Environment International Vol. 3, pp. 395-398
Pergamon Press Ltd. 1980. Printed in Great Britain

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COLLECTION MARKEY FILES
BOX No. 2 OF 6
FOLDER HUMAN IRRADIATION

Development of some Natural Matrix Standards — Progress Report

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Substantial progress has been made in the production of environmental level, natural matrix, radioactivity standards of soil of low organic and low carbonate content, and of human lung and human liver. This is in response to the deliberations of the 1977 ICRM meeting at which the need for such standards was voiced and the criteria suggested. The soil samples have been reduced to optimum particle size and are ready for blending and standardization. A ten year supply of this standard should be ready in a year or less. Reducing the dried lung and liver to less than 74 μm has proven formidable because of the natural fat and oil content. Using cryogenic milling techniques more than three quarters of the total mass of each has been reduced to the desired particle size; the balance will either be reduced or removed and we can then proceed to the final standardization.

Introduction

At the first meeting of the Low-Level Measurement Techniques Group of the International Committee on Radionuclide Meteorology (ICRM), held in Paris in 1976 (Hutchinson and Mann, 1978), several recommendations were adopted. One, specified the development of a program aimed at providing quality control support in laboratories carrying out analyses for radionuclides in the environment and recognized the need for "a supply of natural matrix standards in sufficient quantities and varieties for quality control purposes at the user level" (Hutchinson, 1979). A natural matrix standard was defined as "... a standard of radioactivity which is homogeneously contained in a matrix ... in the same chemical forms that are found in the environment". Bowen (1978) pointed out the necessity for such standards in environmental radiochemistry applications and suggested an assortment of radionuclides, matrices, concentrations and potential sources of supply.

In October 1977, ICRM sponsored a symposium at Woods Hole, Massachusetts, to define more clearly the needs and initiate programs to develop natural matrix radioactivity standards (Hutchinson, 1979). Based upon a number of technical criteria plus the practical workload of cooperating organizations, eight "first round" materials were chosen: human lung and liver, lake and river sediment, lake vegetation and three varieties of soil. Two other important decisions, worth reiterating, were: (a) that the standard should satisfy the needs of the international scientific community for a period of 5-10 yr and (b) that the activity be at the medium concentration levels as defined by Bowen (1978).

This report summarizes the progress to date of the development of three of the standards; the low organic — low carbonate soil, human liver and human lung. The cooperating groups for this soil effort are: Rockwell International's Rocky Flats Plant (RF), the National Bureau of Standards (NBS) of the U.S. Department of Commerce and the Environmental Measurements Laboratory (EML) of the U.S. Department of Energy. The lung and liver standards are being prepared through cooperation of the latter two organizations plus the Los Alamos Scientific Laboratory (LASL).

Rocky Flats soil

Soil from near the Rocky Flats Plant in north-central Colorado was chosen to be one of the natural matrix standards because (a) it fulfills the criteria of low organic-low carbonate content, (b) the contamination concentrations of a number of transuranics are fairly well-known and (c) the logistics and costs of obtaining the samples and shipping then to the EML and NBS laboratories for processing were reasonable.

The Rocky Flats area was contaminated with some transuranic elements some years ago and has been rather thoroughly studied (e.g., Krey and Hardy, 1970; Volchok *et al.*, 1972; Hardy *et al.*, 1978). Figure 1 is an

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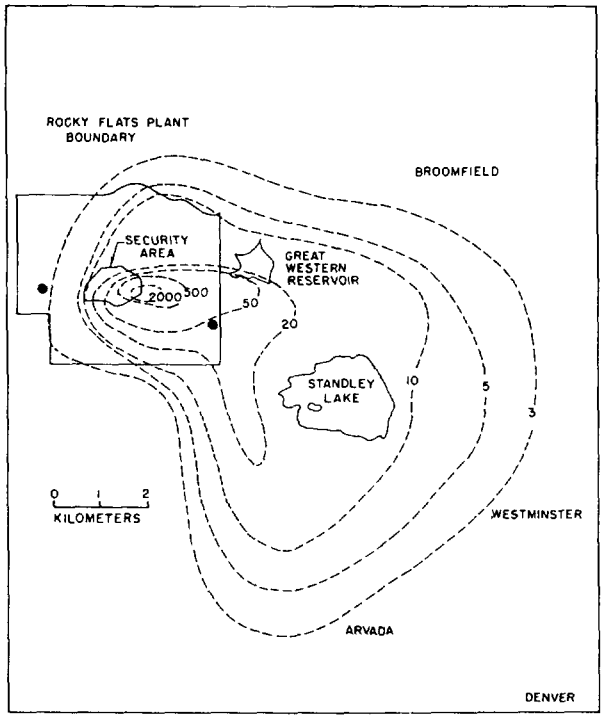

Fig. 1. Map of the Rocky Flats Plant area, Golden, Colorado. The sampling sites are indicated by solid black circles. The isopleths are in units of mCi ^{239}Pu/km^2.

outline map of the Rocky Flats area with superimposed isopleths. Note that dashed lines of ^{239,240}Pu levels are in mCi/km^2. As a matter of convenience throughout this paper ^{239}Pu signifies ^{239}Pu and ^{240}Pu. This is justified because these isotopes of plutonium can not be differentiated by conventional radiochemical techniques.

