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July 1, 1975—September 30, 1976

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AEROSOL SAMPLING AND CHARACTERIZATION FOR HAZARD EVALUATION

July 1, 1975 to September 30, 1976

by

**Ronald C. Scripsick, Douglas C. Gray, Marvin I. Tillery,
Ronald G. Stafford, and Pablo O. Romero**

ABSTRACT

A draft Manual of Recommended Practice for Aerosol Sampling and Evaluation was completed and sent to the Energy Research and Development Administration (ERDA) Division of Safety, Standards, and Compliance (DSSC) for review. The results of the Survey of Sampling Techniques for Defining Respirable Concentration and/or Particle Size Characteristics of Aerosols were published as LA-6087. The need for greater standardization of ERDA aerosol sampling techniques was indicated. The Aerosol Training Course was presented in 11 sessions to 85 persons. General elements of good practice were emphasized, and recommendation of specific sampling devices or procedures was avoided.

A system for estimating dissolution rates of plutonium aerosols was developed. Studies indicate that plutonium aerosols found in the field have a rapid initial dissolution phase followed by a slower secondary phase. Three methods of particle sizing air samples collected on membrane filters were investigated. The most promising was a scanning electron microscope electron microprobe (SEM-EMP) method. An operating plutonium handling facility was a model for development of techniques to evaluate aerosol surveillance systems performance. Airborne contamination records were studied. The physicochemical properties of a plutonium aerosol existing in the facility were investigated in relation to plutonium handling operations. The techniques developed have indicated some areas of the aerosol surveillance system that need improvement.

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I. MANUAL OF RECOMMENDED PRACTICE FOR AEROSOL SAMPLING AND EVALUATION

A draft of this manual was forwarded to the Energy Research and Development Administration (ERDA) Division of Safety, Standards, and Compliance (DSSC) for review. The Table of Contents is shown in Appendix I. Following review by DSSC, a revised draft of this manual will be prepared for review by experts in the aerosol sampling field.

II. SURVEY OF SAMPLING TECHNIQUES FOR DEFINING RESPIRABLE CONCENTRATION AND/OR PARTICLE SIZE CHARACTERISTICS OF AEROSOLS

The results of this survey were published as LA-6087. Fifty-three selected ERDA contractors were mailed a questionnaire to determine the extent to which respirable and particle-size analysis aerosol sampling was performed at each site, to plan visits to contractors having expertise in these techniques, and to determine the need for (and scope of) a proposed aerosol sampling procedures manual. Of the 37 contractors replying to the questionnaire (70% return rate), approximately half performed respirable or particle size sampling, usually on nonradioactive aerosols. The respirable dust concept inherent in assumptions used to set maximum permissible airborne concentrations (MPCs) for radioactive materials was generally ignored. The wide variation in sampling devices used, calibration methods employed, and data handling techniques reported by these ERDA contractors indicate the need for greater standardization of ERDA aerosol sampling techniques.

III. AEROSOL SAMPLING AND EVALUATION TRAINING COURSE

The Aerosol Training Course was presented in 11 sessions to 85 people during FY-1976 and FY-1976A. This course provides attendees a firm background in the following broad fields: air sampling terminology; minimum number of samples to take; total, respirable, and particle-size analysis aerosol sampling methods; and handling aerosol sampling data. General elements of good practice are emphasized, and no attempt is made to recommend specific sampling devices or procedures. The technical level of this course is suitable for senior technicians or for health professionals. Basic sampling considerations are emphasized and developed to arrive at recommendations for good practice. Lectures are presented by personnel with a variety of backgrounds, including operational industrial hygiene and health physics, and aerosol research and development. Attendees' opinions and experiences are incorporated into subsequent courses where appropriate. Appendix II lists organizations which sent one or more people to attend the course in FY-1976. All attendees had responsibilities for operational industrial hygiene, health physics, or aerosol research and development. Table I lists the major topics covered during the course.

Major benefits to ERDA and ERDA contractors from this course are: training of operating personnel with health protection responsibilities; promotion of a common understanding of aerosol behavior and its effect upon sampling techniques; and promotion of common practices for aerosol terminology, minimum sample size determination, and information required in reporting results of aerosol sampling. A new Aerosol Training Course schedule for sessions beginning in

August 1976 and running monthly through August 1977 was prepared and mailed in May 1976 to the following people: ERDA DSSC (distributed in an Environmental Safety and Health Newsletter dated 21 June 1976); past course sessions attendees, participants in the Survey of Sampling Techniques for Defining Respirable Concentration and/or Particle Size Characteristics of Aerosols; and individuals who have telephoned or written requesting information about the Aerosol Training Course or about the Manual of Recommended Practice.

Major changes have been made in several of the course topics to reflect comments and suggestions submitted by the attendees. The most common changes are increases in the quantity and quality of training aids, including slides, demonstration devices, and handouts. Topics which have required major changes in context and organization include: statistical considerations; optical particle analyzers; and radiation counting considerations.

