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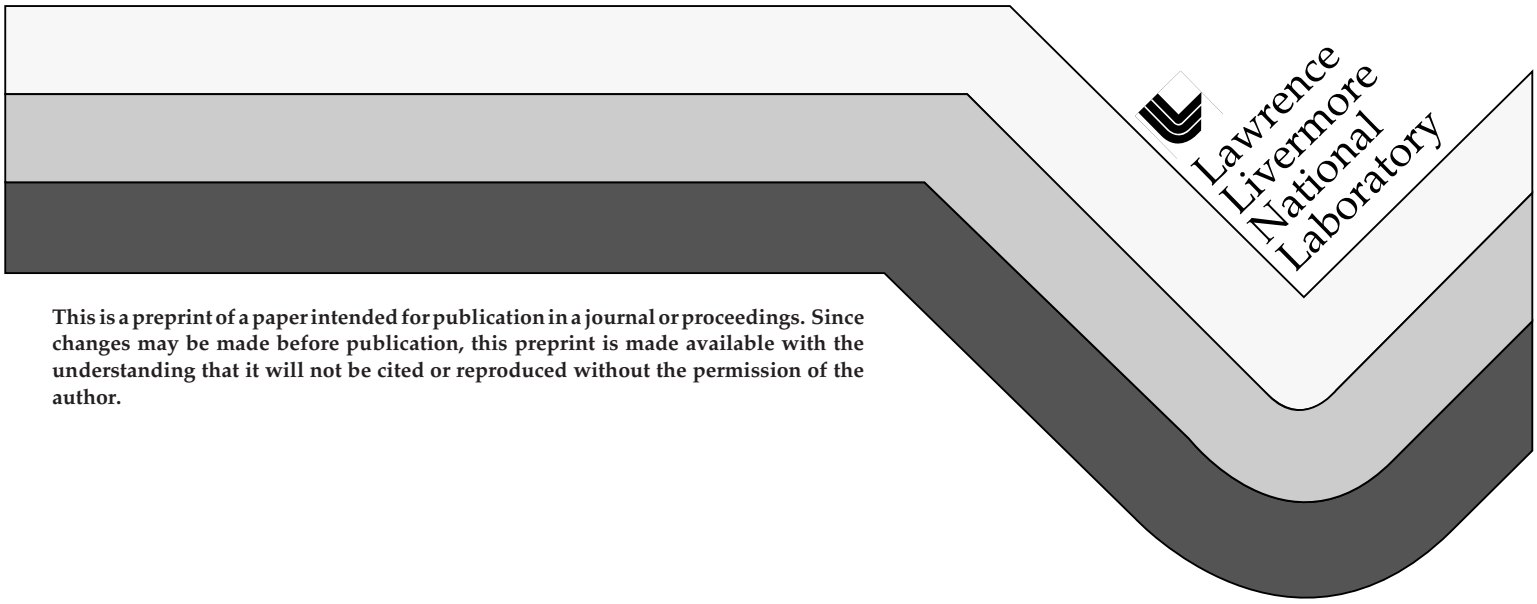

Waste Minimization Measures Associated with the Analysis of ^{137}Cs in Coconut Milk Collected from the Marshall Islands.

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ABSTRACT

The Marshall Islands Environmental Characterization and Dose Assessment Program has recently implemented waste minimization measures to reduce low level radioactive (LLW) and low level mixed LLWMIXED waste streams at the Lawrence Livermore National Laboratory (LLNL). Several thousand environmental samples are collected annually from former US nuclear test sites in the Marshall Islands, and returned to LLNL for processing and radiometric analysis. In the past, we analyzed coconut milk directly by gamma-spectrometry after adding formaldehyde (as preservative) and sealing the fluid in metal cans. This procedure was not only tedious and time consuming but generated storage and waste disposal problems. We have now reduced the number of coconut milk samples required for analysis from 1500 per year to approximately 250, and developed a new analytical procedure which essentially eliminates the associated mixed radioactive waste stream. Coconut milk samples are mixed with a few grams of ammonium-molydophosphate (AMP) which quantitatively scavenges the target radionuclide cesium-137 (^{137}Cs) in an ion-exchange process. The AMP is then separated from the mixture and sealed in a plastic container. The bulk sample material can be disposed of as a non-radioactive non-hazardous waste, and the relatively small amount of AMP conveniently counted by gamma-spectrometry, packaged and stored for future reference.

