) and  
11 calcium (Pill et al., 1978). The species most susceptible to ammonium toxicity was reed Canary  
12 grass; 80 mmoles ammonium nitrate caused a 70% reduction in shoot biomass.

13 The amounts of  $^{137}\text{Cs}$  removed from the soil in shoots of the tested species are shown in  
14 Table 3. The greatest amount of radiocesium, 140 pCi, was accumulated in shoots of cabbage  
15 grown in pots treated with 80 mmoles  $\text{NH}_4^+$ . This represents approximately 0.04% of the  $^{137}\text{Cs}$  in  
16 the pot. It should be remembered, however, that this amount was removed in shoots of 6-week-  
17 old seedlings which produced only 0.14 g of dry mass. Ammonium application was most efficient  
18 in reed Canary grass increasing  $^{137}\text{Cs}$  accumulation in shoots by more than 20-fold. It is possible  
19 that roots of Canary grass might have a very low ability of solubilizing radiocesium from soil  
20 particles. This would explain the very low  $^{137}\text{Cs}$  shoot concentration in plants grown in soil  
21 without  $\text{NH}_4\text{NO}_3$  added. However, total radiocesium accumulated in shoots of this grass was the  
22 least among species tested.

1 Bioaccumulation of radiocesium in shoots was statistically analyzed using ANOVA for a  
2 completely randomized two factorial design. Interactions between species and soil treatment were  
3 not statistically significant ( $p>0.05\%$ ). The amount of  $^{137}\text{Cs}$  accumulated in shoots, however, was  
4 significantly affected ( $p< 0.01$ ) by both, plant species and soil treatment. The abilities of tested  
5 species to accumulate  $^{137}\text{Cs}$  in shoots were significantly different ( $p< 0.01$ ). This ability increased  
6 in order: reed Canary grass < Indian mustard < tepary bean < cabbage. Soil treatments were also  
7 compared using Fisher's least significant difference ( $p<0.01$ ). Compared to control,  $\text{NH}_4\text{NO}_3$ ,  
8 addition significantly increased radiocesium accumulation in shoots. However, the two ammonium  
9 applications were not significantly different from each other.

10 Addition of 40 mmoles of ammonium nitrate increased the bioaccumulation ratio ( $[^{137}\text{Cs}]_{\text{dry}}$   
11  $_{\text{shoot}}/[^{137}\text{Cs}]_{\text{dry soil}}$ ) in all four species. With the exception of cabbage, however, doubling the amount  
12 of ammonium nitrate (from 40 to 80 mmoles  $\text{kg}^{-1}$  soil) had little effect on the value of  
13 bioaccumulation ratio. It is possible that at high concentrations  $\text{NH}_4^+$  competes with cesium for  
14 uptake into roots. Such an effect has been previously described (Shaw and Bell, 1991). Only  
15 cabbage apparently responded to greater addition of  $\text{NH}_4\text{NO}_3$ ; the greatest bioaccumulation ratio  
16 (2.8) was obtained in pots treated with 80 mmoles  $\text{NH}_4\text{NO}_3$  (Fig. 3).

17 In control pots, the value of the bioaccumulation ratio for  $^{137}\text{Cs}$  varied between 0 (reed  
18 Canary grass) and 1.0 (cabbage) (Fig. 3). Comparison of these ratios with those reported in the  
19 literature should be approached with caution. One concern is that some earlier studies have  
20 reported on the bioaccumulation of radiocesium from artificially contaminated soil in which the  
21 initial bioavailability of cesium was high. It is well known that radiocesium bioavailability  
22 decreases as it ages in these experimentally contaminated soil (Nisbet and Shaw, 1994). Such a

1 difference in bioavailability would make the comparison of the  $^{137}\text{Cs}$  bioaccumulation ratios  
2 between plants grown in soil with freshly applied  $^{137}\text{Cs}$  and those grown in radiocesium-aged soil  
3 inappropriate. Comparison of  $^{137}\text{Cs}$  bioaccumulating properties could also be confounded if plants  
4 were grown in different types of soil. Thus, it has been shown that the organic matter content of  
5 the soil has a large impact (up to a factor of 10) on cesium transfer from the soil to shoots (van  
6 Bergeijk et al., 1992). This concern has been previously addressed by Cremers et al., (1988).