IV. ASSESSMENT OF INHALATION HAZARDS

A. Dissolution Study

1. Introduction. Decisions on the treatment of individuals exposed to plutonium aerosols via the inhalation route must be made promptly after exposure to ensure maximum therapeutic benefit.^{1,2} Information on the transportability of the deposited material across the lung air-blood barrier is necessary in selection of the most beneficial therapy.^{2,3,4} Rapid methods of making reliable estimates of this transportability do not exist.

Dissolution of an aerosol in the fluid lining the air passages of the lung is a necessary precursor for its rapid transportation to blood.^{1,3,4,5} It is the rate at which material is transported to blood that is of major concern, and therefore it is the dissolution rate at nonequilibrium conditions that is an important parameter in describing this transportation. This dissolution rate in the lung is defined by both the chemical and physical form of the deposited material.^{3,6} While lung clearance models based on the chemical and physical form of exposure aerosol exist,^{3,7} in many accidental inhalation exposures the individuals charged with making decisions on therapy have no method of rapidly determining these critical parameters.¹

Techniques are being developed at the Los Alamos Scientific Laboratory (LASL) which take into account the chemical and physical form in describing the dissolution of an aerosol involved in an inhalation exposure. The dissolution description is based on the nonequilibrium dissolution of an air sample in a simulant of blood serum at physiological temperature and pH. The air samples being studied are taken from aerosols existing in a plutonium handling facility that have been collected by conventional methods on membrane filter media.

2. Description of System. The solubility system is a modification of a design developed at the Inhalation Toxicology Research Institute (ITRI).^{2,3,8-10} A gravity flow system (Fig. 1) passes a blood serum simulant past a filter sandwich, consisting of three filters (Fig. 2). The air sample, collected on a 5- μ m pore-sized membrane filter made of mixed esters of cellulose, is placed in the sandwich middle. The outer filters are 0.45- μ m pore-sized membrane filters also made of mixed cellulose esters. These filters are encased in a dissolution chamber. Liquid flow past the filters is adjusted by a valve upstream of the dissolution chamber (Fig. 1). Also upstream from the dissolution chamber are a large simulant reservoir (~5 l) and a small secondary reservoir (~200 ml). The simulant held in the smaller reservoir comes into temperature equilibrium with a

37°C water bath before it enters the dissolution chamber. The gas release valve attached to the small reservoir permits complete filling of the small reservoir by allowing the displaced air to pass to the atmosphere. The LASL version of the system also includes a dissolution chamber by-pass and an in-line pH probe. The dissolution chamber by-pass permits purging of the system with simulant and adjusting to the flow rate before initiating flow through the dissolution chamber. The in-line pH probe permits continuous monitoring of the simulant pH as it enters the dissolution chamber. The simulant pH is maintained at 7.4 ± 0.1 . The simulant flow past the filter sandwich carries plutonium from the sampling filter to the collecting bottle. Only that portion of the sample that can penetrate the 0.45- μm pore-sized membrane filters is transported by the liquid flow to the collecting bottle. Penetration is thought to be limited to material in solution.

The serum simulant used was prepared according to the procedure and composition described by Kanapilly et al.⁹ This simulant contained the salts normally found in blood serum. Of special interest here was the inclusion of sodium citrate. The citrate ion is thought to be an excellent chelating agent at physiological pH and therefore may have an important role in complexing plutonium ions into transportable forms.^{3,11} Amino acids were represented by glycine and l-cysteine. The simulant pH was adjusted to ~ 7.4 just prior to its induction into the system by addition of minute quantities of NaOH and HCl. This pH was stabilized by equilibrating with 5% CO₂ in a N₂ atmosphere contained in a balloon vented into the large reservoir (Fig. 1).

3. Data Analysis and Results. Solubility data obtained from a Lovelace Foundation ITRI experiment on ²³⁹Y-fused clay particle are presented in Fig. 3.¹¹ Fractions of remaining activity dissolved per day are plotted as a function of time. Data such as these indicate a two-phase dissolution with a rapid initial phase followed by a slower secondary phase. We felt it was feasible to investigate plutonium aerosols in this manner to determine if a two-phase dissolution theory was applicable. If the secondary phase can be determined or estimated, it is felt that conclusions can be made rapidly concerning the relative solubility rate of plutonium aerosols.

Samples eluted from the system are collected on planchets. The activity detected on these planchets is plotted as cumulative per cent of initial sample activity eluted vs time. Figure 4 is a typical plot of data obtained thus far. These data do suggest a rapid initial phase; however, they are too preliminary to conclude that a secondary phase has been reached.

As a comparative index of the aerosols studied, the cumulative per cent of initial sample activity eluted during a certain initial time period was determined. The time period was arbitrarily chosen to be 100 min. Three aerosols were studied in this manner. The cumulative per cent of initial sample activity eluted during 100 min, chemical form, and particle size data are presented in Table II.