(key words: ^{137}Cs, Marshall Islands, Coconut Milk, Waste Minimization)

1. INTRODUCTION

Bikini and Enewetak Atolls form part of the Marshall Islands in the north Equatorial Pacific Ocean and were used by the United States for testing nuclear weapons (1946-58). Nuclear weapons detonations occurring within the near surface environment produced close-in radioactive fallout leading to widespread contamination of several major residence islands. Under the auspices of the US Department of Energy, International Health Programs (EH-63), the Health & Ecological Assessment Division at Lawrence Livermore has been evaluating radiological conditions at the atolls since 1974. We provide data and updated dose assessments to assist atoll communities with making informed decisions about

resettlement options. Our long-term strategic mission is to ensure the safe resettlement of the four affected atolls—Bikini, Enewetak, Rongelap, and Utirik.

Marshall Island coral soils make cesium-137 (^{137}Cs) much more available for plant uptake than do soils of North America and Europe (Robison at al. 1997a). The major dietary pathway contributing to the estimated dose for returning residents is through consumption of coconut and other locally grown foodcrops containing elevated levels of ^{137}Cs (Robison et al. 1997b, Robison et al. 1982). As a consequence, our environmental characterization and radionuclide monitoring program in the Marshall Islands is focused on the need to accurately measure the concentration of ^{137}Cs in locally grown terrestrial foods and develop reliable dose estimates. We also use large-scale, experimental field programs to develop and evaluate potential remedial measures to reduce ^{137}Cs soil-to-plant uptake.

Several thousand environmental samples are collected annually from the Marshall Islands and returned to LLNL for processing and radiometric analysis. About 50% of all samples collected consist of coconut meat and associated fluids. Historically, we were asked by our sponsors to maintain a permanent Marshall Islands sample archive for future reference. Typically, coconut meats are dried, homogenized and pressed into metal cans for radiometric analyses using high resolution gamma-spectrometry. These same sample cans are stored as part of our permanent sample archive. In the past, we used a simple dehydration method (slow evaporation) in order to concentrate ^{137}Cs in the coconut fluid. This method was very tedious and involved the use of formaldehyde to help stabilize the sample matrix. Sample cans containing dehydrated coconut milk tend to rupture over time, limiting the useful storage life of the sample and, subsequently, created contamination and waste disposal problems. A LLWMIXED waste stream was eventually established at LLNL to dispose of this waste. Here we report on recent efforts to develop a new preparative method for analyzing ^{137}Cs in coconut milk in order to reduce the associated mixed radioactive waste stream.

2. METHODS AND MATERIALS

Ammonium-molydophosphate (AMP) is a relatively insoluble, microcrystalline powder with a high exchange affinity for Cs^+ ions. It has been previously used in our laboratory to concentrate Cs isotopes from large volume water samples (Wong et al. 1994). During the initial methods development work we performed a series of tracer experiments on several hundred coconut milk samples using ^{134}Cs as a yield determinant (Fig. 1). About 8 kg of AMP (AMP-1 ion-exchange crystals) was purchased from Bio-Rad (Richmond, CA) during a special production run. Gamma-spectrometry measurements were performed on high-resolution Ge detectors coupled to a DEC VAXStation operating under Canberra/Nuclear Data systems data acquisition and reduction software. The propagated measurement uncertainties for ^{137}Cs and ^{134}Cs were normally $< \pm 5\%$.

