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TECHNETIUM AND URANIUM: SORPTION BY AND PLANT UPTAKE FROM PEAT AND SAND†

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Abstract—The objectives of this study were to compare the effects of technetium and uranium on the yield and uptake, and to identify the organ of accumulation, of an edible leafy vegetable growing in sandy and peaty soils. In sand, where the soil's sorption capacity is negligible, technetium uptake is four orders of magnitude higher than from peat, suggesting no plant mediation of uptake and thus a constant concentration factor (>50) in an oxidizing environment where technetium is continuously supplied. The technetium is predominantly translocated to the shoots. When soil fixation occurs, as in peat, this becomes the controlling factor in the plant uptake of technetium. In the case of uranium, plant mediation is more significant. Uranium uptake by Swiss chard is up to 80 times higher from sand than from peat. The uranium is restricted to the root system and may only be precipitated on the outer root membrane and may not accumulate in the roots.

INTRODUCTION

TECHNETIUM and uranium in the environment are radioactive elements of concern to the nuclear industry. Technetium-99, produced by the fission of ^{235}U and ^{239}Pu in nuclear reactors, has a half-life of 2.15×10^5 years, and has been dispersed in the environment through weapons testing and from establishments using it for medical (McA64; Be66) or commercial purposes. Uranium occurs naturally in the environment and can be mobilized during mining or milling. Both of these long-lived elements could reach the environment by releases from nuclear power plants, nuclear fuel reprocessing facilities and nuclear disposal vaults.

The study of the chemistry of technetium is still young (Pe37; Co53) whereas that of uranium (Ho73) is relatively well known. The existing knowledge of the behaviour of these elements in the environment has been recently reviewed (Wi79; Pa80; La78; Le79; Sh80). Under oxidizing conditions, the pertechnetate ion (TcO_4^-) and the uranyl ion (UO_2^{2+}) are the most likely forms of technetium and uranium to contaminate the environment. Technetium and uranium both accumulate in plants (Ca78; Ca52) and their mobilities in the environment (Lo70; Sz64) are affected by microorganisms (La77). A concise summary of the effects of technetium on plants has been presented by Berlyn (Be80). At low concentrations (0.1 μg Tc/g dry soil), (Ca78) technetium has been shown to inhibit plant growth and development. Uranium concentrations of 800 μg U/g of ashed plant have been found in black spruce (*Picea mariana*) trees surrounding a uranium ore body in Saskatchewan (Du81), and 1290 μg U/g of ashed peat have been reported for humified peat in Manitoba (Co79). Very little

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has been reported on the accumulation of those elements in edible leafy plants growing in organic soil.

The objectives of the present study were to compare the effects of technetium and uranium on the yield and uptake and to identify the organ of accumulation of an edible leafy vegetable growing in sandy or peaty soil.

MATERIALS AND METHODS

Similar volumes of sand and peat in plastic bags (~3800 cm^3 soil) were brought to field moisture capacity with distilled water and the following amounts of ^{99}Tc (as ammonium pertechnetate) and ^{238}U (as uranyl nitrate, not depleted with respect to ^{235}U) were added to the bags: 0.0, 0.0001, 0.001, 0.01 and 0.1 μg ^{99}Tc/g dry peat, 0.0, 0.00005, 0.0005, 0.005 and 0.05 μg ^{99}Tc/g dry sand, 0, 10, 20, 30 and 50 μg ^{238}U/g dry peat and 0, 5, 10, 15 and 25 μg ^{238}U/g dry sand. The technetium solutions included ^{95m}Tc†, which was added as a γ-emitting tracer (1.33 and 6.66 kBq per bag of dry peat and sand, respectively). This use of a γ-emitting isotope in plants (Ca78) eliminates the tedious chemical separations that are required when a pure β-emitter, such as ^{99}Tc, is used. All treatments were replicated three times in a completely randomized block design. The nutrient requirements for the vegetables were satisfied by a single addition of a complete nutrient solution. The characteristics of the two soils including their natural ^{238}U concentrations, are given in Table 1.

The soil was placed in lysimeters and incubated moist for 70 days. Mature Swiss chard plants were then excavated from a field site, root-pruned and transplanted into the lysimeters. Thermoluminescent dosimeters (LiF; TLD-100, The Harshaw Chemical Co.), both bare and covered (to exclude most α- and β-radiation), were placed in the soil close to the roots. After transplanting, all leaves except the youngest were removed.