The samples studied in runs 15 and 16 (Table II) were taken inside a glovebox in which the machining of plutonium metal was taking place. Microprobe analysis showed the samples to be an oxide form of plutonium. The samples were found to be ²³⁹Pu compound by alpha spectrometry. No particle size data were available.

The sample studied in Run 17 was sampled from a primary shipping container for ²³⁹PuO₂. Microprobe analysis showed the sample to be stainless steel particles with surface contamination of an oxide form of plutonium. The plutonium in the sample was found to be the 238 isotope, using alpha spectrometry. The sample's particle size distribution, measured by an electron microscopy method, has a count median diameter (CMD) of 0.39 μm with a geometric standard deviation (σ_g) of 2.8. This particle sizing method is discussed later.

TABLE I
AEROSOL SAMPLING AND EVALUATION

Major Topics	
What to Sample	Total Particulate Samplers
How to Sample	Respirable Samplers
Aerosol Analysis Considerations	Size Analysis Samplers
Design of Aerosol Sampling Systems	Fibrous Dust Sampling
Sampling Program Management	Surface Sampling
Statistical Considerations	Correction Factors in Radiation Counting
Curve Fitting	Airflow Calibrations

Fig. 1.
LASL dissolution system.

Fig. 2.
Dissolution chamber.

Fig. 3.
Y-Fused clay particles dissolution curve demonstrating two-phase dissolution (based on ITRI data).

Fig. 4.
Typical dissolution curve obtained with the LASL dissolution system. Run 16.

Studies of the long-term dissolution of plutonium aerosols (>100 days) are underway. Preliminary analysis of these data suggests that the initial phase studied thus far does give way to a slower secondary dissolution phase. Further analysis of these data, as well as studies of the long-term dissolution of other samples, are needed to better characterize the dissolution of plutonium aerosol and determine the relation between initial and secondary dissolution.

B. Particle Sizing Study

1. Introduction. Information on the fraction of an aerosol that is capable of depositing along the respiratory tract and the distribution of this fraction along the respiratory tract are important in deciding on treatment for individuals potentially exposed to plutonium aerosols. The particle size distribution of such aerosols is a major factor in estimating the fraction capable of depositing along the respiratory tract and the respiratory tract distribution of this fraction. Therefore, knowledge of plutonium aerosol particle size distribution is important in deciding on treatment for individuals potentially exposed to such aerosols.

Decisions on treatment for individuals potentially exposed to plutonium aerosols must be made promptly after exposure to insure maximum therapeutic benefit. Therefore, information on plutonium aerosol particle size must be made available shortly after exposure to be useful in deciding on treatment.

Methods of estimating plutonium aerosol particle size distribution from air samples collected on membrane filter media were under study. The samples used in the study were collected in a plutonium facility on 5- μ m pore-sized membrane filters. Three methods of particle sizing have been considered: (1) autoradiography, (2) transmission electron microscopy, and (3) scanning electron microscopy-electron microprobe. Investigation thus far has been centered on methods of sample preparation and the time needed for particle size distribution estimation.

2. Autoradiography. Air samples were ashed on stainless steel planchets in a low temperature asher for 1 h at a power <50 W. Studies at LASL have indicated that no detectable activity is lost during the ashing at low power. The planchets were then rinsed in an ultrasonic bath of isopropyl ether. Aliquots of 2 ml were placed on microscope slides coated with Kodak NTB Emulsion in a darkroom for photographic work. The slides were developed following various exposure times dictated by the particle size range perceptible by the technique. Plots of exposure time as a function of diameter for various number of tracks per star are given¹² in Fig. 5 for ²³⁹PuO₂ and in Fig. 6 for ²³⁸PuO₂. Using these plots, one can easily determine what exposure times are appropriate to span the particle size range of interest. After the slides were developed, tracks were counted, using an optical microscope, and particle size was calculated, using a relationship introduced by Leary.¹³ Density estimates have been made, based on chemical form information supplied by microprobe analysis. The frequency and particle size data were fit to a lognormal distribution, and count median diameter, geometric standard deviation, and goodness of fit were determined.

Inaccuracies in particle size estimates, using this technique, arise from the assumption that particles are homogeneous spheres. The field samples being investigated are not made up of homogeneous spherical particles. Comparison of cascade impactor data to autoradiographic data is planned in order to investigate the importance of not fulfilling this assumption.

TABLE II
RESULTS OF DISSOLUTION STUDIES

Run	%-Cumulative Eluted Activity During First 100 min	Chemical & Isotopic Form	CMD μm	σ_g
15	0.17	^{239}Pu Oxide	---	---
16	0.37	^{239}Pu Oxide	---	---
17	0.18	^{239}Pu Oxide	0.39 ^a	2.8 ^a

^aDetermined using transmission electron microscopy.

Fig. 5.

Autoradiographic sizing. Plot of particle diameter vs exposure time for $^{239}\text{PuO}_2$.

Fig. 6.

Autoradiographic sizing. Plot of particle diameter vs exposure time for $^{239}\text{PuO}_2$.