Tracer experiments using ^{134}Cs

Coconut milk samples consisting of 1-2 L of fluid were allowed to thaw and acidified to pH 2-3 with HNO_3. About 4 g of AMP was stirred into the solution and allowed to settle overnight. The supernatant was carefully siphoned off into a secondary container, and the AMP collected in a 50 mL centrifuge tube that deposits the precipitate in a standardized geometry suitable for direct gamma analysis. The average chemical recovery was 96 ± 3 % ($1\ \sigma$; N=1204). The residual bulk materials were filtered and released as sanitary wastewater in full compliance with Federal and City of Livermore regulations.

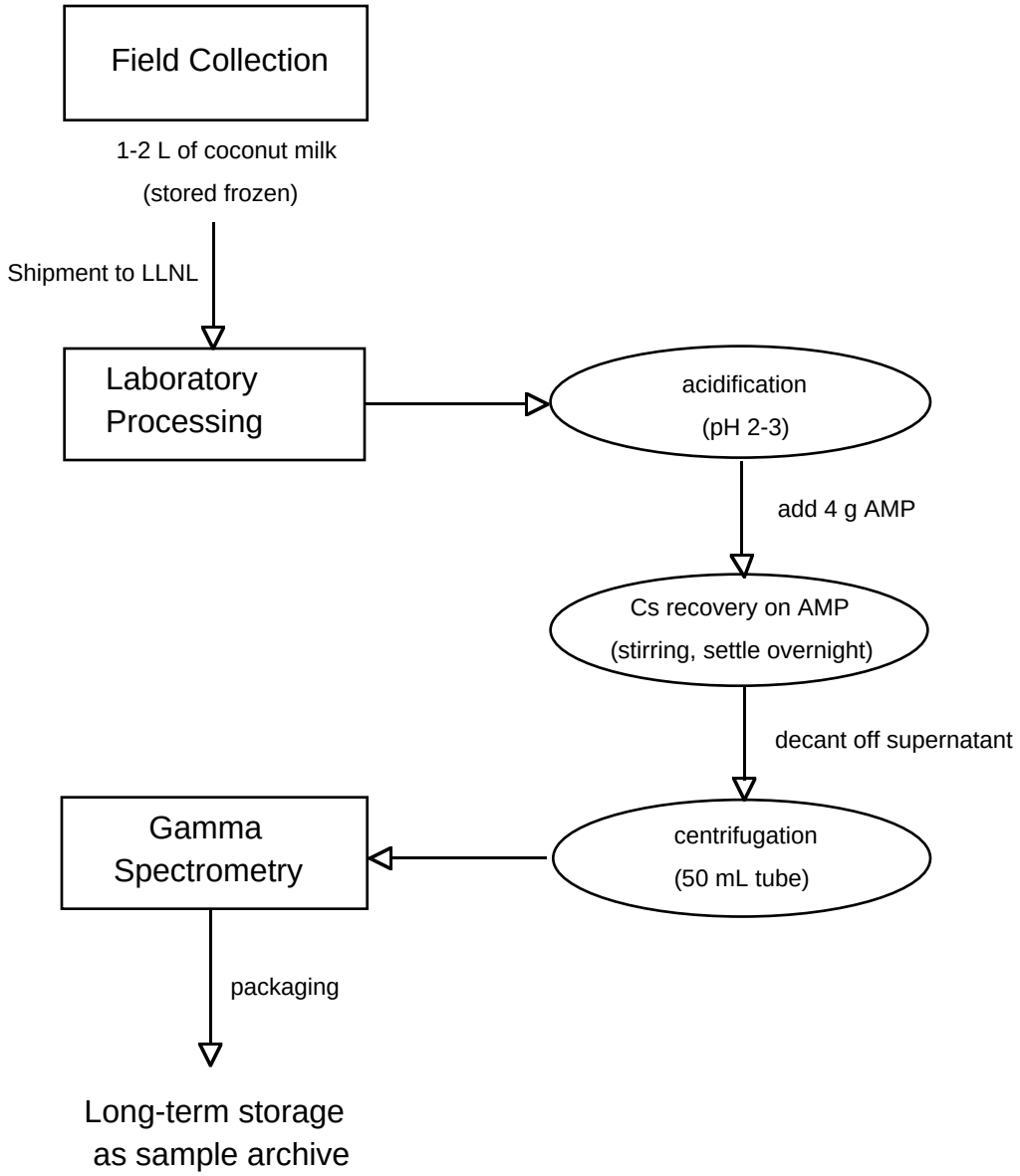

Fig. 1. Outline of steps used for the development of a new radiometric analysis technique for measurement of ^{137}Cs in coconut milk.

3. DISCUSSION

A total quantity of about 1000 liters of LLWMIXED waste has been generated at LLNL in association with the analysis of ^{137}Cs in environmental samples collected from the Marshall Islands. This waste contained about 4×10^6 Bq of ^{137}Cs and up to 8 kg of formaldehyde. Formaldehyde is classified as a federally regulated US RCRA toxic waste and is classified by EPA as a probable human carcinogen under conditions of usually high or prolonged exposure.

We were initially driven to a waste minimization effort because of concerns over breached sample cans within our permanent sample archive. As a result, we decided to re-evaluate our entire process methodology and, if possible, develop a simple and more cost effective method to perform the analyses, reduce the risk of contamination of our laboratory and/or storage facility, minimize the amount of waste generated, and render the sample into a form suitable for long-term storage.