The plants were grown under lights and watered regularly to maintain the soil at field moisture capacity. After 40 days, six to eight new leaves had emerged and the plants were harvested. The whole plant was removed from the lysimeter; the leaves and roots were washed, dried, weighed and ground for analysis for ^{95m}Tc by γ-ray spectroscopy using a Ge(Li) detector and for ^{238}U by delayed neutron activation analysis (DNAA). The dosimeters were removed and subsequently read. Standards were prepared by applying aliquots of a ^{95m}Tc solution to control leaves, roots and soil and carrying out the same preparation technique. Counting geometry corrections were obviated by diluting the standards and samples with wood sawdust. The uranium analysis by DNAA was carried out by the AECL Radiochemical Company, Ottawa.

Batch soil adsorption studies were carried out for technetium in polyethylene vials with air-saturated, synthetic groundwater solutions (see Table 2). Initial technetium concentrations ranged from a carrier-free ^{95m}Tc solution of 3×10^{-10} g/L to $^{95m}Tc + {}^{99}Tc$ solu-

*Table 1. Physical and chemical characteristics of peat and sand**

Characteristic	Peat	Sand
pH (0.01*m* $CaCl_2$)	2.8 ± 0.06	6.4 ± 0.26
(water)	3.8 ± 0.06	8.4 ± 0.0
% $CaCO_3$	4.9 ± 3.0	8.2 ± 1.6
% O.M. (Carbon)	92 ± 5.3 (54 ± 2.6)	-
Free Iron Oxides (μg/g)	1050 ± 26	1382 ± 54
CEC(meq/100g)	64.7 ± 8.1	1.2 ± 0.2
Uranium (μg/g)	0.6 ± 0.1	0.6 ± 0.0
texture	Organic	Coarse Sand

* mean of triplicate values

± = standard deviation

O.M. = organic matter

CEC = cation exchange capacity

†The decay half-lives of ^{95m}Tc and ^{238}U are 60.5 days and 4.5×10^9 years, respectively.

tions of 10^{-4} to 10^{-2} g/L. The relationship between the soil-solution ratio and the distribution coefficient (k_d, Pa80) was determined using ratios varying from 1 to 10 g/10 mL for sand and 0.1 to 1 g/10 mL for peat, in contact with a 10^{-3} g/L $^{95m}Tc + ^{99}Tc$ solution. Soil-solution ratios in the plant uptake study were 1 g/10 mL solution for sand and 0.1 g/10 mL solution for peat. Higher soil-solution ratios (ratios closer to field moisture capacity) could not be used in the soil sorption studies without causing difficulties in sampling the solutions at the end of the equilibration period. The mixtures (each treatment was replicated three times) were rotated gently twice a week and the equilibration period was five weeks. At the end of this period, aliquots of the solution were withdrawn, by centrifugation in sand and extraction by syringe in peat, weighed and counted with a NaI(Tl) well counter.

Table 2. Ion concentrations in synthetic groundwater

Ion	Concentration (mg/L)
Na	8.3
K	3.5
Mg	3.9
Ca	13
HCO_3	+
Cl	5.0
SO_4	8.6
NO_3	0.62
F	0.19
pH	6.5 + 0.5

+ in equilibrium with atmosphere at the final pH.

RESULTS

Dosimetry

Recovery from the soil and subsequent readability of bare TLD chips were very poor. The results (see Table 3) indicate that

Table 3. Mean dose rates from the uranium and technetium soil treatments*

SAND

Initial [U]** (μg/g)	Covered (Gy/h)	Bare (Gy/h)	Initial [Tc]** (μg/g)	Covered (Gy/h)	Bare (Gy/h)
0	3×10^{-7}	6×10^{-6} (n=4)	0	6×10^{-7}	2.5×10^{-6} (n=4)
5	6×10^{-7}	9.6×10^{-6} (n=4)	5×10^{-5}	4×10^{-7}	1.7×10^{-6} (n=2)
10	7×10^{-7}	10.9×10^{-6} (n=4)	5×10^{-4}	4×10^{-7}	2.8×10^{-6} (n=5)
15	9×10^{-7}	12.7×10^{-6} (n=4)	5×10^{-3}	4×10^{-7}	2.7×10^{-6} (n=5)
25	5×10^{-7}	6.2×10^{-6}	5×10^{-2}	4×10^{-7}	2.6×10^{-6} (n=3)