Exposure times of days are often needed to determine the smaller component of the particle size distribution (Fig. 5). Analysis with the microscope is tedious and time consuming, often requiring a week for analysis (Table III). For these reasons, the autoradiographic technique does not now meet the need for rapidly determining the particle size distribution of an accidentally inhaled aerosol.

Techniques are being developed to speed up analysis of autoradiographic data. Statistical manipulations of truncated data are being explored. These manipulations will generate a lognormal distribution which is the best linear fit to the available data.¹³ This will permit estimates of the total distribution parameters to be made from the fraction of the distribution containing the larger particles. Microphotographing equipment will reduce the time needed to analyze the autoradiograph by dividing the work among several people.

3. Transmission Electron Microscopy. The time needed to obtain particle size data by existing transmission electron microscopy (TEM) techniques was investigated. Air samples were prepared¹⁴ by first fusing the filters with acetone vapors and then coating them with carbon and chromium. After coating, the filter material was dissolved with acetone. The remaining coat with the adhering sample was placed onto electron microscope grids. Electron micrographs were made and analyzed, using the Zeiss Particle Size Analyzer.

TABLE III
APPROXIMATE TIME REQUIRED FOR PARTICLE SIZING FROM AIR SAMPLES COLLECTED
ON MEMBRANE FILTERS IN HOURS

Procedure	Autoradiography	Transmission Electron Microscopy	Scanning Electron Microscopy Electron Microprobe
Sample preparation	2	2	2
Micrographing	170 ^a	60	4
Analysis	32	2	2
TOTAL	204	64	8

^aExposure time to define ²³⁹PuO₂ particles as small as 0.1 μ m.

This technique is also time consuming (Table III). The micrographing can take as long as a week, depending on the population density of particles on the filter sample.

4. Comparison of the Particle Size Distributions Obtained by Transmission Electron Microscopy and Autoradiography. Two air samples have been analyzed for particle size distribution by both autoradiography and TEM. A portion of each air sample was analyzed by each technique. In general, autoradiography gave a smaller count median diameter than TEM for each air sample (Table IV).

Several possibilities exist to explain this difference. Inherent to autoradiographic sizing are the assumptions that aerosol particles are homogeneous and a 2π geometry exists in detection of α tracks. Electron microprobe analysis of glovebox air samples indicate that aerosol particles found in the field are often not homogeneous. Examination of the detection configuration involved in autoradiographic sizing shows that the perceptible solid angle is $<2\pi$ steradians. Therefore, adoption of these assumptions yields estimates of particle diameter less than actual particle diameter. Also, there is some question as to whether the technique used to transfer particles to the photographic emulsion is biased against larger particles. From the above considerations, it is thought that autoradiographic sizing tends to underestimate actual particle size.

Conversely, it is thought that TEM sizing may overestimate particle size. It is recognized that particles are lost in the transfer technique employed in TEM sizing.¹⁴ The nature of this transfer technique suggests that transfer may be biased against smaller particles. This bias would result in overestimating median particle diameter. It is thought that some combination of the above considerations explains the difference between size estimates. Further investigation is needed to determine the importance of each consideration especially in relation to aerosol physiochemical character.

5. Scanning Electron Microscopy-Electron Microprobe. Particle size data from the transmission electron microscope may be misleading because the electron microscope's insensitivity to chemical form leads to indiscriminate sizing of particles belonging to the aerosol of concern as well as any extraneous aerosols sampled. While autoradiography is sensitive to the radioactive state of the aerosol, it is not sensitive to chemical form (necessary information for predicting the particle size from autoradiography). The scanning electron microscope-electron microprobe (SEM-EMP) is sensitive to the chemical form of the sampled aerosol. This sensitivity can be used to better characterize both the fraction of sampled aerosol of concern and the size distribution of this fraction.

A technique was developed to obtain particle size information using the SEM-EMP. Air samples are ashed on quartz plates in a low-temperature asher for 1 h at a power <50 W. The quartz plates are then thinly coated (<200 Å thick) with carbon. Micrographs and microprobe grids are produced by the SEM-EMP. Figure 7 is a scanning electron micrograph of a portion of a sample prepared in this manner.

Figure 8 is a micrograph of a sample prepared by a high-temperature ashing technique. Samples prepared in this manner agglomerate, making size analysis impossible. Figure 7 is a micrograph of a sample prepared by the low-temperature ashing technique described above. This preparation allows sizing from micrographs.

Figures 8a and 8b show microprobe grids on the field pictured in Fig. 8. Notice that the entire particle contains plutonium (Fig. 8a), whereas only the "hook" portion contains calcium (Fig. 8b). In this manner, microscopic chemical form of a sample can be investigated.

One air sample, prepared according to the low-temperature ashing technique, has been analyzed, using the SEM-EMP. Information on sample chemical form was obtained by microprobe analysis. The sample was found to be made up of stainless steel particles with surface contamination of plutonium oxide compounds. The micrographs of this sample have not yet been analyzed for particle size.