To satisfy the supply and longevity criteria, we conservatively estimated the sample size as 100 g, the average usage rate as 100 samples per year and the period as 10 yr. This necessitates a final dry homogenized, quantity of 100 kg. The target radioactivity concentration range for ^{239}Pu is 0.05 – 0.1 pCi/g dry soil. In July 1978, three of us (Inn, Feiner and Volchok), with a good deal of help from personnel at the Rocky Flats plant, collected the samples. It was decided to sample two sites, one at or near background level of ^{239}Pu and a second at a somewhat higher concentration than the anticipated final product. Then, after radiochemical analysis of each of the processed samples, an appropriate mixture can be made to achieve the desired concentration. The sites sampled are indicated by solid circles in Fig. 1. Based upon information in Krey and Hardy (1970), the sample from the west side of the plant is expected to be less than 0.02 pCi per g of soil; the east side sample might be as high as 0.2 pCi per g. The samples were coarsely sieved in the field to remove rocks larger than about 1.5 cm diameter. In all, we collected about 600 kg from the east site and about half that much from the west.

After air drying at EML, the samples were milled twice in a Weber Brothers and White stainless steel swing blade mill. The particle size breakdown, after the milling, is shown in Table 1. The relatively small fraction which passed 200 mesh was disappointing as that is one of the limiting criteria of this standard. This criterion is based upon our understanding of the role of particle size in the stability of mixed solid samples (e.g., Bowen and Volchok, 1979). Apparently, mixtures of solids finer than 200 mesh (74 μm) once blended, will not easily segregate under conditions of jarring or vibration and a long shelf life can therefore be expected.

Table 1. Particle size of Rocky Flats soil — after milling at EML (Weight %)

Mesh	West sample	East sample
>20	<1	<1
20–100	58	41
100–200	30	36
<200	12	23

The samples were then taken to the Jet Pulverizer Co. in New Jersey, a commercial firm specializing in the sort of work implied by their name. Upon completion of their pulverizing operation, all of the soil material was substantially smaller than 200 mesh. Table 2 summarizes the mean particle size range for the two soil samples at the present time.

Table 2. Particle size of Rocky Flats soil — after jet pulverizing (Weight %)

μm	West sample	East sample
<4	11	16
4–8	41	39
8–16	45	43
>16	3	2

This is the extent of the progress to date on the low organic-low calcium soil standard. In the next few months each of the soils will be blended to assure homogeneity and then sampled and analyzed for a number of radionuclides including ^{239}Pu. Based on these analyses the "mix" ratio of east and west Rocky Flats soils will be established in order to produce the desired radioactivity concentration in the final product. Appropriate amounts of each will then be thoroughly blended, aliquoted and packaged for homogeneity determination and radioactivity standardization at NBS. These samples will probably be standardized for ^{238}Pu, ^{239}Pu, ^{241}Am and possibly a number of other natural and man-made radio-elements. Our target for making this standard available is before the end of 1980.

Human lung and human liver

The strategy for producing these standards was developed at the Woods Hole Symposium (Hutchinson, 1979) and at subsequent meetings of the cooperating groups. The goal, as with the soil, was to produce a large supply of standardized organ material. Again, we would like to meet the demand for 5-10 yr, supplying standards of about 50 grams each, containing 1-2 pCi per sample. This is a much higher concentration than is found in "background" organs contaminated only by global fallout. Hence, the only practical system for achieving this appears to be to spike a large volume of each tissue type with a radioactively contaminated

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smaller volume of the corresponding tissue. Both the spiked and unspiked tissues, in each case, must be processed to reduce the individual particles to less than 74μm diameter to preclude segregation of the final standardized, homogenized material.

The contaminated organs were obtained through cooperation of the Transuranium Registry Program, Los Alamos Scientific Laboratory. The organs came from two occupationally exposed people who had signed releases stating that their organs could be used for research after their deaths and that the exposure information could similarly be used. Both were killed in automobile accidents. One, a 50-yr old male received all of his Pu exposure in less than a year, at the age of about 20. The other, also male, apparently had a chronic occupational exposure to Pu for the 25 yr prior to his death at age 46. Particle size distribution of the PuO_2 in the lung of the latter individual indicated an activity median diameter of 0.16 μm with a geometric standard deviation of 1.4 (McInroy *et al.*, 1977). The ^{239}Pu concentrations in the livers of the two individuals exceeds 10^3 pCi per kg; the lungs are both more than an order of magnitude higher in ^{239}Pu concentration.

