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## Uptake of natural and anthropogenic actinides in vegetable crops grown on a contaminated lake bed

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### Abstract

Activity concentrations and plant/soil concentration ratios (CRs) of <sup>239,240</sup>Pu, <sup>241</sup>Am, <sup>244</sup>Cm, <sup>232</sup>Th, and <sup>238</sup>U were determined for three vegetable crops grown on an exposed, contaminated lake bed of a former reactor cooling reservoir in South Carolina, USA. The crops included greens and tubers of turnips (*Brassica rapa* var. white-globe), bush beans (*Phaseolus vulgaris*), and husks and kernels of sweet corn (*Zea mays* var. silver queen). Although all plots were fertilized, some received K<sub>2</sub>SO<sub>4</sub>, while others received no K<sub>2</sub>SO<sub>4</sub>. The K<sub>2</sub>SO<sub>4</sub> fertilizer treatment generally lowered activity concentrations for <sup>241</sup>Am, <sup>244</sup>Cm, <sup>232</sup>Th and <sup>238</sup>U, but differences were statistically significant for <sup>241</sup>Am and <sup>244</sup>Cm only. Highly significant differences occurred in activity concentrations among actinides and among crops. In general, turnip greens exhibited the highest uptake for each of the actinides measured, while corn kernels had the least. For turnip greens, geometric mean CRs ranged from  $2.3 \times 10^{-3}$  for <sup>239,240</sup>Pu to  $5.3 \times 10^{-2}$  for <sup>241</sup>Am (no K<sub>2</sub>SO<sub>4</sub> fertilizer). For corn kernels, geometric mean CRs ranged from  $2.1 \times 10^{-3}$  for <sup>239,240</sup>Pu and <sup>232</sup>Th to  $1.5 \times 10^{-3}$  for <sup>244</sup>Cm (no K fertilizer). In general, CRs across all crops for the actinides were in the order: <sup>244</sup>Cm > <sup>241</sup>Am > <sup>238</sup>U > <sup>232</sup>Th > <sup>239,240</sup>Pu. Lifetime health risks from consuming crops contaminated with anthropogenic actinides were similar to the risks from naturally occurring actinides in the same crops (total  $\sim 2 \times 10^{-6}$ ); however, these risks were only  $\sim 0.3\%$  of the risk from consuming the same crops contaminated with <sup>137</sup>Cs. © 1999 Elsevier Science Ltd. All rights reserved.

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## 1. Introduction

Although many measurements of the uptake of actinides from soil by plant crops have been made, rather large variations have been observed (IAEA, 1994). Such variations result from differences in soil characteristics, chemical properties of the radionuclides, crop species and tissues, and other variables. These variations produce large uncertainties for human health and ecological risk assessments unless site- and condition-specific measurements can be made prior to the performance of risk calculations.

This study documents radionuclide uptake by garden crops grown under rather unusual conditions, namely on the exposed lake bed of Par Pond, a former nuclear reactor cooling impoundment (Seel et al., 1995). Par Pond, a large reservoir that served as part of the cooling system for two nuclear materials production reactors on the Savannah River Site near Aiken, South Carolina, USA, was lowered in 1991 due to concerns about the integrity of the dam which forms the impoundment. Because of the presence of radionuclide contamination, the exposed 5.3 km<sup>2</sup> of lake bed was declared an operable unit under the Comprehensive Environmental Response, Compensation, and Liability Act (CERCLA) by the U.S. Environmental Protection Agency in April 1992 (Marcy et al., 1994). One regulatory requirement under CERCLA is a lifetime human health risk assessment of various management options for dealing with contaminated sites, such as the Par Pond lake bed. The work described here was designed to provide site-specific data on the uptake of <sup>239,240</sup>Pu, <sup>241</sup>Am, <sup>244</sup>Cm, <sup>232</sup>Th, and <sup>238</sup>U by crops grown on the lake bed and ingested by hypothetical residents of the area.

