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THE LRL PARTICLE ANALYSIS PROGRAM

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ABSTRACT

A program has been established for the purpose of determining the distribution of individual radionuclides in particulate radioactive material. Particulate samples collected at their point of origin and selected on the basis of the degree to which they represent the true particle population are separated into groups and analyzed on a totality basis. The overall analytical program is described and an example is given of the initial information produced in a particle group separation performed on a filter sample from the SEDAN event.

THE LRL PARTICLE ANALYSIS PROGRAM

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INTRODUCTION

The Particle Analysis Program is concerned with characterizing particulate radioactive material as a function of sedimentation rate. Particulate samples collected at their source are separated in the laboratory into fractions corresponding to discrete sedimentation rate intervals. Each fraction is then subjected to chemical, physical, and radioisotopic analysis in order to provide a complete inventory, according to particle class, of each radionuclide studied. In addition, chemical composition, size distribution, density, and crystallographic character of the particles are determined for each fraction.

The data provided by this program are directly applicable to three main problem areas. Fallout, Particle Formation, and Biological Availability.

FALLOUT: A necessary input to meteorological models of the fall-out process is the distribution of the particulate material in terms of sedimentation rate - what fraction can be treated as a stable aerosol and what fractions will be deposited by sedimentation at various rates. Total beta radiation, total gamma radiation, total mass and individual radionuclides may each be distributed differently in terms of sedimentation rate. The analysis program is designed to provide information from which the various distribution functions can be determined. If the samples from which the distribution functions are determined are representative of the true particle population, then the distribution functions can be used to deduce the absolute amount of individual radionuclides introduced into the various atmospheric reservoirs.

PARTICLE FORMATION: The size, composition, and radionuclide distributions of particles produced by a specific nuclear event will depend upon the energy output of the event, the mass and chemical composition of vaporized material, and on the environment in which the event occurred. Inter-comparison of the distribution functions obtained from a series of events occurring in similar environments can in principle be used to arrive at relationships between parameters of the distribution functions and those of the energy, mass, and composition of the nuclear event. Such relationships would permit the prediction of particle characteristics for specific combinations of energy, mass and composition. The manner in which the distribution of individual radionuclides varies from element to element for a particular event and from event to event as condensation conditions change should provide a valuable insight into the thermodynamics of the condensation process.

BIOLOGICAL AVAILABILITY: In general, each radionuclide will be distributed uniquely among the particle classes, with the volatile elements tending to be concentrated in the finer particles, and the more refractory elements concentrating in the coarser particles. Biological availability of a particular radionuclide may be assessed if both its distribution among the particle groups and the chemical composition of the particles is known. The data provided by the analysis program includes a spectrum of radionuclides ranging from extremely volatile to extremely refractory species.

While the Particle Analysis Program is intended ultimately to include all forms of man-made particulate radioactivity, the initial work has been solely with particulate radioactive debris resulting from nuclear detonations. Two reasons for selecting this problem area are first, that such debris

represents an important fraction of the total burden, and second, that samples suitable for particle analysis are available. These are residual parts of aerial filter papers containing radioactive debris from the initial cloud from a number of nuclear device tests. Radiochemical analysis data on counterparts of the existing filters provide guidance as to the representativeness of the sample. That is, if the most volatile and most refractory radionuclides are present in the correct relative abundance, then the sample may be considered to be representative of the true particle population.

EXPERIMENTAL PROCEDURE

The overall experimental procedure can be described in terms of three main divisions - Sample Processing, Particle Separation, and Analysis.

SAMPLE PROCESSING: The sample as received consists of a few square inches of IPC Type 1478 cellulose fiber filter, in which the particles are entrapped. The experimental objective is to recover the particles from the filter without altering them in any way. The method used is so-called low temperature ashing in which oxygen gas at low pressure is passed through an r.f. field and over the filter sample. Under these conditions a gradual oxidation of the cellulose occurs during which the temperature of the sample is kept below 100°C. Particles released during the oxidation of the cellulose, are electrostatically attracted to the wall of the glass vial in which the ashing is conducted. Recovery is monitored by gamma counting the sample vial before and after ashing, and is found to be essentially one hundred percent. Comparison of pre-and post-ashing gamma spectra indicates no change has occurred. In order to provide for a permanent gamma spectral reference a disc is cut from the original filter

sample and mounted for counting in the same configuration as the final particle samples. The remainder of the filter is ashed in a weighed vial and the ash weight determined.

PARTICLE SEPARATION: The particle separation procedure must provide a series of particle samples in which the particles are divided into groups according to their sedimentation rates. This may be accomplished by sedimentation in a liquid medium. Large particles may be separated into sedimentation rate groups by gravitational settling. Small particles require centrifugation. In practice, particle samples fall into one of two classes, - those in which many milligrams of particulate material are present and the particles are mostly greater than one micron in size, and those in which less than a milligram of material is present and the particles are less than a few microns in size. Clean separations require that the particle volume be kept low relative to the fluid volume (less than 0.5 percent) and that a density gradient be maintained in the sedimentation fluid. For gravitational settling the particle sample is added to the top of a glass column 30 centimeters in length and one inch diameter as shown in Figure 1. The density gradient is maintained by cooling the system from the bottom. The top of the column is sealed and samples are collected at the base of the column. The collecting vessel is changed periodically in order to obtain about 10 equal mass particle fractions. During the changing process, the liquid column is held by 16 mesh wire screen sealed across the bottom of the glass column. Each fraction is filtered through a weighed Millipore filter, weighed, and mounted for counting. Following a sedimentation period of about one day, the column is drained in sections and the particles remaining in each section are treated as individual particle fractions. For centrifugal

separation, centrifuge tubes of the type shown in Figure 2 are used. A density gradient is established by mixing fluids of unequal density. Centrifugation speeds and times are selected to provide ten to twelve particle fractions of approximately equal activity. Again, the column charge is maintained by 16 mesh screen at the base of the centrifuge column. A small aliquot of each particle fraction is used to prepare a microscope slide. The remaining solution is filtered through a weighed .01 micron pore diameter Millipore filter and mounted for counting. A flow diagram of the analysis is shown in Figure 3.

