

**PRELIMINARY ANALYSIS OF FIRST CROP
OF PLANTS GROWN IN SEVEN SOILS
UNIFORMLY CONTAMINATED WITH FOUR
TRANSURANIC ELEMENTS SIMULTANEOUSLY**

Quarterly Progress Report

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ABSTRACT

A forage crop and a grain crop of wheat were harvested from a replicated experiment with about 200 kg soil per container in which seven soils from different locations throughout the U.S.A. were spiked with four different transuranic elements simultaneously. Concentration Ratios (C.R.) for the radioactive elements in plants were obtained as a start to careful examination of parameters involved in the use of these ratios in models. The relative availability of the different radionuclides differed considerably with soil. One order of magnitude differences were common for element pairs. In general the order of plant availability was $Np > Am > Cm > Pu$. The differences that were encountered for soil for radionuclides present in straw were much less pronounced in the grain. The ratio of maximum (usually for the acid sandy soil) to minimum (usually for the soil with high soil organic matter) for straw varied from 1.9 to 2.5 orders of magnitude, but was one order or less for grain. This means that a small and more constant amount of transuranium element is transferred to grain relative to that in straw. The behavior is somewhat analogous to that of calcium. The results are to be very useful in refining of models.

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The seven soils used in this project to represent a wide range of soils of the USA have been described in considerable detail in previous reports. These soils were chosen to provide the necessary range in parameters to more precisely determine and verify transuranic element transfer coefficients being used in models by the NRC. The amounts of spiked transuranic elements used in the study are shown in Table 1.

Wheat was the first crop grown in the large pots which were designed to simulate field conditions. A forage crop was removed when plants were about 10 cm tall. This crop was divided into two portions--lower 5cm. and above 5 cm. so that any plant contamination above the sand barrier could be detected.

The plants were then allowed to produce a crop of wheat grain. Plants were divided into grain, chaff and straw. Yields are shown in Table 2. Even though lime and fertilizers were applied, the acid Lyman soil produced a relatively lower yield of wheat than did other soils.

These plant materials were assayed for ^{237}Np , ^{239}Pu , ^{241}Am , and ^{244}Cm by the methods outlined in the July 1978 quarterly report.

The concentration ratios (C.R.) for the forage crop are in Table 3. It is noted that the analyses are not fully complete at this time and will be soon. Of interest is the fact that generally the upper parts of the plants contained more of each of the transuranic elements than did the lower parts. This indicates that contamination was not present in the system. The CV values were lowest for ^{244}Cm .

Ratios of transuranic element pairs for the forage crop are in Table 4. These indicate the relative availability of the four different transuranic elements. This generally was $Np > Am > Cm > Pu$. Large differences were noted for the different soils. Since plants were not harvested at this stage from the Lyman soil, this point will be discussed later.

Analyses are not fully complete for the mature wheat grain and straw crop and those data which are available are in Table 5. The coefficient of variation (C.V.) for the five replicates (unless incomplete) was less than 50% in around half the cases, occasionally near 100% and often 20% or less. One element was not consistently different in variability from another. Full evaluation of the meaning of the variability encountered here will be made when more data are available.

The maximum over minimum C.R. values for each transuranic element and each plant part are also given in Table 5. Widest ratios were for straw and most narrow for grain. The C.R. values generally were in the order $Np > Am > Cm > Pu$. Lyman soil generally resulted in highest C.R. values and Egbert resulted in the lowest. As much as 2.5 orders of magnitude difference were noted for some elements.

Table 6 serves as a partial summary of Table 5. Within a given plant part, there were wide variations in the ratio of Np/Pu . These reflect in part the high uptake rate for Lyman soil (acid sandy) and the low uptake rate for Egbert (high soil organic matter).

For straw, the Np/Pu ratio of C.R.'s varied from 23 to 538, for Np/Am from 16 to 73, for Np/Cm from 10 to 171, for Am/Pu from 1.46 to 25, for Cm/Pu from 0.14 to 28, and for Am/Cm 0.9 to 10.7. This data set is not fully complete but the wide ratios are of interest.

Eventually the soil characteristics will be used in regression analysis with these and additional data.