The study addressed the following questions: (1) What are the plant/soil concentration ratios (CRs) for three anthropogenic and two naturally occurring actinides in representative food crops that might be grown on the lake bed? (2) Does plant uptake of actinides differ among crops, plant tissue types, and radionuclides at this site? (3) How do the CR values compare with other values in the literature? and (4) What is the comparative magnitude of the hypothetical health risk from consuming crops containing these actinides when compared to risks from consumption of the same crops contaminated with <sup>137</sup>Cs. The latter was addressed by Seel et al. (1995) using data from the same garden plots.

## 2. Study site

The Savannah river site (SRS) is an 800 km<sup>2</sup> land area that was established in 1951 under the Manhattan Project (Bebbington, 1990). For nearly four decades, the SRS served as one of the USA's primary sources of reactor-produced nuclear materials for defense and civilian uses (Westinghouse Savannah River Company, 1992). Plutonium and tritium were the primary materials produced by five reactors located near the center of the SRS.

Par Pond, located near the eastern boundary of the SRS, is a 10.5 km<sup>2</sup> reservoir created in 1958 by construction of an earthen dam across Lower Three Runs Creek,

a tributary of the Savannah River. Par Pond provided a large portion of the water used to cool two reactors: 'P' and 'R'. Actual records of actinide releases from these reactors to the Par Pond system are incomplete (Carlton et al., 1992a). However, based on measured releases of  $^{137}\text{Cs}$  from R reactor (the main source) to the Par Pond system ( $\sim 8.2 \times 10^{12}$  Bq; Carlton et al., 1992b), and radionuclide ratios measured in Pond B (Whicker et al., 1990) and Par Pond sediments, roughly  $2 \times 10^9$ – $2 \times 10^{10}$  Bq of each anthropogenic actinide in this study must have been released to the cooling system. Using  $^{137}\text{Cs}$  inventory estimates and actinide/ $^{137}\text{Cs}$  ratios in sediments, we estimate the anthropogenic actinide inventories in Par Pond itself to range from roughly  $4 \times 10^8$ – $4 \times 10^9$  Bq, and the corresponding average depositions to range from about 40–400 Bq m $^{-2}$ . Depositions (Bq m $^{-2}$ ) are spatially variable in the Par Pond ecosystem due to complex erosional and sedimentation processes (Connor et al., 1997).

In March of 1991, evidence of internal erosion of the Par Pond dam was discovered. For safety reasons, the reservoir water level was lowered about 6 m between 21 June and 19 September 1991, resulting in the exposure of roughly 5.3 km $^2$  of contaminated sediment. This caused a complex series of ecological impacts (Whicker et al., 1993), as well as changes in the dynamics of  $^{137}\text{Cs}$  (Whicker et al., 1997), and probably other long-lived radionuclides. This also raised questions of whether the dam should be repaired and the impoundment refilled, or whether the reservoir should be partially or completely drained for an indefinite period of time (Marcy et al., 1994).

### 3. Materials and methods

#### 3.1. Garden establishment

A garden plot was established in an accessible area previously under 0.5–2.0 m of water. Prior to the drawdown, this location was characterized by extensive macrophyte beds and a several-centimeter-thick surface layer of organic detritus, silt, and clay particles, underlain by a sandy substrate.

Before constructing the garden plot, nine soil samples (0–15 cm in depth) were collected, homogenized, and sent to the University of Georgia's Soil Testing and Plant Analysis Laboratory in Athens, GA, for nutrient assay and measurement of the characteristics necessary to determine the proper fertilizer supplements to grow crops successfully on the site. Results indicated the vertically-mixed soil to be very sandy (range = 78–94%; mean = 89%), low in clay (range = 0–14%; mean = 5%), depleted in organic matter (range = 0.7–2.5%; mean = 1.5%), low in potassium (range = 13–32  $\mu\text{g g}^{-1}$ ; mean = 20  $\mu\text{g g}^{-1}$ ), and acidic (pH range = 3.8–4.2; mean = 4.0). The clay minerals in Par Pond sediments have been found to be 85% kaolinite, 7% illite, and 8% mixed chlorite/vermiculite (Ruhe & Matney, 1980).

