

*Health, Safety,  
and Environment  
Division*

*Annual Report 1988*

*Compiled by  
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**MASTER** 

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## A STUDY OF VENTILATION RATES IN PLUTONIUM-HANDLING LABORATORIES

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The purpose of this 2-1/2 year study is to determine the effect of various ventilation rates on airborne contaminant releases in plutonium (Pu)-handling laboratories. At the present time disparate ventilation rates are used in such facilities with little apparent technical justification. Little is known about the effect of changing ventilation rates on health-related parameters such as contaminant clearance rates, time and position of maximum concentration, mixing factors, and turbulence. In addition, only meager guidance is available to designers concerning the optimum type and location of air inlet and exhaust fixtures, effect of glovebox arrangements on air patterns, and so forth.

Specifically, the primary objective of the study is to compare the effectiveness of different work space ventilation air exchange rates through Pu-handling laboratories, based on the following criteria:

1. The health protection (as indicated by air monitoring, exposure, and bioassay data) provided to workers during operations under known ventilation conditions.
2. The clearance time, concentration distribution and increase time at specific locations, spread time, and other spatial characteristics of tracer gases or aerosols released during simulations of radioactive material release.

During the past year (second year of the study), aerosol release experiments were completed at two Los Alamos laboratories and were started at one laboratory at the Rocky Flats Plant (RFP). The experiments and several aspects of the results to date are discussed.

Similar experiments were performed in each of two laboratories at Los Alamos. The first was a mockup of a Pu laboratory in an empty storage room at the former Pu fabrication facility at TA-21, Room 208, (designated DP-208). A ventilation exhaust system was installed to complement the existing ventilation supply system (Fig. 1). The supply system consisted of six diffuser inlets placed asymmetrically with respect to the center of the room, and four rectangular plenum outlets, one at each corner of the room near the floor, exhausted air. Pu glovebox lines were simulated by cardboard structures.

The second laboratory was at the TA-55 Pu facility. Room 208 in PF-4 was used for the experiments (Fig. 2). No modifications were made to this room, which had two ceiling supply air diffusers and one exhaust grill near the floor in each corner. Three open transfer hoods also extracted some air from the laboratory.

**Aerosol Releases.** Aerosol releases in DP-208 were made at three nominal ventilation rates - 16, 8, and 4 air changes/hr (AC/h) - at three room locations, and with three replicates at each location. A similar number were made in TA-55-208, except that only two ventilation rates, 8 and 4 AC/h, were used, and an additional three releases at each of two locations were made with two generators operating simultaneously to provide much higher concentrations. No known thermal sources were operating in either of the laboratories during the experiments, so buoyancy forces should have been minimal. Likewise, the experiments were all performed with the rooms empty and were not started until after a 10-minute calming period.

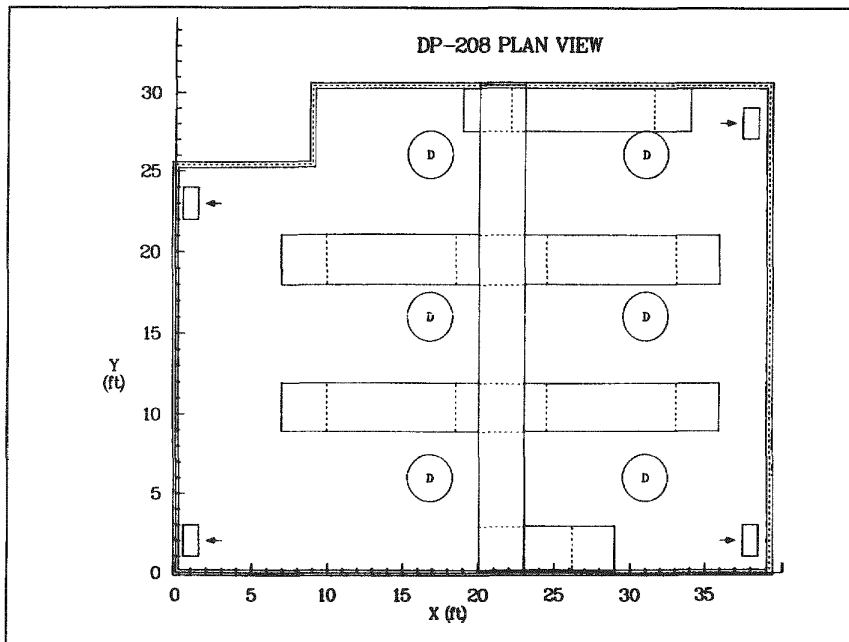


Figure 1. Schematic layout of DP-208 laboratory. Top is north. Rectangles represent glove box lines (E-W) and overhead transfer tunnel (N-S); circles with D are ceiling supply air diffusers; rectangles with arrows are exhaust plenums.