7 **Conclusions**

8 Results obtained in this study suggest that phytoremediation of the  $^{137}\text{Cs}$ -contaminated  
9 BNL soil may be feasible. The BNL soil is contaminated to a depth of approximately 10 cm with  
10 400 pCi of  $^{137}\text{Cs g}^{-1}$ . With good management practices, a fast growing high biomass producing  
11 crop such as cabbage could yield 15-20 tones dry wt  $\text{ha}^{-1} \text{ year}^{-1}$ . Our results suggest that with high  
12 ammonium,  $^{137}\text{Cs}$  bioaccumulation ratio in shoots of cabbage might be as high as 3. Taking these  
13 parameters at face values, decontamination of BNL soil to levels below 100 pCi  $\text{g}^{-1}$  soil would be  
14 attainable in less than 15 years. Unfortunately, as radiocesium is removed from the soil the  
15 bioaccumulation ratio is likely to decrease, making further removal more difficult. It is possible,  
16 however, that implementation of specific management practices could sustain  $^{137}\text{Cs}$   
17 phytoextraction over successive cropping. For example, small increase in  $^{137}\text{Cs}$  transfer with time  
18 was reported in shoots of cabbage grown in peat soil (Nisbet and Shaw, 1994). This agrees with  
19 the observation that organic soils have the ability to sustain radiocesium availability for plant  
20 uptake (Barber and Mitchell, 1963).

21 A major problem with this assessment is that the ammonium rates used in the green house

1 are excessively high and could be not realistically employed for large scale phytoremediation of  
2 radiocesium-polluted sites. It is possible that application of organic fertilizer at the contaminated  
3 site could be important not only for maintaining a high biomass production but also for sustaining  
4 the rate of  $^{137}\text{Cs}$  phytoextraction from soils treated with significantly lower rates of ammonium.

5 The effect of combined application of organic and ammonium fertilizer on radiocesium  
6 phtoextraction is not known and should be the focus of the future research.

7

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1      Table 1. Accumulation of cesium in shoots of several plant species from a nutrient solution  
2      containing 1  $\mu\text{M}$  CsCl.

3

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	Shoot mean dry wt (g)	[Cs] in shoots $\mu\text{g g}^{-1}$ dry wt	Bioaccumulation ratio <sup>†</sup>	Total Cs accumulated in shoots ( $\mu\text{g}$ )
7      broccoli	0.062	14	105	0.87
8      cabbage	0.075	22	165	1.58
9      cauliflower	0.065	14	105	0.91
10     colonial bent grass	0.043	6	45	0.26
11     hairy vetch	1.071	15	113	1.11
12     Indian mustard	0.075	16	120	1.22
13     kochia	0.068	8	60	0.54
14     reed Canary grass	0.060	6	45	0.36
15     red fescue	0.028	5	38	0.14
16     tepary bean	0.079	19	143	1.50

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17     <sup>†</sup>[Cs]<sub>shoots</sub>/[Cs]<sub>solution</sub>

Table 2. Effect of several chemical compounds on  $^{137}\text{Cs}$  desorption from the BNL soil.

	Ammonium salts				Potassium salts				Organic acids				Mineral acids				Reducing agents				
	$\text{NO}_3^-$	$\text{Cl}^-$	$\text{SO}_4^{2-}$	Acetate	$\text{NO}_3^-$	$\text{Cl}^-$	$\text{SO}_4^{2-}$	Acetate	Formic	Acetic	Malic	Citric	$\text{HCl}$	$\text{HNO}_3$	Na-Dithionite	Hydroxylamin-HCl	EDTA				
0.05 M	15	12	15	†	-	7	8	6	0	0	0	0	1	2	1	3					
0.5 M	24	25	22	-	15	14	14	0	0	0	0	0	3	2	1	1	7				
1.0 M	-	-	-	17	18	-	-	-	-	-	-	-	-	-	-	-	-				

†  $^{137}\text{Cs}$  activity in the BNL soil was 400 pCi g dry soil<sup>-1</sup>

† Data not available

1      Table 3. Effect of soil treatment on  $^{137}\text{Cs}$  accumulation<sup>†</sup> in shoots of four plant species.

Soil treatment	Indian mustard		cabbage		tepary bean		reed Canary grass	
	pCi	%	pCi	%	pCi	%	pCi	%
Control (no $\text{NH}_4\text{NO}_3$ )	37	100	69	100	28	100	1	100
40 mmoles $\text{NH}_4\text{NO}_3$	61	165	110	159	100	357	18	1800
80 mmoles $\text{NH}_4\text{NO}_3$	64	173	140	203	118	421	12	1200

10      <sup>†</sup>Determined as:  $[^{137}\text{Cs}]_{\text{shoot}} \times \text{shoot biomass}$

1      **Figure legends**

2      **Figure 1.** Concentration-dependent desorption of  $^{137}\text{Cs}$  from the BNL contaminated soil. Soil was  
3      incubated in  $\text{NH}_4\text{NO}_3$ ,  $\text{NH}_4\text{Cl}$ , or  $\text{KNO}_3$  at concentrations shown. After 2 h, the slurry was  
4      filtered and gamma activity in solution measured.

5      **Figure 2.** Effect of soil treatment with  $\text{NH}_4\text{NO}_3$  on: A)  $^{137}\text{Cs}$  concentration in shoots, and B)  
6      shoot biomass of four plant species.

7      **Figure 3.** Effect of soil treatment with  $\text{NH}_4\text{NO}_3$  on  $^{137}\text{Cs}$  bioaccumulation ratio in shoots of 40-  
8      day-old plants.