PEAT

Initial [U]** (μg/g)	Covered (Gy/h)	Bare (Gy/h)	Initial [Tc]** (μg/g)	Covered (Gy/h)	Bare (Gy/h)
0	5×10^{-7}	2.2×10^{-6}	0	6×10^{-7}	0.8×10^{-6} (n=4)
10	7×10^{-7}	1.7×10^{-6} (n=4)	1×10^{-4}	4×10^{-7}	1.1×10^{-6}
20	5×10^{-7}	1.5×10^{-6}	1×10^{-3}	3×10^{-7} (n=8)	1.5×10^{-6} (n=2)
30	1×10^{-6}	2.4×10^{-6}	1×10^{-2}	2×10^{-7} (n=8)	1.1×10^{-6} (n=4)
50	7×10^{-7}	2.0×10^{-6}	1×10^{-1}	3×10^{-7} (n=8)	1.2×10^{-6} (n=2)

* All values for "covered" dosimeters are an average of 12 observations (4 chips/replicate), and values for bare dosimeters are an average of six observations (2 chips/replicate), unless otherwise stated.

**Uranium dosimeters were implanted for 111 days, technetium dosimeters for 105 days.

the presence of uranium and technetium did not significantly enhance the gamma-ray dose (covered dosimeters) or the alpha/beta dose (bare dosimeters). The gamma dose was about an order of magnitude lower than the alpha dose for all treatments. The alpha dose was, on the average, slightly higher in the sand than in the peat. This may be due to lower moisture content in the sand, reducing the shielding of the α particles from the TLD chips. The external doses received by the roots and shoots of the plants were similar to natural background. The internal doses, due to the uptake of uranium and technetium, were also negligible, as discussed later.

Seedling yield

Analysis of the variance of the dry weight data for the shoots and roots has shown no significant differences due to treatment with technetium (see Table 4). The regression coefficients for dry plant weight (shoots or roots, in peat or sand) versus ^{99}Tc concentration are not significantly different from zero. This confirms visual observations that there was no response of plant yield to the presence of technetium at these concentrations.

A Duncan's Multiple Range Test (Table 4), however, shows that there were significant effects on the yield of roots for plants grown in both uranium-enriched sand and peat. In sand, this effect does not follow a consistent pattern. In peat, however, the control plants had a significantly greater root mass than the treatment plants. This suggests that uranium-enriched peat may inhibit root growth.

Concentration of technetium

Analysis of the variance of the uptake of total technetium (^{95m}Tc and ^{99}Tc) in the shoots and roots of the Swiss chard plants growing in sand and peat showed a significant ($P < 0.05$) effect. There was no difference in the uptake of ^{95m}Tc among the plants growing in sand and, since a constant amount of ^{95m}Tc was added to each lysimeter, this indicates a constant proportion of the ^{95m}Tc was taken up for all treatments. Assuming ^{95m}Tc is an ideal tracer for ^{99}Tc, then the plants growing in sand

Table 4. Mean yields (g) of roots and shoots

TECHNETIUM

Initial [^{99}Tc] (µg/g)	Sand Shoot	Sand Root	Initial [^{99}Tc] (µg/g)	Peat Shoot	Peat Root
0	4.44 a	2.83 a	0	4.36 a	5.07 a
5×10^{-5}	5.18 a	4.19 a	1×10^{-4}	3.25 a	3.01 a
5×10^{-4}	5.30 a	4.14 a	1×10^{-3}	4.06 a	6.04 a
5×10^{-3}	4.57 a	3.14 a	1×10^{-2}	3.90 a	4.84 a
5×10^{-2}	4.93 a	2.53 a	1×10^{-1}	5.61 a	6.05 a

URANIUM

Initial [^{238}U] (µg/g)	Sand Shoot	Sand Root	Initial [^{238}U] (µg/g)	Peat Shoot	Peat Root
0	5.29 a	2.72 ab	0	4.81 a	5.14 b
5	4.09 a	2.86 ab	10	3.83 a	2.96 ab
10	3.97 a	2.04 ab	20	4.12 a	3.96 ab
15	3.42 a	1.51 a	30	3.54 a	2.67 a
25	4.18 a	3.80 b	50	3.50 a	2.83 ab

The means within a column followed by the same letter are not significantly different by Duncan's test at $P < 0.05$.

absorbed a constant proportion of the applied technetium regardless of the actual amounts taken up. These data suggest that, in sand, neither soil-sorption processes nor plant-uptake mechanisms regulate the uptake of technetium and, therefore, the concentration factor is constant.