Soil was tilled to an average depth of 23 cm. A 13  $\times$  5 m garden plot was divided into five randomized blocks. Prior to planting, all blocks were fertilized (70 g m $^{-2}$  lime, 5.6 g m $^{-2}$  superphosphate, 1.4 g m $^{-2}$  ammonium nitrate, 3.5 g m $^{-2}$  magnesium, 0.5 g m $^{-2}$  manganese, 0.8 g m $^{-2}$  iron, 0.4 g m $^{-2}$  copper, 0.3 g m $^{-2}$  boron, 0.6 g m $^{-2}$

zinc, and  $0.0003 \text{ g m}^{-2}$  molybdenum). Two blocks received  $3.8 \text{ g m}^{-2} \text{ K}_2\text{SO}_4$ ; three blocks received no  $\text{K}_2\text{SO}_4$ . The addition of  $\text{K}_2\text{SO}_4$  raised the mean potassium concentration from 20 to  $\sim 25 \mu\text{g g}^{-1}$ . Purple top turnips (*Brassica rapa* var. white-globe) were started from seed in a greenhouse prior to transplanting to the outdoor garden. Bush beans (*Phaseolus vulgaris*), and sweet corn (*Zea mays* var. silver queen) were seeded directly in the garden.

Although resuspension measurements were not made, we believe that our experimental conditions produced data primarily reflective of root uptake from the soil, rather than resuspension. First, the soil was tilled, so only a small fraction ( $< 1\%$ ) of the contamination would be subject to wind resuspension or rainsplash. Secondly, the plot was small, and surrounded by a fabric fence, which excluded animals and reduced wind velocities. Third, the soil was generally moist during the study. Finally, the crops themselves grew rapidly and produced a significant protective cover.

### 3.2. Sampling and sample preparation

After fertilization of the garden plots but prior to planting the crops, a 5 cm diameter  $\times$  20 cm long soil core was taken 1 m from each end of each row and directly in the center of each row ( $n = 15$ ). Similar coring was also carried out following crop harvest. Individual core samples were homogenized and then oven dried ( $78^\circ\text{C}$ ).

Crops were irrigated with uncontaminated well water as necessary, and grown to maturity. After the crops had grown to maturity, they were harvested, thoroughly rinsed in tap water to remove any visible surface soil, patted dry, and stored in a freezer. Plant tissues were thawed at room temperature, oven dried ( $78^\circ\text{C}$ ), ground, and then stored in air tight bottles for actinide analysis. Composite samples from more than one plant were prepared when necessary.

### 3.3. Radioanalytical procedures

Soils and harvested plant tissues were redried at  $80^\circ\text{C}$ , aliquots weighed out and then ashed at  $520^\circ\text{C}$ . The ashed residues from 3-10 g of dry soil or from 2-30 g of dry plant material were extracted three times with near-boiling 6 M HCl. The acid extract solutions were evaporated and converted to 8 M  $\text{HNO}_3$  in order to begin a sequence of ion exchange and radiochemical separations designed to isolate plutonium, americium (plus curium), thorium, uranium, and certain other radionuclides of interest. The separated actinides were electrodeposited onto stainless-steel discs and counted with a low background alpha spectroscopy system. Appropriate isotopic diluents of very high purity were introduced into the initial ashed residues. Details of the methodology can be found in Wahlgren and Orlandini (1982), Fried et al. (1985), Nelson and Orlandini (1986), Kaplan et al. (1994), and Edgington et al. (1996). A technique for the final isolation of uranium is presented here as an example.

After preliminary separation of plutonium and thorium (anion exchange from 8 M  $\text{HNO}_3$ ) the uranium bearing matrix solution was evaporated, converted to 9 M HCl and passed through a column of anion exchange resin. The complex chloride anions of uranium and iron were selectively retained and separated from most other elements in

this step. The uranium and iron were readily removed from the chloride anion column with dilute HCl (0.1-1 M). The eluate was evaporated to dryness. If iron was absent (< 0.5 mg) uranium was immediately electrodeposited and assayed by alpha spectroscopy. To separate uranium from any iron (> 0.5 mg) present, the residue or an aliquot of the uranium-iron fraction was taken up in 1-2 ml 8 M HNO<sub>3</sub> and applied to a small column (5 × 75 mm, 2 ml vol.) of anion exchange resin (Dowex 1 × 8 100-200 mesh). The column was then rinsed with 1 ml aliquots of 8 M HNO<sub>3</sub> to accumulate 6 or 7 ml total rinse. Because iron forms no nitrate complex in strong nitric acid, it was eluted from the resin column with a few ml of rinse. The uranium, however, having a distribution coefficient of 20 ml g<sup>-1</sup> in 8 M HNO<sub>3</sub>, had moved down the column but was effectively retained during the small volume rinse step. The uranium was then readily eluted from the resin column with dilute HCl, electrodeposited and assayed as above.