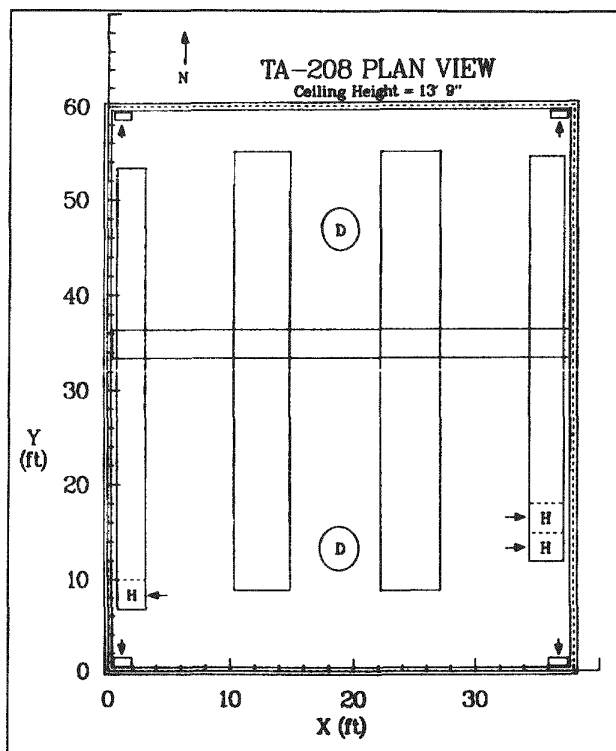


Figure 2. Schematic layout of TA-55-208 laboratory. Same representations as Figure 1, plus squares with H and arrows are open transfer hoods.

The aerosol, sodium chloride doped with a known amount of sodium fluorescein as a tracer, was produced by an air-operated generator at a rate of 200 to 300 mg/min. The sodium chloride aerosol was log-normally distributed, with distribution parameters of 1.0  $\mu\text{m}$  mass median aerodynamic diameter and geometric standard deviation of 2.6, as determined from multiple Andersen impactor samples. The aerosol was sampled, isokinetically where necessary, with membrane filters located at the room exhausts, fume hood faces, and at 72 to 88 diagnostic positions in the room. The diagnostic locations varied with room size and equipment locations, but samplers were always placed on vertical towers at 91, 183, 274, and 366 cm (3, 6, 9, and 12 ft) above the floor at each room location.

After each aerosol release (30-min duration) was complete, the samples were collected and analyzed fluorimetrically for the mass of fluorescein present. In addition to the filter samplers which provided samples of cumulative aerosol, in most of the releases four laser particle counters (LPC) provided real time aerosol number concentration throughout the release. For the final six releases (two generators) in the TA-55 laboratory, an additional four LPCs were used.

**Aerosol release results.** In general, the average concentration increased in proportion to the ventilation decrease (Table I), indicating that the sampling was adequate. In all cases in DP-208, about two-thirds to three-fourths of the sampling locations had concentrations less than the average. The average value for the locations having concentrations greater than average ranged from 1.5 to 4.8 times the average, with the lowest occurring at 8 AC/h. These results from the DP experiments indicate that ventilation effectiveness depends on the criteria used for evaluation. As expected, the average concentration within the room increased with decreasing ventilation rate, but the distribution of concentration with position changed also. The concentration distribution in TA-55-208 showed similar results and trends.

Aerosol distribution for both DP-208 and TA-55-208 has also been depicted in contour graphs such as the four illustrated for TA-55 (Fig. 3). These plots of the average concentrations from all releases at identical conditions help visualize the movement of aerosol in the four sampling planes above the floor. In addition, they provide a visual

comparison between aerosol distributions from different release conditions. Each contour graph maps the concentration defined by the sampling results from the 18 to 22 diagnostic positions in each plane. Perimeter positions where the concentration was known to be zero (inside obstacles, at walls, and so forth) were included in the contour plots. Figure 3 shows the concentration plots of the average of three center releases at 8 AC/h (nominal) at 92- and 366-cm heights on the left, whereas the plots on the right show the same information for a ventilation rate of 4 AC/h (nominal). As an example of their usefulness the following comparisons may be made from the four plots:

- a. The cumulative peak concentrations measured at 4 AC/h are about twice those at 8 AC/h at each height.
- b. The positions of cumulative peak concentrations measured at each height are consistent for each ventilation rate, but they differ slightly for each height at different ventilation rates.
- c. The positions of cumulative peak concentrations at 4 AC/h are close to the release point and apparently exhibit little travel from the release, whereas at 8 AC/h the peak concentration occurs relatively far from the release point.
- d. In all cases the air concentration near the four corner exhausts is low.