The information on sample chemical form provided by microprobe analysis is important in estimating dose commitment distribution. The chemical composition of the sampled aerosol particles gives an indication of particle density. This density is necessary in estimating distribution of material deposited in the lung from information on measured size distribution. The chemical composition of the sampled aerosol particles is also an important factor in predicting dissolution of deposited material.

The main difference between time requirements for TEM and SEM is micrographing time. Particles are lost¹⁴ in the sample preparation technique used for TEM analysis. Therefore, for a given sample the particle population density of the sample analyzed by TEM is lower than that analyzed by SEM. This results in more transmission electron micrographs being taken than scanning electron micrographs. Each transmission electron micrograph requires longer processing time than scanning electron micrographs because TEMs use conventional roll film whereas scanning electron microscopes use Polaroid film. Because more micrographs are needed and the processing time for each micrograph is greater, the time required for transmission electron micrographing is estimated to be 56 h greater than for scanning electron micrographing (Table III).

The time required for SEM-EMP analysis was estimated to be 8 h (Table III). This time is short enough to be of some use in the event of an accidental inhalation exposure.

C. Characterization of an Aerosol Surveillance System

1. Introduction. Techniques are required to evaluate the adequacy of aerosol surveillance systems so that improvements to such systems can be made. An estimate of how well any aerosol surveillance system is operating in a workplace can be obtained by determining how well the system is meeting the task challenging the system. Techniques are being developed to both characterize the challenge to an aerosol surveillance system and estimate the system's response to this challenge.

TABLE IV
COMPARISON OF PARTICLE SIZES (1000X)
BASED ON INTEGRATION OF SIZE PARAMETERS

Filter Sample	Autography		# Particles Analyzed
	CMV µm	µg	
SSSC-500-V	0.27	2.90	116
1-401-E-1	0.13	1.86	365
	TEH		
SSSC-500-V	0.81	2.15	335
1-401-E-1	0.39	2.82	789

Fig. 7.

Scanning electron micrograph of particles found in an air (1000X) sample. The air sample was ashed according to a low-temperature ashing technique. Ashing in this manner eliminates agglomeration evident in the photographs below and thus permits particle sizing.

Fig. 8.

Scanning electron micrograph of an agglomerated particle (1000X). Agglomeration due to high-temperature ashing of air sample.

Fig. 8a.

Intensity surface distribution of Pu Mβ x-ray of (10 00X). Notice entire particle contains plutonium.

Fig. 8b.

Intensity surface distribution of Ca Kα x-ray (100 0X). Shows that calcium is only found in "hook"-like portion of particle pictured.

Two important factors in describing the challenge to an aerosol surveillance system are release character and the physiochemical character of the release aerosol. To characterize possible release aerosols, a survey of aerosols involved in routine plutonium operations has been initiated. The survey involves air sampling the environment of well-defined plutonium operations. Analysis of the dissolution rate and physical properties of these samples provides information on aerosol character in relation to plutonium operation.

Information on release character is provided by studying airborne contamination records. Compiling of records pertaining to releases from a given location permits characterization of such releases as to their rate, cause, magnitude, etc.

System response is indicated by system ability to detect a release, estimate release magnitude, alarm when proper airborne level exists, and isolate releases in space and time. Information on these system abilities is provided by evaluating the data obtained from the surveillance system during actual release conditions.

Data presented in this section are preliminary. Data collection and analysis development are in progress.

2. Description of the System. The system used as a model in this study is an existing aerosol sampling and monitoring system in a plutonium research and development facility at LASL. The surveillance system is typical of those employed by plutonium handling facilities in the U.S. Because of this, the techniques being developed have applicability to other similar surveillance systems.

The plutonium facility consists of six buildings of which five are serviced by the model surveillance system. Each of these buildings is divided into work areas called rooms. These rooms are walled off and interconnected by doors and hallways. A diagram of this facility is shown in Fig. 9.

Many operations which are performed in this facility, such as grinding, milling, and high-temperature ashing, generate aerosols of plutonium. Releases of plutonium aerosol in this facility, to a great extent, are confined to the room in which the release occurs. In this respect, the surveillance system is several small surveillance systems, each serving a specific room.

Most of the work in this facility is carried out in gloveboxes operated at a negative pressure with respect to the work area, and equipped with flexible butyl-neoprene-type gloves and sealed window spaces to permit safe execution of plutonium operations. Materials involved in these operations are taken in and out of the glovebox through bagout ports and/or airlocks. A majority of the infrequent releases that occur in this facility are a result of glove degradation and the transfer of the materials through the bagout ports and/or airlocks.

The work area about the glovebox lines is ventilated to clear airborne releases quickly. The ventilation system is normally arranged so that the general airflow in each room has a component toward the floor. Typically, ventilation inlets are located near ceilings and exhausts are located near floors. The locations of ventilation inlets and exhausts are shown in Fig. 10.