### 3.4. Statistics

Since the data were strongly skewed and characteristic of ln-normal distributions, all statistical analyses were conducted on the normalized, ln-transformed data. Therefore, geometric means and geometric standard deviations are reported throughout. Differences among ln-transformed means were determined from Bonferroni's multiple comparison procedure (SYSTAT, 1992).

## 4. Results and discussion

### 4.1. Activity concentrations

Activity concentrations and associated statistics in garden soil for the actinides studied are shown in Table 1. The geometric mean activity concentrations of the anthropogenic radionuclides (<sup>241</sup>Am, <sup>244</sup>Cm, and <sup>239,240</sup>Pu) ranged from 0.45 to 3.34 Bq kg<sup>-1</sup>; while those of the naturally-occurring actinides were roughly an order-of-magnitude higher, 29 and 25 Bq kg<sup>-1</sup> for <sup>232</sup>Th and <sup>238</sup>U, respectively. The values

Table 1  
Geometric means (GM), geometric standard deviations (GSD), arithmetic means ( $\bar{x}$ ), standard deviations ( $s$ ), standard error of mean (sem), and sample sizes ( $n$ ) for actinide concentrations (Bq kg<sup>-1</sup>) in garden soils. Samples were taken after tilling, but prior to planting

Statistic	<sup>241</sup> Am	<sup>244</sup> Cm	<sup>239,240</sup> Pu	<sup>232</sup> Th	<sup>238</sup> U
GM	1.47	0.45	3.34	28.8	25.1
GSD	1.61	1.44	1.66	1.28	1.59
$\bar{x}$	1.60	0.47	3.69	29.7	27.0
$s$	0.59	0.16	1.48	6.7	8.3
sem	0.16	0.05	0.40	1.7	2.1
$n$	13	12	14	15	15

Table 2

Geometric means of actinide concentrations (mBq kg<sup>-1</sup>) in garden crops. Geometric standard deviations and sample sizes are given in parentheses. Americium and curium data are separated into potassium sulfate (K) and no potassium sulfate (NK) treatments; other radionuclides did not have significant treatment differences, so the results were pooled

Actinide	Bush bean	Corn husk	Corn kernel	Turnip green	Turnip tuber
<sup>241</sup> Am (K)	6.2 (1.6;5)	2.1 (1.2;5)	1.4 (1.4;6)	69.1 (1.3;5)	5.4 (1.8;5)
<sup>241</sup> Am (NK)	11.5 (1.2;5)	2.5 (1.1;5)	1.7 (2.7;5)	84.5 (1.5;5)	8.3 (1.4;5)
<sup>244</sup> Cm (K)	3.6 (1.5;5)	1.3 (1.1;5)	0.3 (2.2;5)	22.1 (1.1;5)	8.9 (2.2;5)
<sup>244</sup> Cm (NK)	8.5 (1.5;5)	1.4 (1.2;5)	0.7 (1.7;5)	19.5 (1.7;5)	6.7 (1.2;5)
<sup>239,240</sup> Pu	1.3 (2.2;8)	1.1 (1.6;10)	0.08 (2.2;10)	8.6 (1.8;10)	0.7 (1.9;10)
<sup>232</sup> Th	27.1 (1.3;10)	16.1 (1.4;10)	0.6 (2.0;10)	69.2 (1.6;10)	8.8 (2.0;10)
<sup>238</sup> U	47.9 (1.3;10)	19.5 (1.3;10)	5.8 (1.9;11)	156.3 (1.5;10)	26.8 (1.7;10)

for <sup>232</sup>Th and <sup>238</sup>U appear to represent natural concentrations, both of which are reported to be in the general range of ~10–50 Bq kg<sup>-1</sup> in soils (NCRP, 1976). Some releases of uranium have been documented at the Savannah River Site, but so far as we are aware, no releases to the Par Pond system were reported (Evans et al., 1992). The values for all actinides are believed to be fairly robust, since they are based on 12–15 well-homogenized soil samples from the garden plot. These values were used in the calculation of plant/soil concentration ratios.

