UPTAKE OF PLUTONIUM, AMERICIUM, CURIUM, AND NEPTUNIUM IN PLANTS CULTIVATED UNDER GREENHOUSE CONDITIONS

M. Pimpl, W. Schmidt
Nuclear Research Center Karlsruhe
Health Physics Division

Abstract:

The root-uptake of Np, Pu, Am, and Cm from three different artificially contaminated soils in grass, maize, spring wheat, and potatoes was investigated under greenhouse conditions in pots filled with 9 kg contaminated soil and in lysimeters with a surface area of 0,5 m² containing the soils in undisturbed profils up to a depth of 80 cm. Only the plough layer of 30 cm was contaminated with Np, Pu, Am, and Cm. Crop cultivation was done corresponding to usual practice in agriculture. Results of the 1st vegetation periode are represented. Transfer factors obtained deviate considerably from those which are recommended for the estimation of long-term exposure of man in the Federal Republic of Germany.

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1. Introduction

The release of actinides into the environment after an accident in a nuclear installation is a potential risk to the population. Of main importance are Np, Pu, Am, and Cm which, due to their long half lifes, will be present for a long time in the root zone of the soils and will be available to plants thus entering the terrestrial food chain. In order to be able to estimate the long-term exposure of the population by ingestion of food contaminated with actinides, a precise understanding of the behavior of actinides in the environment is required. Of particular importance is to deepen the knowledge of the uptake from the soil via the roots into the plant while ruling out any deposition whatsoever. As it is not possible to predict the concentration of actinides in plants which were cultivated on soils with known contamination level by aid of soil-to-plant transfer factors available in literature, aim of a research program performed at the Karlsruhe Nuclear Research Center is the measurement of transfer factors from German soils in crops most common in Germany under climatic conditions which are typical for the region. The experiments were started up in April 1983. First results of the 1st vegetation period are now available.

2. Materials and Methods

The experiments were performed in the fully air-conditioned greenhouse of the Karlsruhe Nuclear Research Center. To maintain the climatic data constant and reproducible a temperature during the day of 20 °C and during the night of 15 °C was defined for a length of day of 16 h with an illumination of at least 3000 lux/m². The moisture of the air was specified to be 70%.

Three types of soil collected in the Schwandorf region were used. The first type of soil was granite weathering soil. This is a poor in carbonate, low humous, shallow and very stonyloamy sand. The cation exchange capacity is 10.0 mval/100 g soil; the pH value of 5.2 lies in the slightly acid range.

The second type of soil is a chalk weathering product. This is a poor in carbonate, low humous, little stony and coarsely silty sand of medium depth. The exchange capacity is 6.3 mval. The pH of the soil of 6.8 lies in the neutral range.

The third type of soil is a normal gley soil collected in the flooding zone of a brook. It is a sandy loam with a pH of 5.4. Since this soil stems from a natural grassland site, no A-horizon was collected for pot experiments but only lysimeters were cut from natural deposits. Via a second vessel the ground water level has been simulated and kept constant at 40 cm depth during the period of vegetation. The soil characteristics determined have been compiled in Table 1.

The vessels used were lysimeters containing soil cut from undisturbed deposits (surface area 0.5 m^2), containers filled with about 250 kg A-horizon (surface area: 0,6 m^2), and Kick-Brauckmann pots containing 8-9 kg A-horizon (surface area: 0.035 m^2).

To contaminate the soil Np-237, Pu-238, Am-241 and Cm-244 dissolved in nitric acid, pH approximatelly 1, were used. In case of the Kick-Brauckmann pots the activity was introduced into 9 kg of moistened soil by a simple irrigation method. The amount weighed for each pot was introduced in layers and slightly compacted whereupon the surface of each layer was uniformly humidified with a measured aliquot of the activity containing solution to be added to each pot before the next layer was placed on the preceding one. 16 contaminated layers of 500 g each were covered with 1 kg soil free of activity. The containers were contaminated by the same method. Each layer corresponds to 10 kg of soil. A second mixing technique was applied in parallel. The activity was appropriately diluted, added to the soil and homogeneously distributed in the moistened soil by a mixer.

The lysimeter vessels were pressed in the field into the soil (depth: 80 cm), filled with the respective type of soil from an indisturbed deposit and subsequently brought to the greenhouse. Before the top soil was contaminated a layer of about 3 cm was removed, stored and later-on reused as a cover. Then, the A-horizon was mechanically loosened down to about 30 cm depth. The surface was uniformly moistened with a defined amount of activity contained in a diluted solution. By repeated manual mixing of the A-horizon the radioactivity added was distributed as homogeneously as possible down to a depth of about 30 cm. This was immediately followed by depostion of the covering layer, slight compaction and sowing.