Statistical analysis of the aerosol releases at DP-208 indicate that the results were significantly different at the three ventilation rates and significantly different for the three release locations, and that there were significant differences in tower locations. When the results were normalized by the ventilation rate, the ventilation rates were not significant, while the other variables were. Several interactions between concentration, sampler location, release position, and ventilation rate were also noted. The analysis indicated good reproducibility in the results, and any changes in the variables caused by ventilation rate changes should be observable for the three rates used. A similar analysis of aerosol sampling in TA-55 is not available yet.

**Table I. Aerosol Sampling In Two Laboratories.**

Ventilation Rate AC/h	Release Location Quadrant	Number of Releases	Average Local Concentration C( $\mu\text{g/L}$ )	Samples <Avg $C_0$ %	Average Ratio $C/C_0$ 62-183 cm
<b>DP-208 Laboratory</b>					
16	Center	3	0.00313	71.0	1.19
8	Center	2	0.00600	67.9	1.04
4	Center	2	0.01327	88.6	1.01
16	Northwest	2	0.00319	73.9	1.02
8	Northwest	3	0.00452	71.0	1.03
4	Northwest	2	0.00752	68.5	0.92
16	Southeast	2	0.00157	76.1	0.84
8	Southeast	2	0.00546	60.3	0.98
4	Southeast	1	0.00771	65.2	0.96
Summary:					
16	All	7	0.00270	73.3	1.04
8	All	7	0.00521	67.1	1.01
4	All	5	0.00985	75.9	0.96
<b>TA-55-208 Laboratory</b>					
8	Center	3	0.0018	60.3	1.01
4	Center	3	0.0028	63.7	1.07
8	Center <sup>a</sup>	3	0.0054	77.6	1.01
8	Northeast	3	0.0013	70.0	1.05
4	Northeast	3	0.0017	71.7	1.05
8	Southwest	3	0.0011	69.2	1.04
4	Southwest	3	0.0021	67.5	1.02
8	Southwest <sup>a</sup>	3	0.0039	67.1	1.05
Summary:					
8	All	9	0.0014	66.5	1.03
4	All	9	0.0022	67.7	1.05
8	All <sup>a</sup>	6	0.0046	72.4	1.03
<sup>a</sup> Two aerosol generators at the same location were used.					