Geometric mean activity concentrations in crops ranged from a low of 0.08 mBq kg<sup>-1</sup> for <sup>239,240</sup>Pu in corn kernels to a high of 156 mBq kg<sup>-1</sup> for <sup>238</sup>U in turnip greens (Table 2). Geometric standard deviations ranged from 1.1 to 2.7, with an average of 1.6 across all crops and actinides. Analysis of variance performed on the ln-transformed data revealed significant differences among the five actinides ( $p < 0.0001$ ), among the various crop tissues ( $p < 0.0001$ ), and that significant crop × actinide interactions occurred ( $p < 0.0001$ ). Results for <sup>241</sup>Am and <sup>244</sup>Cm are shown for plots with (K) and without (NK) potassium sulfate fertilizer because fertilizer effects were significant ( $p = 0.01$ ) for these actinides only; although, a general trend of reduced activity concentration due to potassium sulfate additions was suggested for all the radionuclides studied (Fig. 1). All crop tissues took up less <sup>241</sup>Am when fertilized, while the fertilizer affect for <sup>244</sup>Cm only occurred in bush beans and corn (Table 2).

The apparent effect of K<sub>2</sub>SO<sub>4</sub> fertilizer on <sup>241</sup>Am and <sup>244</sup>Cm was not expected. Fertilization had a dramatic impact on <sup>137</sup>Cs uptake in the same study plots as ours (Seel et al., 1995), in agreement with many other studies, but we are not aware of a similar effect for actinides. It is possible that the SO<sub>4</sub><sup>2-</sup> ion associated with the potassium amendment was involved, rather than K itself. In fact, K<sub>2</sub>SO<sub>4</sub> fertilization was calculated to have increased the initial K concentration by only ~25%. Possible increases in plant growth resulting from the K<sub>2</sub>SO<sub>4</sub> fertilizer application might be expected to cause growth dilution and reduced concentrations. Significantly increased growth of corn in response to K<sub>2</sub>SO<sub>4</sub> fertilizer was observed, but no comparable effect was evident for bush beans or turnips (Seel et al., 1995).

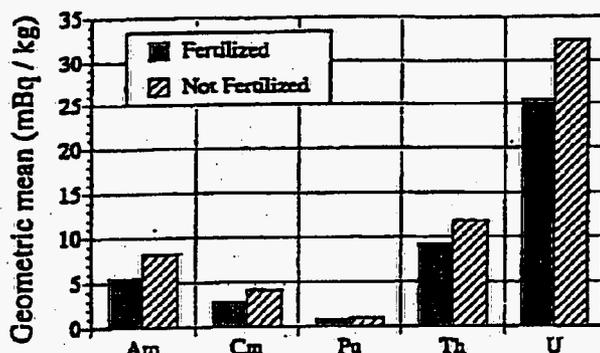


Fig. 1. Geometric mean activity concentrations for radionuclides in crops grown in plots fertilized with and without potassium fertilizer ( $3.8 \text{ g m}^{-2} \text{ K}_2\text{SO}_4$ ). Significant effects from fertilization occurred for  $^{241}\text{Am}$  and  $^{244}\text{Cm}$  ( $p = 0.01$ ).