Regarding the permanent grassland site the gley soil type and the root specific distribution of grass after removal of the mat (1-2 cm) was taken into account; only the topmost 10 cm layer was contamined and the mat placed on the layer as a cover. The respective activity contents of lysimeters,containers and Kick-Brauckmann vessels have been compiled in Table 2. The first period of vegetation lasted from June until October 1983. Potatoes, variation "Saphir", maize, variation "Cargill", and spring wheat, variation "Kolibri" were cultivated.

The nutrient requirements were satisfied with "Blau" complex fertilizer (12-12-17-2). In the pot experiments 1 g of fertilizer was mixed with 1 kg soil, in the lysimeters and containers the initial fertilization was spread over the soil surface, the amount being 14 g N/m². Additional nitrogen fertilizer in the form of calcium ammonium nitrate was supplied to promote the emergence of ears in maize and the emergence of vanes in cereals; the amounts being 0.25 g N/pot and 6 g N/m² in containers and lysimeters. Both before contamination and following the first cutting after contamination the grassland lysimeter received a nitrogen supply of 15 g N/m² each. During the period of vegetation aphicide in the form of 0.1% Phosdrin solution was administered to maize and potatoes.

The potatoes were harvested after complete perishing of the foliage; wheat and maize were harvested at the time of full ripeness. The plants were divided into edible parts and parts not suited for consumption. In case of maize, also complete plants were brought to analysis. After drying at 105 °C and complete ashing with nitric acid at 550 °C the samples were subjected to radiochemical analysis.

3. Results and discussion

The concentrations of Pu, Np, Am, and Cm measured in the cultivated plants have been compiled in Tab. 2 together with the corresponding soil concentrations. The transfer factors calculated are shown in Tab. 3.

It should be mentioned that the results represented are of guideline nature with limited validity, because sowing was done immediately after contamination of the soils. Equilibrium between soil and added activity had not been installed and therefore a change of the transfer behavior due to an aging effect of the actinides in soils must be awaited before a final statement of the transferfactors obtained will be possible. Nevertheless, some trends can be recognized in the results of the first vegetation period which are worth to be discussed in detail.

Root uptake of different plant species

When comparing the root uptake in the different plants cultivated it can be seen that the highest concentrations of the nuclides investigated are measured in grass. Likewise very high concentrations were detected in young potatoe plants, whereas maize plants showed a evidently lower uptake. The lowest concentrations were measured in maize ear and wheat grain. The transfer factors reflect the similar trend. For all actinides investigated, the transfer factor for grass is about 10 to 100 times higher than the value obtained for wheat grain.

Influence of the soil type on the root-uptake:

Comparing the uptake in maize ear and wheat grain of the two different agricultural soils used, it can be seen that in case of the granite weathering soil for Np the transfer factor is higher up to a factor of 100 compared to the chalk weathering soil. For Pu, Am, and Cm no conclusion can be drawn at the moment concering the dependence of the root-uptake on the soil type, because all concentrations measured in maize ears and wheat grains up to now are below the lower detection limits. An explanation for the different Np-transfer could be found in the different pH-values of the two soils used: pH = 5.2 in granite weathering and pH = 6.8 in chalk weathering. Whilst from literature data no dependence of the transfer factor from soil pH-value can be detected /1/, extraction experiments show a higher exchange capacity for actinides and with that a higher plant availability at pH = 5 compared to pH = 7 /2/.

Transfer behavior of the different nuclides used:

The transfer factors of the different actinides used increase in the order of Pu $<$ Am $\simeq$ Cm $<$ Np. The transfer factors obtained up to now show no significant difference in the uptake behavior of Am and Cm. Compared to Am and Cm the transfer factor of Pu is lower one order of magnitude for all plants cultivated whereas the uptake of Np is significantly higher. The transfer factors measured for Np are up to 1000 times higher than for Pu.

Influence of the way of contamination on the root-uptake

The respective mixing technique for activity introduction into the soil exerts a great influence on the root-uptake by the plant. Probably this influence depends on the period of time during which the equilibria between the soil and the added actinides establish. In this case the variation of transfer factors caused by different mixing techniques should disappear when soil equilibrium will be reached after some vegetation periods. With the contamination technique mainly used - a irrigation technique - the uptake is higher up to one order of magnitude compared to contamination of the soil by mechanical mixing.

