

TRANSURANIC ELEMENT TRANSPORT IN AGRICULTURAL SYSTEMS (SOIL TO FOOD CHAIN TRANSFER OF NUCLEAR FUEL CYCLE RADIONUCLIDES)

Quarterly Progress Report
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QUARTERLY PROGRESS REPORT

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This quarterly report documents some of our analytical procedures for assay of four transuranium radionuclides. The procedures are being used in the assay of wheat grain and wheat plants from our large container experiment conducted at UCLA. The data will be available for the Annual Report.

After the wheat harvest, soybeans were planted as the second crop. This experiment is progressing satisfactorily.

Some small container experiments are being made with stable Cs and stable Sr as factors influencing plant uptake and distribution of radioactive Cs and Sr. We suspect that this is an item of importance in aging of radionuclide contamination, especially under waste disposal and storage conditions.

Studies continue with ^{241}Am in individual small containers, and the studies are being expanded to other transuranium elements.

At Berkeley the scale experiments with wheat grown on identical soils having identical treatments to those at UCLA, but with variable containers size, were harvested and are being analyzed. Analytical cross-checking is being made between our groups. Soybeans and are also being grown at Berkeley. An additional experiment at Berkeley in which soil pH was altered is in progress.

A review of transuranium element uptake entitled "Radionuclide Uptake by Plants" by H. Nishita, A. Wallace, and E. M. Romney was prepared during the quarter. Copies as UCLA No. 12-1158 were sent to NRC to be prepared for distribution.

ANALYTICAL RADIOCHEMISTRY OF THE TRANSURANICS
IN AGRICULTURAL PLANT SYSTEMS

By

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ABSTRACT

A method is described for the isolation and determination of the transuranium metals in various agricultural plant systems. Precipitation and ion exchange techniques are used to isolate, essentially carrier free, the isotopes of ^{237}Np , 239 , ^{240}Pu and ^{241}Am from 1 - 50 g plant samples. Chemical yields range from $\geq 80\%$ for Np to $\geq 90\%$ for Pu and Am. The samples are sufficiently free of contaminant metals after analysis that direct radioassay can be done without time-consuming electroplating techniques being applied. Alpha spectra are essentially free of scatter or self absorption. Transuranic contamination in the chemical reagents used in the analysis as well as the slight cross contamination between samples by way of the glass ion exchange columns limit our sensitivity. However, as these areas of contamination are identified and corrected, sensitivities of 0.1 d/m/g of plant are not unrealistic goals in future analyses.

ANALYTICAL RADIOCHEMISTRY OF THE TRANSURANICS IN AGRICULTURAL PLANT SYSTEMS

This report covers the radioanalytical chemistry techniques developed by this laboratory to assess the cycling behavior of ^{237}Np , 239 , ^{240}Pu , ^{241}Am and ^{244}Cm in several different agricultural systems. A primary requirement of the method to be used by this laboratory is its adaptability to the kinds of plant material to be analyzed and to the equipment available to us. As a result, we examined many analytical techniques in these reports as well as adding additional steps where needed to assure quantitative isolation and determination of the desired isotopes.

The major difference of our methods from those described by (1-6) is the elimination of time-consuming electroplating techniques for final mounting of the samples. The final fractions before counting are essentially carrier-free and contain less than 0.001 mg of solid material. Evaporation and flaming produce a sample that yields alpha spectra essentially free of scatter or self absorption.

In the procedure, plant samples (1-50 g) are ashed at 525°C for 48 hours. The ashes are transferred to 250 ml Teflon beakers and solubilized by successive fuming with 10 ml portions of concentrated HNO_3 , HF and 60% HClO_4 . The silica is removed as the volatile tetrafluorides. The remaining salts are converted to soluble chlorates and nitrates during the fuming step. The salts are dissolved in hot distilled water and diluted to known volumes sufficient to maintain the salts in solution.

Routinely, three aliquots (20% of the samples) are placed in 50 ml tygon centrifuge tubes. Ten milligrams of iron as Fe^{+3} are added to the samples

and the transuranics are co-precipitated with iron hydroxide using ammonium hydroxide as the precipitation agent. Reduction of the actinides to oxidation states suitable for ion exchange separation of the desired isotopes is achieved by dissolving the hydroxides in $\text{HCl-NH}_2\text{OH}$ and heating the samples in a water bath for 1 hr. The transuranics are separated from many of the metal contaminants by selective absorption on AG50W-X4 cation exchange resin contained in water jacketed glass ion exchange columns (1). The columns are preheated and maintained at 50°C . Selective washings of the pregnant resin beds with 50 ml of 0.1 M HCl, 25 ml of 1 M HCl, 50 ml 0.1 M HCl in CH_3OH , 25 ml of 1.0 M HCl in CH_3OH and finally 8 ml of 1.5 M HCl free the transuranics of most of the associated metal contaminants. The $^{239,240}\text{Pu}$, ^{244}Cm , ^{241}Am and ^{237}Np are stripped from the resin by the addition of 12 M HCl (40 ml). The eluates are evaporated to dryness in 50 ml teflon beakers. The trace residues are dissolved in 1 ml of 16 M HNO_3 . The nitric acid phases are transferred to stainless steel planchets and evaporated to dryness under a heat lamp. The planchets are flamed to a dull redness using a bunsen burner. The samples are then alpha counted using silicon surface barrier alpha detectors in conjunction with a 4095 channel analyzer. The analyzer permits counting of 8 samples simultaneously. Each set of 16 samples processed during any one run consists of four samples run in triplicate plus two standards and two reagent blanks.