Table 3

Ranking of crops, by actinide, in order of lowest to highest  $\ln$ -transformed activity concentrations. For a given actinide, activities in crops are shown to differ significantly ( $p < 0.05$ ) if not connected by an equal sign, or not connected with a common underline. Crops are corn kernel (CK), corn husk (CH), turnip tuber (TT), bush bean (BB), and turnip green (TG)

Actinide	Crops compared	
	Lower activity	Higher activity
$^{241}\text{Am}$	(CK = CH)	(TT = BB) < TG
$^{244}\text{Cm}$	CK < CH	(BB = TT) < TG
$^{239,240}\text{Pu}$	CK < (TT = CH = BB)	< TG
$^{238}\text{U}$	CK < (CH = TT = BB) <sup>a</sup>	< TG
$^{232}\text{Th}$	CK < (TT = CH = BB) <sup>b</sup>	< TG

<sup>a</sup>CH < BB.

<sup>b</sup>TT < BB.

Rankings in activity concentrations by crops and tissues for each actinide are shown in Table 3. Significant ( $p < 0.05$ ) differences among crops are shown. In all cases, turnip greens exhibited higher uptake than all other crops and corn kernels had the lowest. Only in the case of  $^{232}\text{Th}$ , did turnip tubers have significantly different activity concentrations from bush beans. Differences in actinide uptake among plant species and among different tissues have been observed by others. Pinder et al. (1990) also showed that corn kernels exhibited very low accumulation of Pu isotopes relative to other parts of the corn plant; and Schreckhise and Cline (1980) have shown legumes to accumulate more activity than grasses, and seeds to accumulate much less than other plant tissues.

The comparative uptake of the actinide elements by plants is affected by numerous chemical, physical and biological conditions in the soil. The combined effects of these conditions, as well as the individual chemical properties of the actinides, tend to affect

the relative sorption of these elements to soil particles. For example, detailed studies by Wildung and Garland (1980) have led to the expectation that the order of sorption is  $\text{Pu}^{2+} > \text{Am}^{3+} \sim \text{Cm}^{3+} > \text{Np}^{5+}$ . Presumably, only the soluble fraction of these elements would be available for plant uptake, and the higher the degree of sorption to soil particles, the lower the soluble fraction. Furthermore, Kaplan et al. (1994) found Am and Cm to exhibit greater mobility than Pu and Th in groundwater at the Savannah River Site. These studies lend credence to our observations on relative plant uptake of Pu, Am and Cm. We believe that differences in the 'effective' cationic charge (Choppin and Rao, 1984; Choppin et al., 1997) expected among the actinides in surface soils ( $3^+$  for Am and Cm;  $3.3^+$  for U;  $4^+$  for Pu and Th) explain the relative differences in their mobility and plant uptake.

#### 4.2. Concentration ratios

Geometric mean CRs across all crops are shown by actinide in Fig. 2. Geometric means of plant/soil CRs when separated by crop ranged from a low of  $2 \times 10^{-5}$  for  $^{239,240}\text{Pu}$  in corn kernels to a high of  $5 \times 10^{-2}$  for  $^{241}\text{Am}$  in turnip greens growing in the plots without  $\text{K}_2\text{SO}_4$  fertilizer (Table 4). Results for  $^{241}\text{Am}$  and  $^{244}\text{Cm}$  are shown for plots with (K) and without (NK)  $\text{K}_2\text{SO}_4$  fertilizer, since the differences were statistically significant for these actinides. Geometric standard deviations for specific crop-actinide combinations ranged from 1.1 to 2.7. It is clear that geometric mean CRs for  $^{241}\text{Am}$  and  $^{244}\text{Cm}$  are similar to each other, but substantially higher than values for the other actinides. CRs for  $^{232}\text{Th}$  and  $^{239,240}\text{Pu}$  were also similar to each other, but lower than the other actinides.  $^{238}\text{U}$  appeared to be intermediate between Th-Pu and Am-Cm in terms of CRs. The similarity of plant uptake for Am and Cm, and the higher uptake of Am and Cm than Pu, has been observed before. Schreckhise and Cline (1980), using soil from the Hanford site in Washington (USA) and different plant species (alfalfa, barley, peas, and cheatgrass) found the order of plant uptake to be  $\text{Np} > \text{Cm} = \text{Am} > \text{Pu}$ , in agreement with this study. Schultz et al. (1976) also