Influence of the size of cultivation vessels and the fertilization

The influence of the vessel size on the transfer behavior of actinides cannot yet be clearly defined because of scarcity of data available. Using Kick-Brauckmann pots and lysimeters there seems to be no significant difference in the uptake behavior. These two extremely differing vessel sizes show during the period of vegetation a great difference in evapotranspiration and besides that there are dissimilar amounts of fertilizier - related to the surface area in case of the lysimeter and related to the weight in case of the pot experiment.

Comparison of the results with data from literature

For Np the transferfactors obtained are in the order of magnitude of 10^{-3}. Extrem values are transfer factors applicable to wheat grains and maize ears of about $2 \cdot 10^{-4}$ compared with $6 \cdot 10^{-2}$ for grass. The measured transfer factors for wheat grains are in excellent agreement with results proposed by Eriksson /3/, who indicates values of $1.2 \cdot 10^{-4}$ to $3.5 \cdot 10^{-2}$. Slightly lower transfer factors were measured by Wallace /4/, the order of magnitude being between $3 \cdot 10^{-5}$ ans $1.4 \cdot 10^{-3}$.

For Pu-238 the transfer factors indicated in literature undergo great variations. For wheat grains Smith et al. /5/ found values up to $2.4 \cdot 10^{-4}$, whilst Wallace /4/ indicates a value of not more than 10^{-7}. Eriksson /3/ found values between 10^{-5} and $6 \cdot 10^{-7}$, which are in good agreement with the results for wheat grains proposed in this paper. For grass the measured transfer factors in the range of $1.7 \cdot 10^{-4}$ to $2.9 \cdot 10^{-4}$ are very much lower than values indicated by Frissel /6/ and Cawse /7/, who measured transfer factors up to $1.1 \cdot 10^{-1}$ and $6.5 \cdot 10^{-1}$, respectively. It must be proofed if deposition and resuspension would have increased these transfer factors obtained in field experiments.

For Am-241 transfer factors obtained for wheat grain are in the range of $\leq 2 \cdot 10^{-6}$ to $\leq 6 \cdot 10^{-5}$. These results agree with values measured by Wallace /4/, Eriksson /3/, and Schulz /9/ who indicate transfer factors in the range of 10^{-7} to 10^{-5}. The transfer factor of $6 \cdot 10^{-4}$ measured for grass is much lower than values proposed by Frissel /6/, obtained in field experiments.

For Cm-244 only few data ara available in literature. The measured transfer factors in the range of $\leq 3 \cdot 10^{-5}$ for wheat grains agree with data from Wallace /4/, but they are much lower then values from Smith /8/ who proposed transfer factors between 1.9 and $3.6 \cdot 10^{-3}$. The transfer factor determined for grass is about $7 \cdot 10^{-4}$ and is in agreement with data from literature /10/.

4. Conclusions

Some of the transfer factors determined, which are to be used to judge the exposure pathways to man, deviate considerably from the basis of calculation used in the Federal Republic of Germany /11/.

For Np a transfer factor of $2.5 \cdot 10^{-3}$ is specified. Compared with the results obtained this value is too low by a factor of 10, because even for wheat grain a transfer factor of $1.5 \cdot 10^{-2}$ could be attained.

The basis of calculation for Pu is $2.5 \cdot 10^{-4}$. This value conforms to the values determined for grassland, but in case of maize and likewise wheat the transfer factors are much lower than suggested by the basis of calculation.

A similar evaluation can be made for Am for which the basis of calculation is also $2.5 \cdot 10^{-4}$. Unlike for Pu, the transfer factors in case of grassland plants are slightly higher than $2.5 \cdot 10^{-4}$.

For Cm the basis of calculation is $2.5 \cdot 10^{-3}$. The transfer factors measured for grassland are lower at least by a factor of 10, for wheat grains the obtained values are by more than 100 times lower.

In conclusion the guideline nature of the described results derived from the first vegetation period should be underlined. It remains to be examined how in the long run above all aging of the transuranics in the soil in general and parameter variation in particular will exert an influence on the transfer of these elements from the soil into the plant. With the knowledge then available the pathway of Np, Pu, Am, and Cm to man by soil-to-plant transfer via root upstake can be judged more reliable than today.

