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TITLE: SAMPLING OF SOILS FOR RADIOACTIVITY: PHILOSOPHY, EXPERIENCE, AND RESULTS

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SAMPLING OF SOILS FOR RADIOACTIVITY: PHILOSOPHY, EXPERIENCE, AND RESULTS

by

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and Edward H. Essington*

INTRODUCTION

The importance of Surface-Atmosphere interactions is emphasized by the title of this conference; a surface source term for surface constituents is of prime importance to an understanding of those interactions. I will discuss the philosophy and development of methods used in the Nevada Applied Ecology Group (NAEG) program to obtain reliable data for the calculation and description of that source term.

The present discussion will be limited to the NAEG experience as it relates to the current emphasis on "surface" plutonium. However, it should be noted that a similar approach may be applied to a study of any surface entity.

HISTORICAL

Fisher at Rothemstead was among the early workers to approach sampling techniques in a scientific manner. His philosophy set a useful pattern for solution of general agricultural problems and served as a basis for many scientific studies

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until the advent of the atomic age. Early methods yielded samples which were satisfactory for the more or less homogeneous nature of constituents and the gross concentrations present. Conservation augers, coring tools, and even shovels were used with some degree of success.

However, the nonhomogeneity of many radionuclides distributed in the environment, their extremely low concentrations, and the sophisticated analytical procedures available today, have changed the approaches to both sampling and sample preparation. Experience over the past eight years in the development of methods and their use in solving the newer problems has indicated the need for more care in determining the mission and a more demanding approach to sampling techniques.

DEFINITION OF MISSION

The worth of the results is directly related to the preliminary consideration and thoroughness with which the mission is defined. If the questions of "why, what, where, when, and how" cannot be answered in an acceptable manner, it is probable that the mission should be redefined.

The first viewgraph presents three of the especially important items which must be considered in defining the mission. The three interrelated factors are: (1) Choice of analytical method, (2) choice of sampling method, and (3) choice of sample preparation method. The mission as noted may be defined by any group of persons with knowledge of the problem. However, the present legal climate would dictate group qualifications which are above argument.

It should be noted that the intent here is to concentrate on the NAEG problem. However, a similar approach is applicable to surveillance, incident, accident, or basic research problems.

Input for mission definition and planning must be supplied by those who have the expertise necessary to answer the posed questions; such expertise should come from; (1) the producer of the material in question, (2) a person familiar with the matrix for analyses, (3) one who can properly prepare the matrix for analyses, (4) the analyst, and (5) a statistician who guides the design, the sampling, the reduction of data and who can remain cognizant of developments as they relate to the direction and degree of success of the mission.

It should be obvious that the above approach may be overly ambitious for some sampling programs. If so, the mission should be redefined to include a less sophisticated design or approach.

The interaction of all persons concerned is a must. The sampler must know how, where and what to sample. The preparation laboratory must know what the analyst requires. The analyst must be able to assay a sample size and configuration dictated by the mission. All of the above must relate to the nature of the matrix and all must work within the constraints of the mission. The statistician's consideration of the data will indicate any changes necessary so that the maximum amount of useful information can be obtained with a minimum of cost and effort. Some guidance as to an acceptable degree of error is mandatory.

SPECIAL PROBLEMS

It is important to note that in the real world at least two major sources of radioactive material in the environment are encountered. These are (1) worldwide fallout related, and (2) incident or accident related.

In the first case, time and meteorological factors have resulted in a relatively homogeneous population of particle sizes which fall earthward at a relatively constant rate and which are relatively uniformly distributed as determined by local meteorology.

In the second case a heterogeneous population of particle sizes may be expelled and distributed locally in a nonuniform manner.

In either case, the approach to sampling, sample preparation, sample analyses, and interpretation of results present unique problems which require their own special treatment.

Of high priority is the requirement for a representative sample free of cross-contamination. Sampling and associated analyses are expensive, cross-contamination generates data which are misleading, and actions based on such information may be costly. Special techniques are required such as decontamination of equipment, proper labeling and documentation, double bagging, prevention of surface disturbances, and stratification of the area of interest based on levels of radioactivity to be expected. Proper licensing and shipping requirements should be observed. We would be remiss if we permitted a variable to exist which could have been controlled.