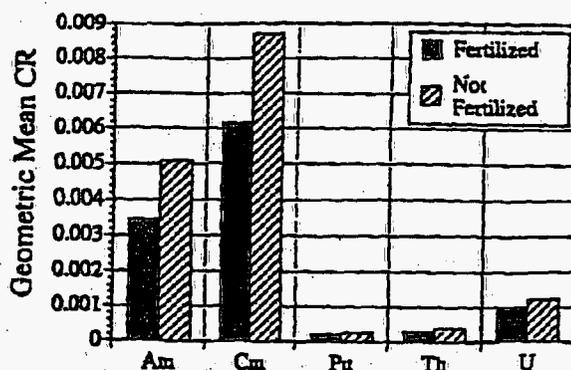


Fig. 2. Geometric mean concentration ratios ( $\text{Bq kg}^{-1}$  dry plant per  $\text{Bq kg}^{-1}$  dry soil) for radionuclides in crops fertilized with and without potassium fertilizer ( $3.5 \text{ g m}^{-2} \text{ K}_2\text{SO}_4$ ).

Table 4

Geometric means of concentration ratios ( $\text{Bq kg}^{-1}$  dry plant/ $\text{Bq kg}^{-1}$  dry soil) of actinides in garden crops. Geometric standard deviations and sample sizes are given in parentheses. Americium and curium data are separated into  $\text{K}_2\text{SO}_4$  (K) and no  $\text{K}_2\text{SO}_4$  (NK) treatments; other radionuclides did not differ significantly from fertilization

Actinide	Bush bean	Corn husk	Corn kernel	Turnip green	Turnip tuber
$^{241}\text{Am}$ (K)	0.0039 (1.6:5)	0.0013 (1.2:5)	0.00087 (1.4:5)	0.0432 (1.3:5)	0.0034 (1.7:5)
$^{241}\text{Am}$ (NK)	0.0072 (1.1:5)	0.0016 (1.1:5)	0.0011 (2.7:5)	0.0528 (1.5:5)	0.0053 (1.4:5)
$^{244}\text{Cm}$ (K)	0.0077 (1.5:5)	0.0027 (1.1:5)	0.00073 (2.2:5)	0.0469 (1.2:5)	0.0189 (1.3:5)
$^{244}\text{Cm}$ (NK)	0.0180 (1.5:5)	0.0030 (1.2:5)	0.0015 (1.7:5)	0.0416 (1.7:5)	0.0143 (1.2:5)
$^{239,240}\text{Pu}$	0.00036 (2.2:8)	0.00029 (1.6:10)	0.000021 (2.2:10)	0.0023 (1.8:10)	0.00019 (1.9:10)
$^{232}\text{Th}$	0.00091 (1.3:10)	0.00054 (1.4:10)	0.000021 (2.0:10)	0.0023 (1.6:10)	0.00029 (2.0:10)
$^{238}\text{U}$	0.0018 (1.3:10)	0.00072 (1.3:10)	0.00021 (1.9:10)	0.0058 (1.5:10)	0.00099 (1.7:10)

found Am uptake to exceed Pu uptake. One study, however, found the soil to plant transfer of Pu to be greater than that of Am (Bunzl & Kracke, 1994).

The magnitude and range of CRs found in this study appear to be generally similar to values obtained in other studies where root uptake is the primary mechanism of accumulation (IAEA, 1994). Greenhouse studies, and field studies of well-aged deposits in humid, densely vegetated areas generally tend to reflect root uptake more than rain/wind resuspension and subsequent foliar deposition of contaminated soil particles. We believe, based on the conditions in our study (aged Pu in vertically mixed soil in a humid, densely vegetated, and wind-sheltered site), that root uptake was the primary mechanism of plant contamination. Our observed ranges of CR values were quite large, often two orders-of-magnitude, depending on the actinide, crops and plant tissue. The ranges of values in the literature are also comparably large, and have been shown to vary with soil pH, organic matter, various soil amendments, and basic physical and microbial properties of the soil (Adriano et al., 1980; Wildung & Garland, 1980).