The original ashed filter sample is divided into five portions, one of these is mounted directly on a Millipore filter as a post-ashing control sample, the remaining four are used to prepare sets of particle fraction samples by sedimentation. The number of sedimentation runs per original sample is dictated partly by the variety of subsequent analysis requirements and partly by the need to know how the sedimentation rate distribution varies with the density of the sedimentation fluid. Briefly, the particle separation experimental procedure is as follows:

- (1) The weighed, ashed particle sample is suspended in a known volume of isobutyl alcohol. The volume used is determined by the requirement that the particle volume should not exceed 0.5% of the isobutyl alcohol volume.
- (2) Three twenty-five percent aliquots of the suspension are taken for sedimentation and mounted filter sample preparation. One five percent aliquot is taken for sedimentation preparation of a set of particle samples suspended in liquid. One five percent aliquot is mounted as a control sample. During the aliquoting process, the suspension is vigorously mixed by a bubbling air through the suspension.

- (3) Suitable sedimentation fluids are those which are moderately viscose, whose densities exceed that of isobutyl alcohol but are less than two gms per cubic centimeter, and which have no solvent effect on the particles. N-butyl alcohol, chlorobenzene, and carbon tetrachloride have been used.

ANALYSIS: The four sets of particle samples produced by the sedimentation are required for four separate analytical programs.

SET I: is reserved for permanent reference and is used for non-destructive analysis - principally gamma spectrometry and x-ray fluorescence.

SET II: is committed to radiochemical analysis for those isotopic species whose half-life and abundance combine to make them of interest but which cannot be determined by instrumental analysis.

SET III: is committed to neutron activation analysis. First to short term activation and gamma spectral analysis; ultimately to long term activation and chemical separation.

SET IV: which consists of particle samples suspended in an organic medium is allocated for optical and electron microscopic examination and to x-ray diffraction and electron microprobe analysis.

Existing LRL analytical facilities are used wherever possible. Analyses such as those in Set I and III which constitute a continuing heavy workload are accomplished by analysis facilities provided especially for particle work.

PROGRAM STATUS

Since its inception in April of this year, the Particle Analysis Program has been engaged in procedural development in connection with the

particle separation problem and with equipment procurement in connection with the analysis problem. The ashing and sedimentation procedures described above were developed and tested using aerial filter samples collected at other than the source. The first application of these procedures to a prime sample was in October. A portion of an aerial filter sample taken at 0 + 8 minutes in the particulate cloud resulting from the SEDAN event was ashed and separated into sedimentation rate fractions using gravitational sedimentation in normal butyl alcohol. The distribution of mass and gross gamma activity among the particle fractions is given in Table 1. These data in themselves are insufficient to permit us to calculate the atmospheric sedimentation rate of the particle groups and are presented only because they may be of some general interest. The additional information required will derive from sedimentation runs in media of different density than the normal butyl alcohol and on particle size measurements on the individual fractions. Although these analyses are in process, the data from them is not available at the time of this writing.

Further analysis of the SEDAN particle samples to obtain distribution data on individual isotopes must await the installation and calibration of low background, high resolution, gamma-gamma and beta-gamma coincidence counting systems the final portions of which are in the process of arriving at the laboratory. It is anticipated that by the time the counting systems are operational, it will be possible to maintain a steady input of particle fractions prepared from filter samples covering a variety of events from previous years. We believe that a substantial amount of useful data will be produced.

TABLE 1

Particle Sample	Distance Fallen* (cm)	Time Removed (min)**	Weight (mg)	Cumulative Weight	Cumulative Weight Fraction	Gamma Activity (cpm)	Cumulative Activity	Cumulative Activity Fraction
1	31.7	16.5	13.46	13.46	.208	1736	1736	.196
2	31.7	31.5	10.98	24.44	.378	1386	3122	.353
3	31.7	46.9	8.57	33.01	.510	1105	4227	.478
4	31.7	61.2	5.23	38.24	.591	763	4990	.565
5	31.7	91.5	6.73	44.97	.695	888	5878	.665
6	31.7	125	5.23	50.20	.776	804	6682	.756
7	31.7	191	4.48	54.68	.846	710	7392	.837
8	31.7	252	1.91	56.59	.875	324	7716	.873
9	31.7	310	0.81	57.40	.888	174	7890	.893
10	31.7	436	1.51	58.91	.911	259	8149	.922
11	31.7	1339	2.21	61.12	.945	367	8516	.964
12	15.2	1339	1.50	62.62	.968	149	8665	.981
13	4.8	1344	1.08	63.70	.985	95	8760	.992
14	4.8	1349	0.97	64.67	1.000	75	8835	1.000

* Particles introduced onto 30.7 cm column of n-butyl alcohol in 2 cm layer of iso-butyl alcohol. Range in distance traveled is 30.7 to 32.7 cm, "Distance Fallen" values correspond to midpoint of range.

** Sedimentation rate calculations should include column temperature gradient:
Base 4.5°C, 8 cm 5.6°C, 16 cm 11°C, 32 cm 20°C.

FIG. 1

GRAVITATIONAL SEDIMENTATION COLUMN

FIG. 2

CENTRIFUGAL SEDIMENTATION TUBE

FIG. 3

ANALYSIS FLOW DIAGRAM