APPROACH TO MINIFY PROBLEMS

Controllable Variables

In any sampling, analysis, and interpretation study, a series of variables will appear. Some can be controlled; some cannot be. The next viewgraph presents a list of controllable variables. These are (1) cross-contamination, (2) sample activity level, (3) alternate sampling points or sampling methods, (4) matrix particle size definition, (5) sampling stratification, and (6) subsample size. In each case listed, the variable can be controlled by the approach taken at some step or steps; however, changes should be in accord with the defined mission. Two examples from the list are discussed, (1) cross-contamination and (2) subsample size.

The Palomares incident of 1967 and the current survey of the Nevada Test Site (NTS) presented common problems with common goals; these were (1) determine the areal levels, and (2) determine the vertical distribution of radioactivity. The data obtained provide the source term for the final calculation of inventory. Experience at Palomares served as a base from which the present NTS sampling procedure was developed. Prevention of cross-contamination received primary consideration as the first controllable variable.

The next series of slides briefly illustrates the sequence followed for obtaining surface samples and profile samples at NTS. A preliminary survey on an established grid using a FIDLER or comparable survey instrument defined stratification. Selections from a table of random numbers defined the sampling points within

a stratum. An arrangement for survey of the specific sampling point is shown in the next slide. The FIDLER has been placed on a support ring; the ring is 12.5 cm in diameter and for surface samples is 5 cm deep, thus defining the area and volume to be sampled; the radioactivity at the ^{241}Am 60 keV peak* is documented.

In the next slide the ring has been pressed into the soil to the 5 cm depth for actual removal of the confined volume. Disposable plastic spoons are used for the removal of soil. Profile samples may be obtained using the ring technique in which case soil exterior to the ring is removed to the designated depth and the ring again pressed into the soil so as to define the volume of the second or subsequent cuts. Excess soil is replaced and the sampling point appropriately marked to avoid future sampling of the same point.

A preferential method for obtaining soil profile samples is shown in the next slide. A 10 x 10 x 2.5 cm stainless steel scoop is used. In this method, a trench is dug to the desired depth; the exposed front of the trench is faced vertically using a spatula and a side to side motion; scrapings are removed from the bottom of the trench. The scoop is inverted and pressed into the surface; soil is cleared from the area around the scoop to the proper depth. The scoop is removed, placed in the upright position, pressed into the exposed block as shown in the next slide and removed with an upward motion; samples are bagged as above. The process is continued to the desired profile depth.

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* The observed value is used to estimate the plutonium concentration using a factor of 10.

A supply of scoops is desirable so that sampling may proceed while equipment is being decontaminated. Field weight and oven dry weight (110°C for twenty-four hours) are determined.

Results obtained from an initial study of the procedure conducted at Area 13 NTS are shown in the next slide. A standard base is not available in such an approach, but since every effort was made to avoid cross-contamination, we believe that the low activity found in the lower profile reflects some other phenomena such as "solubility" and subsequent transport.

The second controllable variable to be discussed relates to choice of subsample size. As noted above, some guidance to activity levels is provided by the FIDLER stratification survey and by the FIDLER data obtained at the specific sample site. However, a further refinement is included in that each bagged sample is screened for activity on a Ge(Li) detector system at the preparation laboratory, again using the ^{241}Am 60 keV peak as an indicator of approximate plutonium concentration.

From data obtained above, the analyst now has a basis for judgment relative to subsample size. If the mission permits, the mass of matrix to be subjected to analyses can be adjusted so that the radioactivity in the majority of samples will fall within a workable narrow range. The analytical procedure can now be adjusted to accommodate the range expected and since samples in general will contain about the same level of activity, problems of gross cross-contamination are reduced.

Variables Requiring Further Study

Certain variables which are not amenable to control are shown in Viewgraph #3; Sample Size with Respect to Particle Distribution will be discussed because of its marked effect on results from replicates.

The problems encountered in sampling for worldwide fallout are less severe due to the more nearly homogeneous size and distribution of particles. Increasing the number of samples will solve a part of the problem; however, guidance from the statistician on cost-benefit analysis and from definition of the mission will be desirable.