Concentration ratios for  $^{137}\text{Cs}$  in our garden plots ranged from 0.22 to 6.8 (Seel et al., 1995) compared to geometric mean CRs of  $2.1 \times 10^{-5}$ – $5.3 \times 10^{-2}$  for the actinides. The unusually high CRs for  $^{137}\text{Cs}$  were attributed to the sandy, acidic, nutrient-poor soil, which lacked significant quantities of 2:1 lattice clays that irreversibly bind Cs. Apparently, these soil properties did not greatly affect the comparative CR values for the actinides. For example, Trabalka and Garten (1983) showed ranges of CR values for U, Th and Pu in crops grown on clay-rich flood plain soils in Tennessee that were very similar to our values in South Carolina. Our CR values for  $^{241}\text{Am}$ , however, tended to be about an order-of-magnitude higher than those from the Tennessee study. It is possible that this might have been due to the lower pH of our

Table 5  
Summary of calculations to estimate the potential effective actinide dose to a hypothetical 30 year resident consuming vegetables grown on the Par Pond lake bed

Radionuclide	Mean effective concentration <sup>a</sup> (mBq kg <sup>-1</sup> )	30 year consumption <sup>b</sup> (kg)	Activity ingested (Bq)	Dose factor <sup>c</sup> (Sv Bq <sup>-1</sup> )	Dose (mSv)
<sup>241</sup> Am	4.3	6 × 10 <sup>3</sup>	26	9.84 × 10 <sup>-7</sup>	0.026
<sup>244</sup> Cm	1.8	6 × 10 <sup>3</sup>	11	5.45 × 10 <sup>-7</sup>	0.006
<sup>239</sup> Pu	0.4	6 × 10 <sup>3</sup>	2.4	9.56 × 10 <sup>-7</sup>	0.002
<sup>232</sup> Th	6.0	6 × 10 <sup>3</sup>	36	7.38 × 10 <sup>-7</sup>	0.027
<sup>238</sup> U	13	6 × 10 <sup>3</sup>	78	6.88 × 10 <sup>-8</sup>	0.005
Total anthropogenic actinides					0.034
Total naturally occurring actinides					0.032
Total for all actinides					0.066

<sup>a</sup> Concentrations are computed as arithmetic means across all crops studied, on a fresh weight basis.

<sup>b</sup> Based on 200 kg year<sup>-1</sup> (fresh weight) × 30 year. From U.S. EPA (1991).

<sup>c</sup> Eckerman et al. (1988).

soil (3.8-4.2 vs. 7-7.5) rather than clay content differences, since Adriano et al. (1980) demonstrated a roughly order-of-magnitude depression of <sup>241</sup>Am uptake by liming southeastern U.S. soils to reduce their acidity. The decrease in adsorption of the trivalent f-elements with decreasing pH has been demonstrated for SRS soils and sediments (Clark et al., in press). At the pH of the soil used in this study, sorption isotherms suggest that only 50-60% of the trivalent actinides will be sorbed.

#### 4.3. Comparative ingestion health risks

Ingestion of vegetable crops grown on the Par Pond lake bed represents one potential exposure pathway to a hypothetical human resident. The potential effective dose and health risk from actinides via vegetable consumption using an EPA CERCLA Guidance Scenario involving a 30 year self-sufficient resident (U.S. EPA, 1991) were calculated. The effective dose was estimated from the product of the mean actinide concentration across crops (mBq kg<sup>-1</sup>), the total 30 year consumption (kg), and the effective ingestion dose factor (Sv Bq<sup>-1</sup>), as shown in Table 5. The effective dose estimates were then summed over the actinides measured.

It is evident that the anthropogenic and naturally occurring actinides contribute about equally to the effective 30 year dose of ~0.07 mSv from all actinides (Table 5). This value is only about 0.3% of the effective ingestion dose from <sup>137</sup>Cs, which was comparably estimated as ~22 mSv (Seel et al., 1995). The effective dose from actinide ingestion via vegetable crops is over two orders-of-magnitude less than the 1 mSv year<sup>-1</sup> limit for the general public (ICRP, 1992). The effective anthropogenic actinide dose, when multiplied by the most current risk factor for fatal and non-fatal cancer, as well as severe hereditary effects, namely 7.3 × 10<sup>-2</sup> Sv<sup>-1</sup> (ICRP, 1992), produces a lifetime risk of about 2 × 10<sup>-6</sup>.

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