Literature

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Tab. 1: Chemical and physical properties of the soils

		Granite Weathering	Chalk Weathering	Glisoil
pH ($CaCl_2$)		5.2	6.8	5.4
Organic C	%	2.1	1.5	7.1
Clay	%	5.8	5.5	
Silt	%	19.8	32.9	
Fine sand	%	10.0	17.0	
Total N	%	0.14	0.08	0.40
Ca	meq/100g	5.4	4.4	
T-value		10.0	6.3	
S-value		8.1	5.5	
Content of nutrients (one step extraction)				
P_2O_5	ppm	140	440	140
K_2O	"	580	370	200
MgO	"	120	50	
B	"	0.34	0.30	
Na	"	24	18	
Cu		2.5	3.2	
Mn	"	129	314	
Mo	"	0.24	0.47	

Tab. 2: Concentrations of Np, Pu, Am, and Cm in soils and plants

kind of container	Nr	soil	activity in pCi/g dry soil				Plant 1983	activity in fCi/g dry plant			
			^{237}Np	^{238}Pu	^{241}Am	^{244}Cm		^{237}Np	^{238}Pu	^{241}Am	^{244}Cm
pot	9	chalk weathering	71	204	218	186	maize	455	5.6	9.8	2.0
"	10		77	220	235	200		349	5.6	12.5	7.7
"	11		74	212	226	193		487	7.8	16.8	7.5
"	12		74	212	226	193		516	15.1	13.0	9.3
pot	15	chalk weathering	75	214	228	195	summer wheat (grain)	-	-	≤12.8	≤5.1
"	16		70	200	214	183		62	≤4.7	≤ 1.8	1.8
"	17		75	214	229	195		48	-	-	≤5.9
"	18		73	211	225	192		31	≤9.5	≤ 6.0	≤3.4
pot	33	granite weathering	86	296	354	218	summer wheat (grain)	751	≤7.8	≤15.9	2.4
"	34		85	294	351	216		1297	≤7.2	≤ 4.1	≤2.4
"	35		85	294	351	216		618	≤4.5	-	-
"	36		85	292	349	216		1135	≤7.0	4.4	3.1
pot	37	chalk weathering	83	286	342	212	maize	872	12.5	138	40
"	38		83	286	342	212		1598	18.2	187	121
"	39		83	286	342	212		1616	15.4	171	62
"	40		83	286	342	212		1030	13.0	112	32
lysimeter	C 1	chalk weathering	284	206	219	176	potatoes (young plants)	17400	97	554	150
"	C 2	chalk weathering	142	206	219	88	maize (young plants)	1330	26	10	7
"	C 3	chalk weathering	142	206	219	88	summer wheat (grain)	104	≤3.9	≤5.4	1.5
								109	≤4.3	-	-
								-	-	≤1.4	1.2
								36	≤1.9	≤0.6	≤0.6
"	C 5	granite weathering	81	234	249	50	maize (ear)	505	≤0.4	-	-
"	C 6	granite weathering	81	234	249	50	summer wheat (grain)	388	≤1.4	-	-
								501	≤2.1	-	-
								274	2.4	-	-
								339	≤4.3	-	-
"	C 7	glisoil	178	515	548	165	pasture 1.cut	≤0.2	≤1.8	1.6	0.4
							2.cut	8030	87	345	85
							3.cut	10150	94	307	157
							4.cut	8200	149	348	121
container	C 8	granite weathering	33	372	136	56	summer wheat (grain)	948	≤6.9	-	-
								389	≤2.4	-	-
"	C 9	granite weathering	41	457	167	69	maize (ear)	108	≤1.4	-	-
"	C 11	chalk weathering	32	365	133	82	summer wheat (grain)	21.5	14.0	4.1	3.5
								19.2	23.7	-	-
"	C 12	chalk weathering	32	363	132	82	maize (ear)	3.7	1.2	-	-