There was a significant increase in the concentration of ^{95m}Tc in the shoots of plants growing in peat as the level of applied ^{99}Tc increased. If the plants behaved similarly in both soils, these data suggest a saturation of the sorption capacity of the soil.

Our analyses show a striking difference in total technetium uptake by Swiss chard from sand and peat (see Fig. 1). Measured on a dry weight basis, the technetium uptake from peat was up to four orders of magnitude lower than from sand. This reduction is due to the immobilization of technetium in the peat, possibly due to reduction to far less soluble TcO_2 (Pa80) or to sorption as ionic species on the peat. Another explanation is that the acidic conditions in the peat inhibit the plant's uptake activity.

Concentration of uranium

Analysis of the variance of the uptake of uranium showed no differences in concentration of uranium in the shoots or roots of Swiss chard growing in peat, nor in the shoots of chard growing in sand. The uptake of uranium by the roots of chard growing in sand, however, was highly significant ($P < 0.005$). Consequently, total plant uptake (root plus shoot) of uranium from peat was not significant, but total plant uptake of uranium from sand was significant ($P < 0.01$) (see Fig. 2). Measured on a dry weight basis, the uptake of uranium from sand was up to 80 times greater than from peat.

The mean uranium concentrations associated with the roots growing in sand were 0.2,

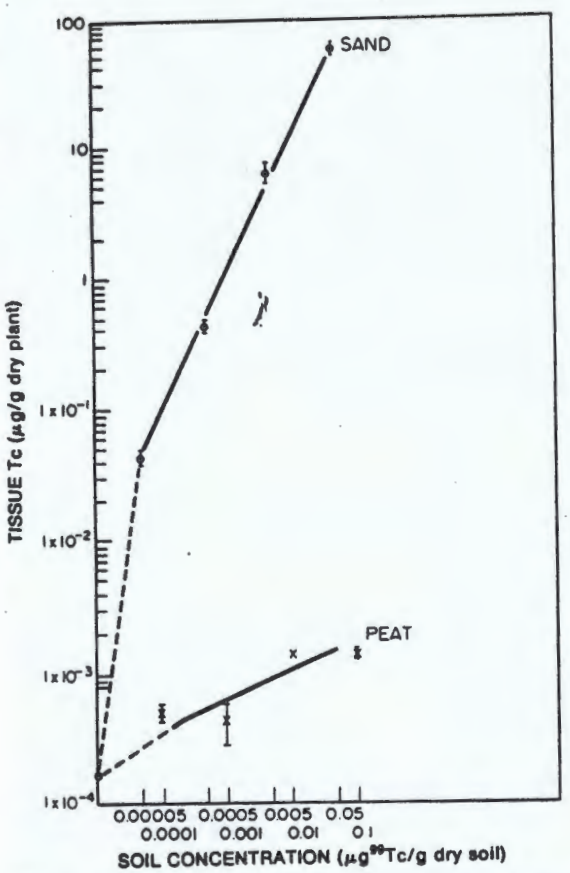

FIG 1. Tissue and soil technetium concentrations for Swiss chard plants growing in sand and peat (error bars denote standard deviation).

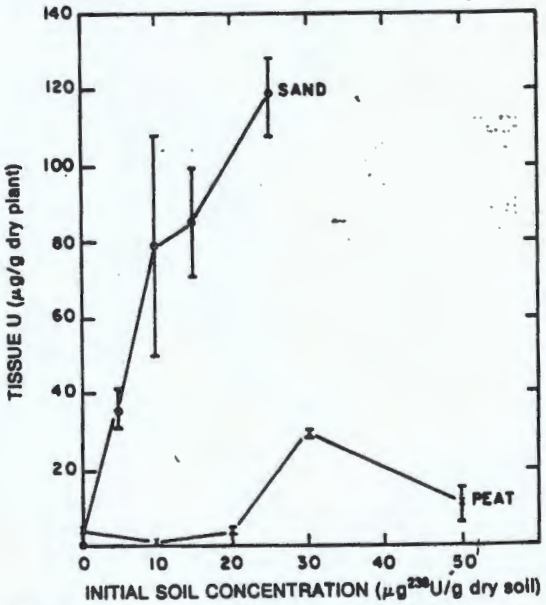

FIG. 2. Tissue and soil uranium concentrations for Swiss chard plants growing in sand and peat (error bars denote standard deviation).