The aerosol sampling and monitoring system is designed to detect releases of airborne activity in the work area. The fixed head samplers are located in the work area adjacent to the glovebox lines ~2 m above the floor. They are denoted in Fig. 10 by circles with letters next to them. A sample is taken each day from every sampler in the facility. Normally, samples are collected for 24 h. During periods of actual or potential release, special samples are taken in the vicinity of the release. These special sampling periods are called "Special Air Tests." Both the daily and special samples are collected at a flow of between 0.042 and 0.057 m³/min on fibrous filters made of cellulose and asbestos and are analyzed with a Wide Beta II gas flow proportional counter. Daily samples that indicate an airborne concentration ≥ 4.0 (dis/min)/m³ are reported in an

"Above LASL Guide Number Air Count Report." Special samples indicating an airborne concentration ≥ 440 (dis/min)/m³ are reported in a "Special Air Test" report.

Continuous air monitors (CAM) with alarm capabilities are located near the room exhausts of the ventilation system ~1 m above the floor. They are denoted by rectangles on Fig. 10. Both filter samples and monitoring information are provided by these monitors. The filter samples are collected on 5- μ m pore-sized membrane filters made of mixed esters of cellulose at a flow of between 0.028 and 0.042 m³/min. The sampling is continuous with filters being changed every 24 h. Continuous monitoring of the activity buildup on the filter is provided by a silicon diffuse-junction detector. The output of this detector passes through two single-channel pulse height analyzers. One analyzer is set to detect the α energy of ²³⁹Pu and the other to the α energy of the Rd-Th daughters. The outputs of these analyzers are such that the Rd-Th background is subtracted. The alarm of the monitor is activated whenever pulse height analyzer output is greater than a preset count rate. The triggering level is currently ~60 dis/min. The airborne concentration indicated by the gross α activity on the filter sample and the probable cause for the alarm are reported in a "CAM Alarm Report" made out for all alarm occurrences.

3. Glovebox Survey of Aerosols at a Plutonium Facility. Strategies for surveillance of aerosols in the work environment are affected by the aerosol chemical and physical form. For radionuclides, airborne maximum permissible concentrations (MPCa) are in part dependent upon the dissolution properties of the hazard aerosol. This parameter is important in estimating minimum concentration which the surveillance system must be able to detect.

Aerosol physical form also affects this necessary limit of detection. In addition, physical form affects such sampling parameters as filter surface collection efficiency, penetration, and sampler flow rate.

To investigate aerosol character, air samples are taken from inside a glovebox in which a plutonium operation is taking place. Sampling location, sampling conditions, operation description, and chemical and isotopic form of materials involved in the operation are recorded on the form shown in Fig. 11. These samples are then analyzed for dissolution and physical properties, using the techniques described earlier.

Two work locations have been sampled. Analyses of these samples are underway.

4. Retrospective Study of Airborne Contamination Records. Information on release character, such as release frequency, magnitude, and cause, can be obtained by studying the release records of a particular location. This information is important in determining sampling strategies.

The records of potential and actual releases occurring between July and December 1975 have been compiled according to release location. All existing records for a given event were combined. A "contamination incident record" form was filled out for each event. A copy of this form is shown in Fig. 12. In addition, a copy of all pertinent release records was attached to this form. These completed forms were grouped according to release location.

While not all releases occurring in a work area are of equal severity, their mere occurrence does indicate the existence of a potential hazard and the need for monitoring. The relative release frequency at locations within a work area may be used as an indication of the need for monitoring at these locations. Estimates of the relative release frequency at various sampling locations in Building 150 were calculated from the compiled records (Table V). These relative release frequencies are the fraction of the total number of releases occurring in a room that occur near a given sampler location. Instances of potential as well as actual releases were included in the

Fig. 9.

Diagram showing buildings and rooms of a LASL plutonium handling facility being used as a model in the aerosol surveillance system study.

Fig. 10.

Diagram of typical workroom in model plutonium handling facility showing location of air samplers and monitors and room ventilation inlets and exhausts.

Bldg. _____ Date _____
Room _____ GLOVEBOX AEROSOL SAMPLE Time _____
Location _____
SAMPLE NO. _____
Flow rate _____ LPM Sampling duration _____ Min
Filter type _____ Collection on _____ up _____ down
Filter's _____ side _____ side
Filter holder type _____ (as found in package)

Materials being worked with in the sample area:

Activities being carried out in the sample area:

Fig. 11.

Form used in glovebox survey of aerosols.

Bldg. _____ Date _____
Room _____ CONTAMINATION INCIDENT RECORD Time _____
Head _____
How was incident reported? CANAI _____ ALGRATL _____ ALGNACR _____ BOR _____
SAT _____
Fixed head monitor data supplied? yes _____ no _____
Continuous air monitor data supplied? yes _____ no _____
Surface contamination data supplied? yes _____ no _____
Rose swipe data supplied? yes _____ no _____
In vivo data supplied? yes _____ no _____
CANAI - Continuous Air Monitor Alarm Information
ALGRATL - Above LASL Guide Routine Air Test Log
ALGNACR - Above LASL Guide Number Air Count Report
BOR - Radiation Occurrence Report
SAT - Special Air Test
ACT - Airborne Contamination Test

Fig. 12.