Tab. 3 Transferfactors, calculated on dry matter base

Plant	Soil type		^{237}Np	^{238}Pu	^{241}Am	^{244}Cm
Pasture	Glisoil	Lys. C 7	$4.5\cdot10^{-2}$	$1.7\cdot10^{-4}$	$6.3\cdot10^{-4}$	$5.2\cdot10^{-4}$
			$5.7\cdot10^{-2}$	$1.8\cdot10^{-4}$	$5.6\cdot10^{-4}$	$9.5\cdot10^{-4}$
			$4.6\cdot10^{-2}$	$2.9\cdot10^{-4}$	$6.4\cdot10^{-4}$	$7.3\cdot10^{-4}$
Summer wheat (grain)	chalk weathering	Lys. C 3	$7.3\cdot10^{-4}$	$\le1.9\cdot10^{-5}$	$\le2.5\cdot10^{-5}$	$\le1.7\cdot10^{-5}$
			$7.7\cdot10^{-4}$	$\le2.1\cdot10^{-5}$	-	-
			-	-	$\le0.6\cdot10^{-5}$	$\le1.4\cdot10^{-5}$
			$2.5\cdot10^{-4}$	$\le0.9\cdot10^{-5}$	$\le0.2\cdot10^{-5}$	$0.7\cdot10^{-5}$
		Lys. C 11	$6.7\cdot10^{-4}$	$3.8\cdot10^{-5}$	$\le3.1\cdot10^{-5}$	$\le4.3\cdot10^{-5}$
			$6.0\cdot10^{-4}$	$6.5\cdot10^{-5}$	-	-
		pot 15	-	-	$\le5.6\cdot10^{-5}$	$\le2.6\cdot10^{-5}$
		pot 16	$8.9\cdot10^{-4}$	$\le2.4\cdot10^{-5}$	$\le0.8\cdot10^{-5}$	$\le0.9\cdot10^{-5}$
		pot 17	$6.4\cdot10^{-4}$	-	-	$3.0\cdot10^{-5}$
		pot 18	$4.2\cdot10^{-4}$	$\le4.5\cdot10^{-5}$	$\le2.7\cdot10^{-5}$	$\le1.8\cdot10^{-5}$
	granite weathering	Lys. C 6	$47.9\cdot10^{-4}$	$\le0.5\cdot10^{-5}$	-	-
			$61.9\cdot10^{-4}$	$\le0.9\cdot10^{-5}$	-	-
			$33.8\cdot10^{-4}$	$\le1.0\cdot10^{-5}$	-	-
			$41.9\cdot10^{-4}$	$\le1.8\cdot10^{-5}$	-	-
		Lys. C 8	$287.3\cdot10^{-4}$	$\le1.9\cdot10^{-5}$	-	-
			$117.9\cdot10^{-4}$	$\le0.6\cdot10^{-5}$	-	-
		pot 33	$87.3\cdot10^{-4}$	$\le2.6\cdot10^{-5}$	$4.5\cdot10^{-5}$	$\le1.1\cdot10^{-5}$
		pot 34	$152.6\cdot10^{-4}$	$\le2.5\cdot10^{-5}$	$\le1.2\cdot10^{-5}$	$\le1.1\cdot10^{-5}$
		pot 35	$72.7\cdot10^{-4}$	$\le1.5\cdot10^{-5}$	-	-
		pot 36	$133.5\cdot10^{-4}$	$\le2.4\cdot10^{-5}$	$\le1.3\cdot10^{-5}$	$\le1.4\cdot10^{-5}$
Maize (young plant)	chalk weathering	Lys. C 2	$9.4\cdot10^{-3}$	$12.6\cdot10^{-5}$	$4.6\cdot10^{-5}$	$\le2.3\cdot10^{-5}$
Maize (ear)		Lys. C 12	$0.1\cdot10^{-3}$	$0.3\cdot10^{-5}$	-	-
Maize (total plant)		pot 9	$6.4\cdot10^{-3}$	$2.8\cdot10^{-5}$	$4.5\cdot10^{-5}$	$1.1\cdot10^{-5}$
		pot 10	$4.5\cdot10^{-3}$	$2.6\cdot10^{-5}$	$5.3\cdot10^{-5}$	$3.9\cdot10^{-5}$
		pot 11	$6.6\cdot10^{-3}$	$3.7\cdot10^{-5}$	$7.4\cdot10^{-5}$	$3.9\cdot10^{-5}$
		pot 12	$7.0\cdot10^{-3}$	$7.1\cdot10^{-5}$	$5.8\cdot10^{-5}$	$4.8\cdot10^{-5}$
		pot 37	$10.5\cdot10^{-3}$	$4.4\cdot10^{-5}$	$40.4\cdot10^{-5}$	$18.9\cdot10^{-5}$
		pot 38	$19.3\cdot10^{-3}$	$6.4\cdot10^{-5}$	$54.7\cdot10^{-5}$	$57.1\cdot10^{-5}$
		pot 39	$19.5\cdot10^{-3}$	$5.4\cdot10^{-5}$	$50.0\cdot10^{-5}$	$29.3\cdot10^{-5}$
		pot 40	$12.4\cdot10^{-3}$	$4.6\cdot10^{-5}$	$32.8\cdot10^{-5}$	$15.1\cdot10^{-5}$
Maize (ear)	granite weathering	Lys. C 5	$6.2\cdot10^{-3}$	$\le0.2\cdot10^{-5}$	-	-
Maize (ear)		Lys. C 9	$2.6\cdot10^{-3}$	$\le0.3\cdot10^{-5}$	-	-
Potatoes (young plant)	chalk weathering	Lys. C 1	$61.3\cdot10^{-3}$	$47.1\cdot10^{-5}$	$253.0\cdot10^{-5}$	$85.2\cdot10^{-5}$