34.7, 75.8, 82.1 and 114 μg ^{238}U/g dry root for the treatments containing 0, 5, 10, 15 and 25 μg ^{238}U/g dry soil. The highest concentration value for uranium associated with a root was 140 μg ^{238}U/g dry root. Based on unit weight, the dried root had from 4.5 to 7.5 times the amount of uranium in the dry soil. Assuming a bulk density for dry sand of 1.7 g/cm^3 and a root density of ~0.45 g/cm^3, then, on a volume basis, the root accumulated from 1.2 to 2 times the amount of uranium in the soil. It is not clear whether the uranium is in or on the root (i.e. associated with very persistent soil particles either struck to the roots or precipitated on the outer root membrane (Ku39)).

Internal radiation dose

Based on a wet to dry weight ratio of 9 for Swiss chard tops (actually, the ratio is 9.5 ± 0.5, $n = 3$), the highest value (58 μg total Tc/g dry plant, see Fig. 1) can be converted to a concentration of total Tc/g wet plant. This gives a concentration of 6.4 μg total Tc/g wet plant. Since such a small portion of this is ^{95m}Tc, and the gamma dose which would be attributed to it was so small, we can assume that we have about 6.4 μg ^{99}Tc/g wet plant. The β-dose from ^{99}Tc would then be 2.9×10^{-10} Gy/h absorbed dose rate in the plant tissue. This is lower than natural background suggesting that the internal radiation levels experienced by the plants containing even the greatest amounts of technetium were negligible.

Since no significant accumulation of uranium was observed in or on the plants, no calculation for internal radiation due to uranium was made.

Plant concentration factors for technetium

The plant concentration factor (C.F.) (Ba76), as applied to the environmental assessment of radionuclide migration in the foodchain, is defined as:

$$\text{C.F.} = \frac{\text{concentration in plant } (\mu\text{g or Bq/g wet plant})}{\text{concentration in soil } (\mu\text{g or Bq/g dry soil})}$$

Since the final ^{95m}Tc soil counts were very low, the activity in the soil has been estimated based on the number of half-lives that occurred between the spiking and counting dates, and subtracting that detected in the combined root and shoot. The percentage of technetium removed by the plants growing in the sand was as high as 93%, illustrating that plant uptake rather than soil sorption predominated. This was obvious from the lack of detection of ^{95m}Tc in the sand soil.

The C.F. estimates at the end of the experiment (Table 5) show no significant correlation with ^{99}Tc concentration in either the sand or the peat. However, Table 5 does show that the C.F. for uptake from sand is approx. 60–180 times greater than for the peat. In a system where technetium is continuously supplied, this suggests that the C.F. could be very large before a lethal concentration is reached. The C.F. of 50 suggested by Till (Ti78) is far too low for such a non-adsorbing soil system; however the C.F. of

Table 5. Mean plant concentration factor vs initial ^{99}Tc concentration: sand and peat

SAND		PEAT	
Initial [^{99}Tc] (µg/g)	C.F.	Initial [^{99}Tc] (µg/g)	C.F.
0	1.7×10^3	0	18
0.00005	2.1×10^3	0.0001	12
0.0005	2.0×10^3	0.001	11
0.005	3.0×10^3	0.01	32
0.05	2.6×10^3	0.1	40

30–1200 suggested by Routson and Cataldo (Ro77a) is in agreement with our findings. The lethal concentration for Swiss chard is >0.05 μg ^{99}Tc/g dry soil.