Plant part discrimination for the wheat plant parts are in Table 7. Egbert soil with the least uptake of transuranic elements had lowest discrimination ratios. This means that concentrations in grain may not be fully related to that in straw. The levels in grain tended somewhat toward uniformity in spite of straw levels. Lyman soil generally resulted in highest plant part ratios indicating that the highest levels in stems did not necessarily result in proportionately higher amounts in grain.

This experiment is not fully complete and additional crops have been grown and more also will be grown in these same containers to give long-time effects. Full synthesis will be made when more data are available.

In the section that follows, there is evidence of a good correlation between transuranic element uptake and chemical extractability from soil.

EXTRACTABILITY OF TRANSURANIC ELEMENTS IN SOILS

A previous report (Annual Report, 1978, NUREG/CR-1057) presented the extractability of ^{237}Np , $^{239,240}\text{Pu}$, and ^{241}Am from the seven soils that are being used for plant uptake experiments. The extractability was determined by using 0.05 M calcium chloride as the extractant. This report gives the extractability of ^{237}Np , $^{239,240}\text{Pu}$, ^{241}Am , and ^{244}Cm by using 1.0 M ammonium acetate, which had been pH-adjusted to the same pH as the soil being treated. The procedure, otherwise, was the same as described previously (Annual Report, 1978, NUREG/CR-1057).

The extractability of the transuranics with respect to different soils are shown in Table 8. A consistent extractability effect that occurred among the four transuranic elements studied was as follows: Egbert < Yolo < Aiken < Sharpsburg < Lyman. Except with $^{239,240}\text{Pu}$, the extractability of the transuranics from the Aiken soil was greater than that from Holtville and less than that of Malbis. Comparison with previous results (Annual Report, 1978), the 1 M ammonium acetate consistently extracted greater amount of the transuranics than 0.05 M calcium chloride. The major part of this difference is considered to be the stronger complexing action of the ammonium acetate.

Table 1. Picocuries per g. of transuranic elements mixed simultaneously into seven different soils.

<u>Soil</u>	<u>^{237}Np</u>	<u>^{239}Pu</u>	<u>^{241}Am</u>	<u>^{244}Cm</u>
Aiken	352	35236	3524	3524
Egbert	536	53591	5359	5359
Holtville	250	24963	2496	2496
Lyman	320	31969	3197	3197
Malbis	279	27871	2787	2787
Sharpsburg	305	30469	3047	3047
Yolo	282	28233	2823	2823

Table 2. Yield (g dry wt) in grams per container--wheat experiment.

Wheat (forage crop)			
<u>Soil</u>	<u>>5 cm</u> *	<u>0-5 cm</u> *	<u>Total</u>
Aiken	116.52	27.30	143.82
Egbert	84.24	22.40	106.64
Holtville	117.75	28.41	146.16
Lyman	Not sampled		
Mablis	99.62	26.16	125.78
Sharpsburg	127.53	30.77	158.30
Yolo	148.90	26.16	175.06
Wheat (mature crop)			
<u>Soil</u>	<u>Grain</u>	<u>Chaff</u>	<u>Straw</u>
Aiken	1601.1	509.0	676.7
Egbert	1581.5	475.5	521.6
Holtville	1493.1	591.5	722.6
Lyman	130.4	150.6	117.4
Mablis	1278.1	515.9	623.3
Sharpsburg	1405.4	605.8	678.5
Yolo	1417.2	565.7	752.3

* Plants were divided at below and above 5 cm from sand level.

Table 3. Transuranic element concentration ratios (CR) of the wheat forage crop (data are preliminary/subject to revision).