Form used in retrospective study of airborne contamination records.

TABLE V
RELATIVE RELEASE FREQUENCIES^a IN THE VICINITY
OF AIR SAMPLERS AND MONITORS IN ROOMS OF BLDG. 150

Release Location	Rooms						
	601	602	603	604	605	606	607
A	---	0.31	---	0.34	0.36	0.23	0.33
B	---	---	0.71	0.07	0.27	0.19	0.11
C	---	0.12	0.21	0.10	0.13	0.04	---
D	---	---	0.07	---	0.06	0.11	0.14
E	---	0.12	---	---	---	0.21	---
F	---	0.13	---	0.38	0.11	0.06	0.44
G	---	---	---	0.10	---	0.02	---
H	---	---	---	---	0.05	0.09	---
Shop	---	---	---	---	---	---	---
N.E. CAM	---	---	---	---	---	---	---
S.E. CAM	---	0.06	---	---	---	0.02	---
Total ^b Releases	0	16	14	29	39	47	9

^aRelative release frequency = (number releases in the vicinity of a given air sampler or monitor during study period)/total number releases in room during study period.

^bReleases from multiple locations are not included in the relative frequency estimate for individual release location. However, these multiple location releases are included in "total # releases." Therefore, release rate estimates for a particular room may not sum to 1.00.

relative release frequency estimation. The total number of releases occurring in each room during the period examined are listed across the bottom of Table V.

5. Evaluation of Special Air Tests. Whenever a recognized release or potential release occurs, the filters of area samplers in the vicinity of the release are replaced with clean filters. These filters remain for the duration of the release. An estimate of the airborne concentration in the vicinity of the sampler is made from the filter sample activity. The purpose of these tests, called special air tests (Sec. IV.C.2), is to detect the location and magnitude of any release that might occur and isolate it in time. As of May 24, 1976, for this study, the filters of the CAMs were made part of these tests. These CAM filters are now changed, along with the area sampler filters during a special air test. Tests of this sort are providing information on the performance of the aerosol sampling and monitoring system during actual or potential release conditions.

For a given release, the resultant airborne concentration estimate from each sampler is divided by the highest concentration estimate made for this release. This quotient, called concentration factor (CF), indicates dispersion of a release in vicinity of a given sampler. These CFs are with respect to a given release location. During a three-month study, five releases occurred in the vicinity of sampler F in Room 606. The CFs at the room air samplers, with respect to release location F, are shown for all five releases in Table VI. For these five releases, every air sampler failed at least once to detect a release (CF = 0), and the maximum sampled airborne concentration estimate was not made twice by any sampler. In general, the maximum sampled airborne concentration estimate does vary from release to release for a given release location, and every sampler studied has failed to detect a release.

To better characterize the sampling variability indicated above, averages of CF over the number of releases occurring at a particular release location during the three-month study period

were made. These CFs are shown along with their range in Table VII. The CFs for CAMs in Table VII demonstrates the variability of CAM coverage as a function of release location. For instance, the CAM coverage for a release at G ($CF_{NE\ CAM} = 0.53$ and $CF_{SE\ CAM} = 0.33$) is better than the coverage for releases at B ($CF_{NE\ CAM} = 0$ and $CF_{SE\ CAM} = 0$), D ($CF_{NE\ CAM} = 0.10$ and $CF_{SE\ CAM} = 0$), and F ($CF_{NE\ CAM} = 0.03$ and $CF_{SE\ CAM} = 0.06$). This indicates that for a given release concentration, a release at G is more likely to trigger a CAM alarm than a release at B, D, and F. From Table V, the area in the vicinity of sampler B in Room 606 has a high release frequency, relative to release locations D, F, and G in Room 606. Since risk of a relatively high air concentration existing without setting off an alarm is directly related to relative release frequency and inversely related to maximum CAM CF, for a given release location, by dividing the release frequency by this CAM CF, a relative estimate of this risk can be made. Table VIII shows such estimates for samplers B, D, F, and G. Because of the poor CAM coverage in the vicinity of B and the relatively high release frequency there, the relative risk of having a release not announced by the CAM alarm is high. These data indicate that a relatively high air concentration at B would probably not be detected by the monitoring system and could persist until the next day, when it would probably be detected by the sampling system. Discovering, via the sampling system, would come too late to prevent exposure.