Soil sorption of technetium

There is essentially no sorption of technetium on sand (see Tables 6a and b). Even at high soil-solution ratios, k_d is nearly zero. This agrees with results obtained by Routson *et al.* (Ro77), and with those obtained for soils with a low organic-carbon content by Landa *et al.* (La77). In contrast, sorption on peat is higher (see Table 7a) and agrees with data reported by Landa *et al.* (La77). Our results show an increase in k_d with increased peat-solution ratio (Table 7b) and also an increase in k_d with a decrease in initial technetium concentration (Table 7a). Although we could not obtain the peat-solution ratio of 600 g peat to 2600 mL water (field moisture capacity), our results show that even at a 260 g peat to 2600 mL water ratio, 90% of the technetium is tied up and that little would be available for uptake by the Swiss chard.

CONCLUSIONS

(1) In sand, where the sorption capacity of the soil is negligible, the plants absorbed a constant proportion of the applied technetium, suggesting no plant mediation of uptake and thus a constant concentration factor between the soil solution and the plant, within a concentration range of 5×10^{-5} to 0.1 μg Tc/g dry soil.

(2) When soil sorption occurs, as in peat, this becomes the controlling factor in the uptake of technetium by plants.

(3) In the case of uranium, plant mediation plays a more important role, since essentially no uranium translocation occurs in the plant. Uranium may be excluded from the plants by an active root-membrane selection process.

(4) Technetium uptake by Swiss chard from sand is up to four orders of magnitude higher than from peat at the same level of applied technetium, and the technetium is predominantly translocated to the shoots.

(5) Uranium uptake by Swiss chard from sand is up to 80 times higher than from peat at the same level of applied uranium.

Table 6(a). Distribution coefficient and percent adsorption of technetium for sand

Initial [Tc] (g/L)	k_d* (mL/g)	% Adsorbed*
3×10^{-10}	0.24 ± 0.06	2.6 ± 0.7
1×10^{-4}	0.02 ± 0.02	0.3 ± 0.3
3×10^{-4}	0.04 ± 0.03	0.8 ± 0.4
1×10^{-3}	0.10 ± 0.08	1.0 ± 0.5
3×10^{-3}	0.07 ± 0.06	0.7 ± 0.6

Table 6(b). Distribution coefficient and percent adsorption of technetium for sand as a function of the soil-solution ratio. Initial technetium concentration of 10^{-3} g/L

Soil/Solution (g/10 mL)	k_d* (mL/g)	% Adsorbed*
3.1 ± 0.3	0.06 ± 0.01	1.7 ± 0.4
6.2 ± 0.1	0.02 ± 0.03	0.9 ± 1.6
10.1 ± 0.1	0.02 ± 0.01	1.6 ± 1.5

* average of triplicate experiment.

± = standard deviation.

Table 7(a) Distribution coefficient and percent adsorption of technetium for peat

Initial [Tc] (g/L)	k_d* (mL/g)	% Adsorbed*
3×10^{-10}	28 ± 10	28 ± 12
1×10^{-4}	23 ± 4	34 ± 4
3×10^{-4}	15 ± 1	22 ± 3
1×10^{-3}	15 ± 8	11 ± 4
3×10^{-3}	6.4 ± 1.3	10 ± 4
1×10^{-2}	4.7 ± 0.6	5.6 ± 1.2

Table 7(b). Distribution coefficient and percent adsorption of technetium for peat as a function of soil-solution ratio. Initial technetium concentration of 10^{-3} g/L

Soil/Solution (g/10 mL)	k_d (mL/g)	% Adsorbed
0.31 ± 0.04*	33 ± 8*	50 ± 9*
0.49 ± 0.08+	40 ± 6+	66 ± 8+
0.74*	95*	88*
0.93 ± 0.02+	93 ± 3+	90+
1.15*	68++	89*

* average of triplicate experiment

\+ average of duplicate experiment

++ low value, probably due to incomplete separation of solid and solution.

± = standard deviation.

However, as previous studies suggest, it is restricted to the root system and may only be precipitated on the outer root membrane and may not accumulate in the root.

(6) Root yields were reduced with >10 μg ^{238}U/g soil in both peat and sand; however, shoot yields were not significantly affected.

(7) No yield effects were found for Swiss chard growing in either technetium-contaminated peat or sand; Swiss chard continues normal growth at technetium levels (0.1 μg Tc/g dry soil) found toxic to soybean.

(8) A plant concentration factor for technetium of 50 used in dose assessment models appears to be too low for a non-adsorbing soil system, such as sand.

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