Sample *	^{237}Np			^{239}Pu			^{241}Am			^{244}Cm		
	n	Mean CR	CV%	n	Mean CR	CV%	n	Mean CR	CV%	n	Mean CR	CV%
<u>Aiken</u> (>5cm) (0-5)	4	2.60×10^{-2}	24.9	3	6.52×10^{-5}	27.0	5	1.03×10^{-3}	46.5	5	7.49×10^{-4}	7.9
	5	1.48×10^{-2}	41.3	5	5.91×10^{-5}	48.4	4	1.87×10^{-3}	67.4	5	6.09×10^{-4}	15.0
<u>Egbert</u> (>5) (0-5)	2	3.02×10^{-3}	86.1	1	8.51×10^{-5}	-	3	4.66×10^{-4}	54.3	-	-	-
	-	-	-	-	-	-	2	9.32×10^{-4}	45.2	-	-	-
<u>Holtville</u> (>5) (0-5)	5	1.03×10^{-2}	18.6	4	5.01×10^{-4}	34.3	5	3.46×10^{-4}	88.7	3	3.74×10^{-5}	22.4
	5	6.22×10^{-3}	51.9	2	3.43×10^{-4}	33.8	3	3.87×10^{-4}	38.2	3	4.14×10^{-5}	29.7
<u>Lyman</u> not sampled												
<u>Mablis</u> (>5) (0-5)	5	3.94×10^{-2}	13.4	4	1.65×10^{-4}	21.0	5	9.42×10^{-4}	98.4	5	3.61×10^{-4}	16.1
	5	2.45×10^{-2}	47.3	5	1.45×10^{-4}	27.9	5	5.91×10^{-4}	29.3	5	3.22×10^{-4}	15.8
<u>Sharpsburg</u> (>5) (0-5)	5	4.11×10^{-2}	4.2	5	8.42×10^{-5}	33.6	5	3.98×10^{-4}	68.8	5	2.80×10^{-4}	8.1
	5	1.40×10^{-2}	31.6	5	4.14×10^{-5}	36.2	3	2.90×10^{-4}	43.1	5	2.21×10^{-4}	16.9
<u>Yolo</u> (>5)	5	9.97×10^{-3}	11.6	5	2.58×10^{-4} **	130.6	1	1.20×10^{-4}	-	5	5.61×10^{-5}	11.3
				4	1.08×10^{-4}	11.4						
	5	6.08×10^{-3}	30.9	5	8.89×10^{-5}	14.8	3	3.33×10^{-4}	108.7	5	4.96×10^{-5}	16.7

* 0.5 cm and 75 cm are height of plant from sand layer.

** Five replications.

Table 4. Ratios of concentration ratios (or relative availability) for pairs of transuranic elements (wheat forage crop).

Sample	Plant section	$^{239}\text{Np}/^{239}\text{Pu}$	$^{237}\text{Np}/^{241}\text{Am}$	$^{237}\text{Np}/^{244}\text{Cm}$	$^{241}\text{Am}/^{239}\text{Pu}$	$^{244}\text{Cm}/^{239}\text{Pu}$	$^{241}\text{Am}/^{244}\text{Cm}$
<u>Soil</u>							
Aiken	(5cm)	399.	25.2	34.7	15.8	11.5	1.38
	(0-5cm)	250.	7.91	24.3	31.6	10.3	3.07
Egbert	(5)	35.5	6.48	-	5.48	-	-
Holtville	(5)	20.6	29.8	275.	0.691	0.0747	9.25
	(0-5)	18.1	16.1	150.	1.13	0.121	9.35
Lyman	Not sampled						
Mablis	(5)	239.	41.8	109.	5.71	2.19	2.61
	(0-5)	169.	41.5	76.1	4.08	2.22	1.84
Sharpsburg	(5)	488.	103.	147.	4.73	3.33	1.42
	(0-5)	338.	48.3	63.3	7.00	5.34	1.31
Yolo	(5)	92.3	83.1	178.	1.11	0.519	2.14
	(0-5)	68.4	183.	123.	3.75	0.558	6.71

Table 5. Concentration ratios for the mature wheat crop.