It is well known that AMP is very effective in scavenging cesium ions from large volume sea water samples (Folson and Sreekumaran, 1970). Using 400 mg of powered AMP per liter of sea water, distribution coefficients of about 50,000 are attainable. Similarly, we have found that ^{137}Cs present in coconut milk is very efficiently scavenged onto a few grams of powdered AMP. The small amount of AMP used is transferred into a centrifuge tube and the samples conveniently counted, packaged and stored for future reference. This refined analytical technique for analysis of ^{137}Cs in coconut juice not only lead to the elimination of our mixed radioactive waste stream but greatly improved the general quality of our data and archival integrity of our samples. We are now able to make reasonable estimates of the ^{137}Cs concentration in coconut milk based on the activity ratio in associated meat samples as a function of the normalization weight (Fig. 2). Consequently, we have reduced the total number of coconut milk samples required for analysis under the Marshall Islands Dose Assessment and Radioecology Program from over 1500 samples per year to approximately 250 (Fig. 3).

4. CONCLUSION

Small quantities of ammonium-molydophosphate (AMP) can be successfully used to quantitatively scavenge ^{137}Cs from 1-2 L of coconut milk in an ion-exchange process. The bulk sample material can be disposed of as a non-radioactive non-hazardous waste and the relatively small amount of AMP conveniently counted by gamma-spectrometry, packaged and stored for future reference. We have also found that this technique offers improvements in data quality along with savings in the cost per analysis and use of storage space compared with earlier methods.