Local type fallout such as from an explosion or fire presents problems which are more complex. A variation encountered among samples or among aliquots from the same sample of up to five orders of magnitude is not uncommon. The proper definition of the mission will have recognized this. An increased number of samples will be of some value - again at an increased cost. The approach taken by NAEG has been to conduct a preliminary sampling, judge the extent of the problem and obtain guidance from the statistician.

The distribution and particle size associated with the incident-accident case is not known; however, its extent is indicated in the next slide. This lack of knowledge presents problems

related to sampling. For example, a single sphere of plutonium oxide 44 μm in diameter produces about 87,000 dis/min; this is about the same activity to be found in 4.3 million grams of soil at worldwide fallout levels. If the assumption is made that there is one such particle and one sampling for each 1,000 square feet, the chances of always obtaining that particle in the sample taken are ridiculously small.

Whether the particle is or is not present in the sample presents problems relative to interpretation of data which have not been solved; this uncontrollable variable may present a major reason for disagreement among certain groups relative to levels of activity associated with certain unfortunate incidents. (We do not have a solution but we will welcome suggestions.)

APPLICATION OF METHODS

The importance of a defined mission has been discussed and emphasized; Dr. Gilbert has reviewed the need for a designed and defined approach. The importance of sample preparation methods should be stressed. Consultation with the analyst is important; the sample presented to him must be of a size, configuration, and radioactivity level compatible with the analytical method to be used.

Historically samples are normally ground, weighed and analyzed; however, where radioactivity exists, the air cleaning and

decontamination of equipment necessary to assure discretely representative matrices is prohibitive. The NTS project has developed a ball milling procedure which uses a one gallon paint can fitted with a compression lid and protected by a brass sleeve.

Each sample is milled and stored in its own container, thus eliminating decontamination and air cleaning problems. The sample may or may not be sieved prior to aliquoting. If it is sieved, both the "less than" and "greater than" material should be weighed to permit a back calculation to the field condition. Although data are available which indicate a very small percentage of activity associated with the "greater than" fraction when mesh sizes of 50 to 100 are used for sieving, some number of those fractions should be analyzed as part of Quality Assurance.

Quality assurance is a must and, although it should be relegated to a discussion of analyses, it is mentioned here to emphasize the importance of accommodating Quality Assurance at the time of sampling and at each subsequent step. A related topic is that of method and/or laboratory certification. Results from analysis of standards are reviewed to indicate laboratory capability or problem areas prior to analysis of the "real life" samples.

RESULTS

When I was a student in organic chemistry we had a running argument with our learned professor, Henry Gilman, to the effect

that there were two types of organic synthesis (1) "paper synthesis" and (2) "laboratory synthesis". We have been discussing "paper" soil sampling; for the remainder of the time we will present data related to the application of the philosophy.

The Soils Element of the NAEG developed, in part, from existing techniques a "standard" analytical procedure for the determination of plutonium in soils. Although all contract laboratories involved do not use the procedure as written, a comparison of results is interesting. Comparisons are made for "within" laboratories and "among" laboratories; data relate to "standard" samples and "field" samples.

The next slide shows results obtained from an interlaboratory comparison of a field soil sample.

The next slide shows results obtained (by participating laboratories) on aliquots from a common sample.

The next slide shows results obtained by participating laboratories on prepared quality control meat samples.

The next slide compares results obtained from the FIDLER Survey, the laboratory Ge(Li) scans and wet chemistry analyses.

The next slide shows the relative importance of profile samples in the calculation of total inventory.

SUMMARY

For the NAEG about 2400 analyses have been performed representing about 1800 soil samples; the information serves as a basic source term for the other Elements of the NAEG such as

Resuspension, Field Ecology, Biological Studies, and the final Modeling as all of these relate to transfer of plutonium to man.

We have presented an approach to sampling which has been developed over the past eight years and with which we have had a fair degree of success. The approach is not presented as the final word; that certain problems exist has been pointed out. However, we do believe that the organization proposed, tested, and in use can provide proper answers to questions which may be posed by the public, the legal profession, and those of you who are interested in Surface-Atmosphere interactions. Dr. Anspaugh will discuss this aspect more completely in his paper.