Sample	^{237}Np			^{239}Pu								
	n	Mean CR	C.V.%	n	Mean CR	C.V.%	n	Mean CR	C.V.%	n	Mean CR	C.V.%
Grain												
Aiken	5	3.87×10^{-4}	28.7	5	3.63×10^{-6}	81.8	2	7.38×10^{-5}	-	5	5.96×10^{-6}	10.6
Egbert	2	2.06×10^{-4}	64.1	4	1.83×10^{-6}	54.1	-	-	-	-	-	-
Holtville	5	1.38×10^{-4}	79.7	5	4.75×10^{-6}	82.7	4	8.48×10^{-5}	53.8	-	-	-
Lyman	4	1.43×10^{-3}	109.8	4	9.47×10^{-7}	45.0	2	4.56×10^{-5}	71.7	5	1.22×10^{-5}	20.2
Mablis	5	5.51×10^{-4}	98.0	5	2.51×10^{-6}	37.9	3	3.61×10^{-5}	17.0	-	-	-
Sharpsburg	4	3.51×10^{-4}	30.8	4	4.60×10^{-7}	51.1	5	2.91×10^{-5}	48.1	-	-	-
Yolo	5	3.16×10^{-4}	63.9	5	1.66×10^{-6}	45.6	3	1.72×10^{-5}	22.0	-	-	-
Max/min		10.4			10.3			4.9 (?)			2.0 (?)	
Chaff												
Aiken	5	6.40×10^{-3}	24.8	5	1.44×10^{-5}	43.1	5	3.84×10^{-4}	59.3	5	2.31×10^{-4}	18.3
Egbert	5	3.36×10^{-4}	50.0	5	2.43×10^{-6}	28.3	5	6.38×10^{-5}	149.1	3	2.80×10^{-6}	33.2
Holtville	3	2.68×10^{-3}	21.7	3	4.68×10^{-5}	22.2	3	1.19×10^{-4}	97.5	5	3.18×10^{-5}	134.6
Lyman	5	3.69×10^{-2}	109.6	5	5.71×10^{-5}	104.7	5	5.94×10^{-4}	13.4	5	4.45×10^{-4}	30.6
Mablis	5	7.75×10^{-3}	16.0	5	1.39×10^{-5}	22.4	5	1.23×10^{-4}	14.6	5	8.82×10^{-5}	14.2
Sharpsburg	5	1.02×10^{-2}	36.6	4	1.71×10^{-5}	46.6	5	1.94×10^{-4}	26.8	5	7.29×10^{-5}	20.4
Yolo	5	4.61×10^{-3}	57.5	5	2.34×10^{-5}	48.3	5	1.49×10^{-4}	88.6	5	5.22×10^{-5}	141.6
Max/Min		109.8			23.5			9.3			158.9	
Straw												
Aiken	5	1.57×10^{-2}	15.7	5	2.92×10^{-5}	20.4	4	7.15×10^{-4}	38.0	5	8.19×10^{-4}	15.1
Egbert	5	5.34×10^{-4}	42.1	5	1.99×10^{-6}	62.8	2	2.68×10^{-5}	32.5	5	6.34×10^{-6}	39.4
Holtville	5	4.54×10^{-3}	13.0	5	1.94×10^{-4}	49.1	4	2.84×10^{-4}	41.5	5	2.66×10^{-5}	33.4
Lyman	5	7.93×10^{-2}	102.8	5	4.87×10^{-5}	179.1	5	2.19×10^{-3}	25.7	5	1.97×10^{-3}	21.6
Mablis	5	1.92×10^{-2}	16.8	5	4.95×10^{-5}	13.0	5	5.61×10^{-4}	9.3	5	3.10×10^{-4}	7.7
Sharpsburg	5	2.51×10^{-2}	19.4	5	4.35×10^{-5}	17.4	5	3.64×10^{-4}	21.0	5	2.40×10^{-4}	15.0
Yolo	5	1.02×10^{-2}	44.7	5	8.96×10^{-5}	-2.1	5	1.39×10^{-4}	18.7	5	7.86×10^{-5}	20.0
Max/Min		148.5			244.7			81.7			310.7	

Table 6. Ratios of concentration ratios (or relative availability) for pairs of transuranic elements (crop grown to maturity).

Soil	$^{239}\text{Np}/^{239}\text{Pu}$	$^{237}\text{Np}/^{241}\text{Am}$	$^{237}\text{Np}/^{244}\text{Cm}$	$^{241}\text{Am}/^{239}\text{Pu}$	$^{244}\text{Cm}/^{239}\text{Pu}$	$^{241}\text{Am}/^{244}\text{Cm}$
Grain						
Aiken	107.	5.24	64.9	20.3	1.64	12.4
Egbert	11.3	-	-	-	-	-
Holtville	29.1	1.63	-	17.9	-	-
Lyman	1510.	31.4	117.	48.2	12.9	3.74
Mablis	15.3	-	14.4	-	-	-
Sharpsburg	763.	12.1	-	63.3	-	-
Yolo	190.	18.4	-	10.4	-	-
Chaff						
Aiken	444.	16.7	27.7	26.7	16.0	1.66
Egbert	138.	5.27	120.	26.3	1.15	22.8
Holtville	57.3	22.5	84.3	2.54	0.679	3.74
Lyman	646.	62.1	82.9	10.4	7.79	1.33
Mablis	558.	63.0	87.9	8.85	6.35	1.39
Sharpsburg	596.	52.6	140.	11.3	4.26	2.66
Yolo	197.	30.9	88.3	6.37	2.23	2.85
Straw						
Aiken	538.	22.0	19.2	24.5	28.0	0.873
Egbert	268.	19.9	84.2	13.5	3.19	4.23
Holtville	23.4	16.0	171.	1.46	0.137	10.7
Lyman	163.	36.2	40.3	4.50	4.05	1.11
Mablis	388.	34.2	61.9	11.3	6.26	1.81
Sharpsburg	577.	69.0	10.5	8.37	5.52	1.52
Yolo	114.	73.4	130.	1.55	0.877	1.77