Based on the foregoing, we hold that this proposed procedure will not violate the spirit of the definition(s) of "natural matrix standard" (Bowen, 1978; Hutchinson, 1979; Bowen and Volchok, 1979). It seems reasonable to assume that at the time of death of the two individuals, the plutonium remaining in their lungs and livers, after so many years since the initial contaminating event, must be in physical and chemical forms that can be considered "natural". This is not to say that any or every contamination situation will result in similar metabolic histories; we have no way of knowing that. But we do believe that if and when these standards are completed and made available, they will provide the best approximation to real-life samples at much more easily measurable radioactivity concentrations. To provide 100 samples of each organ per year for ten years with each sample weighing 50 grams, the total mass of standard would have to be 50 kg of dried tissue or 200 – 250 kg of raw material. This is approximately 400 individual whole human lungs and more than 100 livers (HEW, 1979). It was therefore logistically tempting to dilute the contaminated human tissue with more easily obtainable material, such as bovine organs. We were dissuaded from this path by experienced analysts (Boyd, 1979) convinced that there are large enough differences between human and bovine lungs and livers (relating to analytical difficulties) to jeopardize the integrity of the final product. Therefore the decision was taken to use human organs as the diluent.

The general protocol for producing the human lung and liver standards is as follows: (a) complete the collection of the organs, (b) radiation sterilization of the samples for protection of the laboratory personnel, (c) preparation of the samples for freeze drying, (d) freeze drying, (e) pulverization to the appropriate particle size, (f) blending, (g) homogeneity determination, (h) packaging and (i) standardization.

The diluent human lungs and livers were collected through the Transuranic Registry and from local sources in the Maryland-Washington, D.C. area. Only "healthy" specimens, that is, organs unaffected by pathogens were requested. By mid-winter of 1979, 220 kg of liver and 77 kg of lung had been accumulated. Even though the total lung sample was only about one third of that originally planned the difficulty and slowness of accumulating these organs persuaded us to proceed with the processing, recognizing that some sacrifice in either the number of final aliquots or their size would result.

Initial processing of the radioactively contaminated organs was carried out at EML. The material was sliced, reduced in size in a food processor when possible, freeze dried and then further reduced in a "V" type blender with intensifier bar. The particles, at the completion of these preliminary operations, were mostly greater than 700μm in diameter. The samples were then re-frozen and stored, pending further size reduction.

The accumulated background diluent organs were kept frozen until radiation sterilization by a commercial contractor. They received 6 Mrad from a ^{60}Co source. The material was then ground in a meat grinder and freeze dried. The processing to this point had reduced the liver to a dry weight of about 50 kg and the lung to about 16 kg.

Reduction of the lungs and livers to the optimum particle size of 74 μm or less has proven to be more difficult than anticipated. Principally, the presence of fats and oils which constitute some 30% of the dry weight, cause a stickiness and plating out of materials on the walls as the particle size is reduced. Cryogenic milling, therefore seemed most likely to succeed. In a test run, a stainless steel shatterbox mill at NBS was used quite successfully to reduce the particle size but was found to be impractical in terms of time and manpower. We then hired a commercial firm to cryogenically hammermill the diluent material. At this point about 75% of each organ has been reduced to particles of less than 74 μm diameter.

This is where we are today on the human lung and human liver standards; it must be said that the text of this paper does not adequately reflect the maze of twists and turns, the trials and errors the numerous disappointments and the occasional successes which brought us here. At least one more processing step is required to either separate the acceptable (less than 74 μm material) from the coarser fraction, or to reduce the coarse material to the desired size range. We prefer the latter alternative because the former will cost us some 20% of the sample and this we can hardly afford for the lung. Following the successful resolution of the sizing problem, the contaminated organs will also be reduced to less than 74 μm. Then appropriate amounts of the active and diluent organs will be thoroughly blended to achieve the desired radioactivity levels. After assurance of homogeneity, aliquots will be bottled and the final radioactivity standardization at NBS will take place. It is presently anticipated that these organs will be standardized for ^{238}Pu, ^{239}Pu, ^{241}Am and possibly for a number of naturally occurring radionuclides. We are confident that these standards will also be available in 1980.

Acknowledgements — We are indebted to V.T. Bowen (WHOI) and J.H. Harley (EML) for the guidance and advice in the early stages of this effort, to R.E. Yoder, M.V. Werkema and C.T. Illsley (RF) for

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cooperation in obtaining the soil samples and we particularly wish to acknowledge J.M.R. Hutchinson (NBS) for the continued support and encouragement and for his total immersion in the organ pretreatment stages.

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