DISCUSSION

Chemical Yields

Chemical yields are determined by analyzing background plant samples containing known tracer quantities of ^{237}Np , $^{239,240}\text{Pu}$, ^{241}Am and ^{244}Cm run in parallel with the samples. The standards contain identical amounts of plant material as the samples. The standards are run identically as the

samples. The quantity of tracer activity that is added to the standard plant samples is plated directly on stainless steel planchets. The chemical yields are determined by comparing the tracer activity recovered in the plant standards after analysis to that of the directly plated out standard activity counted under identical conditions.

Chemical yield values (Table 1) range from 94-98% for $^{239,240}\text{Pu}$, ^{241}Am to between 80-90% for ^{237}Np . ^{244}Cm values are not yet available. The validity of this technique will depend upon how close the standards duplicate the samples in size and chemical composition.

Table 1

	^{237}Np	% Trace Recovery $^{239,240}\text{Pu}$	^{241}Am
	81.0	96.7	94.0
	84.0	101.0	93.3
	76.7	94.2	94.3
	83.8	91.3	89.8
	88.1	96.4	98.2
	87.3	99.1	94.3
	90.4	95.3	97.2
\bar{x}	84.5	96.3	94.4
sd	4.6	3.2	2.7

Reagent Blanks

In order to assess the levels of trace transuranics in the chemical reagents being used in the analysis and to measure cross contamination between samples, two reagent blanks are run with each set of 12 samples. The

reagent blanks are started at the point of solubilization of the ashed plant material. Equal quantities of reagents are added and the blanks are analyzed identically as the sample in terms of reagents added and sample mounting and radioassay. It is of interest to note that instrument backgrounds due to electronic noise for our counting systems are less than 0.001 counts per minute. However, the reagent blanks (Table 2) show net backgrounds 136 times higher than the instrument background for ^{237}Np , and 200 times higher for $^{239,240}\text{Pu}$ and ^{241}Am using detector 1. Detector 2 shows net background increases of 69 times higher for ^{237}Np , 100 times higher for $^{239,240}\text{Pu}$ and 270 times higher for ^{241}Am .

Table 2
Reagent Blanks Net c/m

	Detector 1			Detector 2		
	^{237}Np	$^{239,240}\text{Pu}$	^{241}Am	^{237}Np	$^{239,240}\text{Pu}$	^{241}Am
	0.10	0.12	0.47	0.03	0.08	0.06
	0.10	0.10	0.08	0.04	0.11	0.31
	0.20	0.14	0.29	0.03	0.06	0.16
	0.09	0.17	0.37	0.03	0.11	0.13
	0.09	0.11	0.14	0.07	0.09	0.31
	0.18	0.32	0.48	0.22	0.16	0.41
	0.09	0.10	0.08	0.04	0.11	0.31
	0.24	0.56	0.36	0.07	0.06	0.48
\bar{x}	0.136	0.20	0.29	0.07	0.10	0.27
sd	0.06	0.16	0.17	0.06	0.03	0.15

We have not identified the exact source of this trace activity in the reagent blanks. However, it seems evident that this trace alpha activity in the reagent blanks is due in part to residual activity left on the glass ion exchange columns even after acid washings and in part to trace transuranic contamination of the chemical being used in the analysis. World-wide fallout could be the source of this trace activity in the reagents. We have been able to significantly reduce this background activity by discarding the ion exchange resin after each run and by washing the columns in hot 16 M HNO_3 and finally washing them in a .01 M EDTA solution.

We attempted regeneration of the resin beds after each run by washing with 150 ml of concentrated HCl. Trace activity could still be detected in subsequent HCl washings. We concluded that the resin columns had to be changed after each set of samples in order to avoid significant cross contamination between samples.

CONCLUSIONS

Conclusions at this point are that group separations of Np, Pu, Am and Cm from 1-50 gm agricultural samples can be achieved using our technique with chemical yields greater than 80%.

Our sensitivity at present, is limited by the high count rates of the reagent blanks. However, studies are continuing to reduce these reagent blank levels to a point where sensitivity levels approaching 0.1 d/m/gram plant will not be unrealistic goals in future analyses.

Developmental work has been completed in analyzing ^{244}Cm in our plant systems. We will include ^{244}Cm in our group separation technique as described above in the very near future.

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