TABLE VI
CONCENTRATION FACTORS (CF) FOR RELEASES
IN THE VICINITY OF SAMPLER F IN ROOM 606

Sample Location	Releases at F				
	1	2	3	4	5
A	---	---	---	0.02	0.11
B	---	---	---	0.18	0.51
C	---	---	---	0.02	0.25
D	0.50	1.0	---	0.08	0.27
E	---	---	---	1.0	.28
F	---	---	---	0.23	1.00
G	---	---	1.0	0.09	0.19
H	1.0	---	---	0.004	0.12
Office	---	---	---	---	---
N.E. CAM	---	---	---	0.04	0.26
S.E. CAM	---	---	---	0.02	0.11

TABLE VIII
ESTIMATE OF RELATIVE RISK OF NO ALARM FOR RELEASES
NEAR SAMPLERS B, D, F, AND G IN ROOM 606

	Release Location			
	B	D	F	G
Relative Release Frequency	0.19	0.11	0.06	0.02
$CF_{max-cam}$	0	0.10	0.06	0.53
Estimate of Relative Risk of No Alarm (Relative Release Frequency/ CF_{max})	∞	1.10	1.00	0.04

TABLE VII
RANGE AND AVERAGE OF CONCENTRATION FACTORS
(Maximum/Average/Minimum)

Monitoring Locations	Release Locations									N.E. CAM	S.E. CAM
	A	B	C	D	E	F	G	H	Shop		
A	---	1/0.5/0	---	0.55/0.28/0	0.50/0.25/0.0001	0.11/0.03/0	0.35/0.12/0	0.29	---	---	---
B	---	---	---	---	1.0/0.50/0.0001	0.51/0.14/0	---	---	---	---	---
C	---	---	---	---	---	0.25/0.05/0	---	0.18	---	---	---
D	---	---	---	---	0.0001/5 × 10 ⁻³ /0	1.0/0.37/0	---	---	---	---	---
E	---	1/0.61/0.22	---	1/0.50/0	3.50/0.25/0	1.0/0.26/0	0.35/0.12/0	---	---	---	---
F	1.0	---	---	---	---	1.0/0.25/0	---	0.18	---	---	---
G	---	0.22/0.11/0	---	---	---	1.0/0.26/0	1.0/0.53/0	---	---	---	---
H	---	---	---	---	0.0001/5 × 10 ⁻³ /0	1.0/0.22/0	1.0/0.33/0	0.18	---	---	---
Shop	---	---	---	1/0.78/0.55	---	---	1/0.59/0	---	---	---	---
N.E. CAM	---	---	---	---	0.07/0.04/0	0.26/0.06/0	1/0.33/0	0.82	---	---	---
S.E. CAM	1.0	---	---	0.20/0.10/0	1/0.50/0	0.11/0.03/0	1/0.53/0	1.0	---	---	---
# No Release Detected	---	3	---	---	---	---	---	---	---	---	---
# Release Detected	1	2	---	2	2	5	3	1	---	---	---

~2 m above the floor (Fig. 10), releases at the glovebox level would not be expected to be sampled due to the direct airflow. Any part of a release sampled would be expected to be due to unsteady airflow within the room.

Release location is not isolated by high concentration estimates in the vicinity of release. In many instances, the release is not even detected by the sampler nearest release location. Instances of sampling and monitoring systems independently failing to detect an entire release have been discovered. There has been no investigation into the possibility of these systems simultaneously failing to detect a release.

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APPENDIX I

Manual of Recommended Practice for Aerosol Sampling and Evaluation

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APPENDIX II

AEROSOL SAMPLING AND EVALUATION TRAINING COURSE

List of Organizations Sending One or More Attendees to This Course

1. Aerojet Nuclear Company
Idaho Falls, Idaho	10. ERDA Los Alamos Office
Los Alamos, New Mexico
2. Argonne National Laboratory
Idaho Falls, Idaho	11. ERDA Nevada Operations Office
Las Vegas, Nevada
3. Atlantic Richfield Hanford Company
Richland, Washington	12. Flanders Filters, Incorporated
Washington, North Carolina
4. Babcock and Wilcox Company
Lynchburg, Virginia	13. Hudson Health Center, Ohio University
Athens, Ohio
5. Battelle Northwest Laboratory
Richland, Washington	14. Lawrence Berkeley Laboratory
Berkeley, California
6. Colorado State University
Fort Collins, Colorado	15. Lawrence Livermore Laboratory
Livermore, California
7. E. G. and G, Incorporated
Las Vegas, Nevada	16. Lovelace Foundation
Albuquerque, New Mexico
8. E. G. and G., Incorporated
Santa Barbara, California	17. Los Alamos Scientific Laboratory
Los Alamos, New Mexico
9. ERDA Albuquerque Operations Office
Albuquerque, New Mexico	18. Mason and Hanger Company
Amarillo, Texas

19. National Lead Company of Ohio
Cincinnati, Ohio	24. Savannah River Plant
Augusta, Georgia
20. NASA White Sands Test Facility
Las Cruces, New Mexico	25. Texas A and M University
College Station, Texas
21. Nuclear Fuel Services
West Valley, New York	26. Union Carbide Corporation
Paducah, Kentucky
22. Reynolds Electrical and Engineering
Las Vegas, Nevada	27. United Nuclear Industries, Incorporated
Richland, Washington
23. Rocky Flats Plant
Golden, Colorado	28. Zia Company
Los Alamos, New Mexico