Table 7. Plant part discrimination ratios.

Soil	^{237}Np			^{239}Pu			^{241}Am			^{244}Cm		
	$\frac{\text{Straw}}{\text{Chaff}}$	$\frac{\text{Straw}}{\text{Grain}}$	$\frac{\text{Chaff}}{\text{Grain}}$	$\frac{\text{Straw}}{\text{Chaff}}$	$\frac{\text{Straw}}{\text{Grain}}$	$\frac{\text{Chaff}}{\text{Grain}}$	$\frac{\text{Straw}}{\text{Chaff}}$	$\frac{\text{Straw}}{\text{Grain}}$	$\frac{\text{Chaff}}{\text{Grain}}$	$\frac{\text{Straw}}{\text{Chaff}}$	$\frac{\text{Straw}}{\text{Grain}}$	$\frac{\text{Chaff}}{\text{Grain}}$
Aiken	2.5	40.6	16.5	2.0	8.0	4.0	1.8	9.7	5.2	3.5	137.4	38.8
Egbert	1.6	2.6	1.6	0.8	1.1	1.3	0.4	-	-	2.3	-	-
Holtville	1.7	3.3	1.9	4.1	40.8	8.9	2.4	3.3	1.4	0.8	-	-
Lyman	2.1	55.4	25.8	8.5	514.2	60.3	3.7	48.0	13.0	4.4	161.5	36.5
Mablis	2.5	34.8	14.1	3.6	19.7	5.5	4.6	15.5	3.4	3.5	-	-
Sharpsburg	2.5	71.5	29.1	2.5	94.6	37.1	1.9	12.5	6.7	3.9	-	-
Yolo	2.2	32.3	14.6	3.8	54.0	14.1	0.9	8.1	8.7	1.5	-	-

Table 8. Extractability of ^{237}Np , $^{239,240}\text{Pu}$, ^{241}Am , and ^{244}Cm by 1.0 M ammonium acetate, which had been pH-adjusted to the same pH as the soil being treated.

Soil series	Soil extract pH*	^{237}Np			$^{239,240}\text{Pu}$			^{241}Am			^{244}Cm		
		Soil activity d/m/g	Extractable d/m/g	Extractable %**	Soil activity d/m/g	Extractable d/m/g	Extractable %**	Soil activity d/m/g	Extractable d/m/g	Extractable %**	Soil activity d/m/g	Extractable d/m/g	Extractable %**
		$\times 10^2$			$\times 10^4$			$\times 10^3$			$\times 10^3$		
Egbert	7.04	11.9	35.56	2.99	11.9	63.61	0.05	11.9	14.66	0.12	11.9	15.83	0.13
Holtville	7.88	5.5	88.65	16.2	5.5	407.22	0.74	5.5	18.83	0.34	5.5	8.67	0.16
Yolo	6.54	6.3	77.62	12.32	6.3	153.89	0.24	6.3	41.30	0.65	6.3	50.72	0.81
Aiken	6.24	7.8	136.47	17.50	7.8	277.73	0.36	7.8	124.80	1.60	7.8	158.00	2.03
Malbis	6.54	6.2	129.47	20.88	6.2	180.84	0.29	6.2	122.59	2.00	6.2	176.83	2.85
Sharpsburg	6.09	6.8	180.65	26.57	6.8	284.97	0.42	6.8	195.58	2.88	6.8	251.30	3.70
Lyman	5.65	7.1	177.73	25.03	7.1	4012.90	5.65	7.1	465.80	6.56	7.1	396.28	5.58

* Soil extract pH after 72 h equilibration period.

** Percent of soil activity (dose).

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