Acknowledgments

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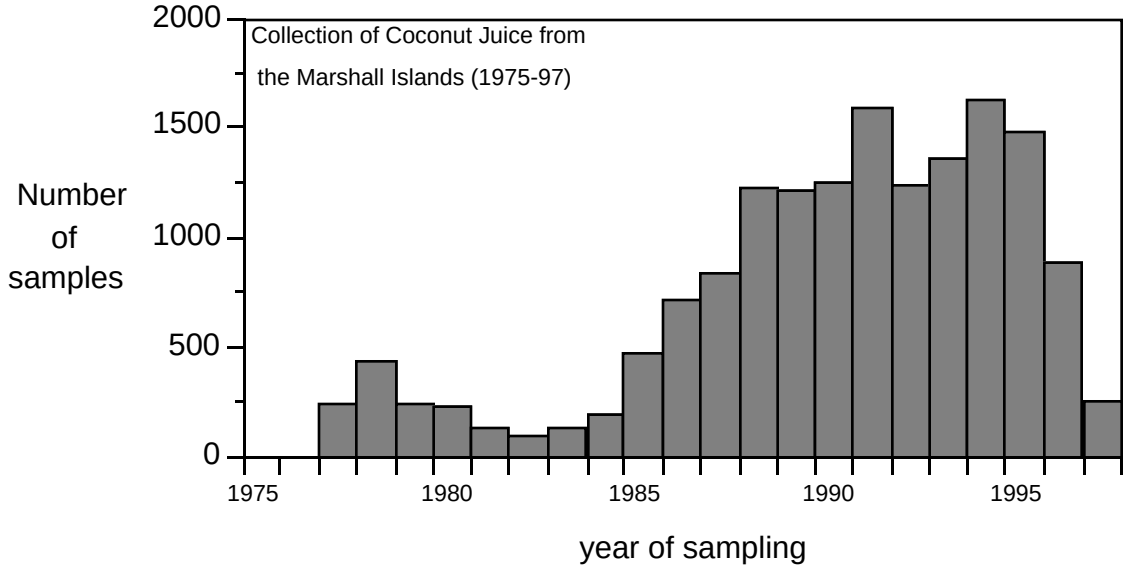

Fig. 2. A plot showing the correlation between the concentration of ^{137}Cs in coconut milk and associated meat (expressed as an activity ratio) versus the normalization weight.

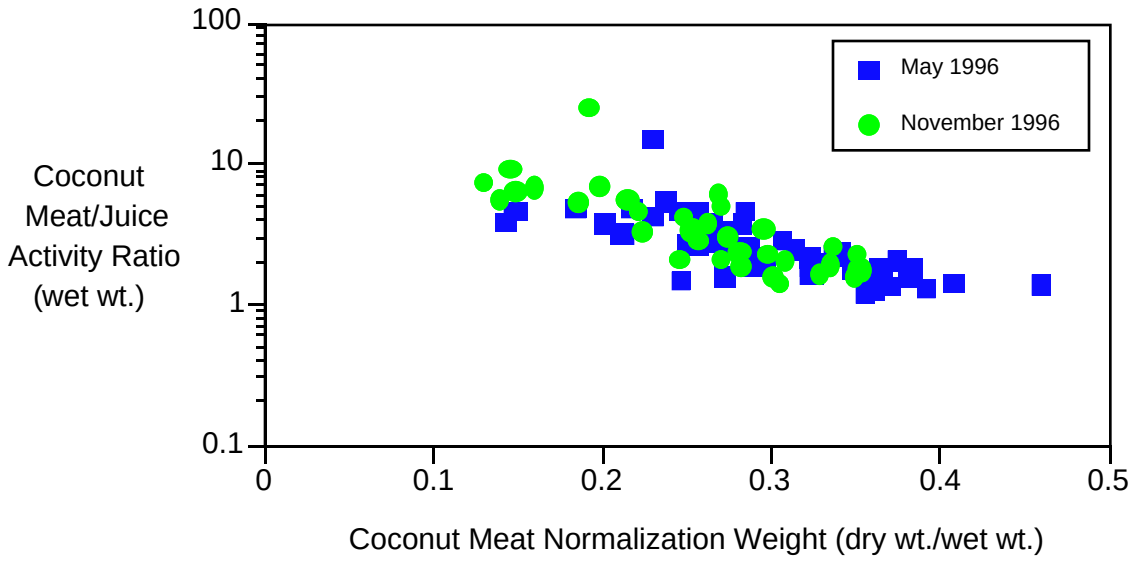

Fig. 3. A plot showing the reduction in the number of coconut milk samples collected from the Marshall Islands (1974-97).

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