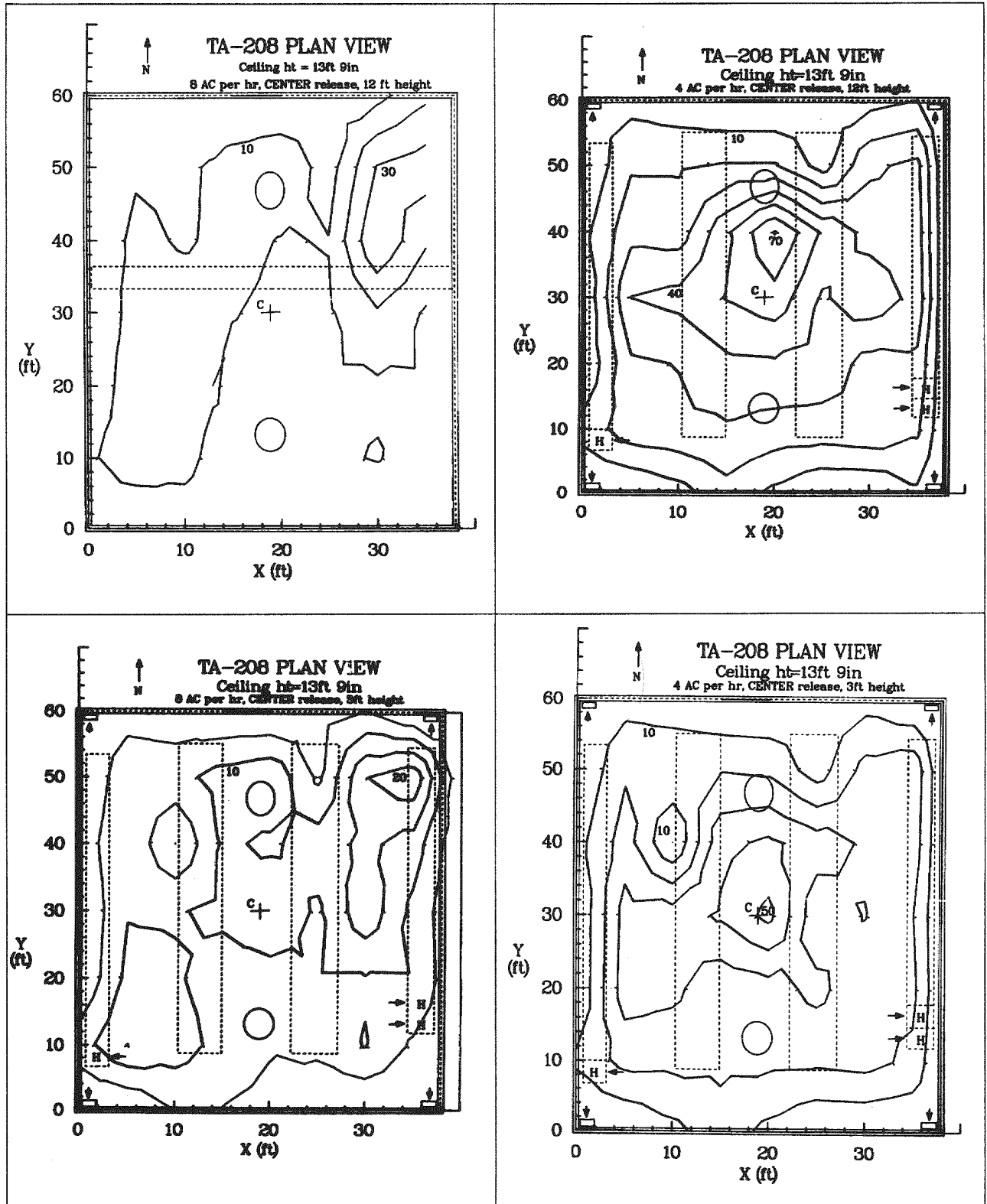


Figure 3. Aerosol concentration contour plots, TA-55, Room 208 at 8 and 4 AC/h (nominal) and 12 and 3 ft above floor. Numbers on contours represent  $\mu\text{g}/\text{L} \times 10^4$ .

**LPC Analysis.** Results from the laser particle counters (LPC) provided considerable information concerning the transient portions of the aerosol experiments. LPC total counts versus time were plotted in the formats shown in Figs. 4 and 5 to define concentration risetime, decay time, and lag time until LPC response (a function of distance from release and effective air movement velocity). Using the gas dilution equations,

$C = C_f(1 - \exp(-Qt/V))$  for the ideal concentration rise and

$C = C_0(\exp(-Qt/V))$  for the ideal concentration decay,

calculated values for instantaneous, perfect mixing may be compared with experimental values obtained from the graphs to compare rise and decay 1/2-times and obtain an effective mixing factor at each LPC. These may be compared for each release condition and ventilation rate. In each equation, C is the aerosol concentration at time, t;  $C_f$  and  $C_0$  are the final and initial concentrations, respectively; Q is the ventilation rate; and V is the free air volume of the room.

The averaged results appear in Tables II and III. Table II lists the decay or clearance 1/2-times, calculated from the LPC records for all aerosol runs in DP-208 for each ventilation rate. The clearance times (minutes) and standard deviations are shown for each release location and the average of all locations. From this average clearance 1/2-time, the ventilation rate in air changes per hour can be calculated using the exponential decay relationship.

More interesting are the calculated ideal rise 1/2-times for each release location, as determined graphically from LPC data (Table III).

The experimental rise 1/2-times are considerably less than the theoretical times because theory assumes instantaneous, perfect mixing, whereas the aerosol cloud actually mixes slowly but moves relatively fast with the air movement toward nearby downwind sensors. As an observation, the difference in 1/2-times, although not statistically significant, suggests that the aerosol concentration rises faster for lower ventilation rates. This is not an illogical trend because it is likely that at lower ventilation rates the aerosol cloud that reaches the photometer is

more concentrated because of poor mixing, thus producing a faster rise in LPC response. The decrease in the averaged standard deviations with increasing ventilation rate, although small, may indicate that increased mixing efficiency at higher ventilation rates decreased the variability of the 1/2-times observed.

The 50% detection times for both DP-208 and TA-55-208 (Table IV) were obtained from graphs of the risetime for each of the aerosol release groups (center, northwest, southeast). This is the time at which the trace of the LPC data plot reaches 50% of the final aerosol concentration. (This time is not the same as the rise 1/2-time; it will be the same only if the risetime line coincides with the data trace that passes through 50%.) No calculation or judgement is involved in obtaining the 50% detection time. It is an unequivocal point on the risetime graph where the concentration reaches 50%. Figure 4 illustrates the difference between rise 1/2-time and 50% detection time. For each ventilation rate and release location the minimum time that any of the four LPCs indicated 50% concentration is listed in Table IV. This is analogous to a listing of the first continuous air monitors to alarm in radioactive material releases. Surprisingly, in all cases in DP-208 the lowest ventilation rate produced the shortest 50% detection time, by a significant amount. Thus, in this particular laboratory, apparently a high ventilation rate and better mixing increased the LPC response time to the aerosol release.

An analysis, identical to that performed for DP-208, of LPC data obtained during the releases in TA-55-208 showed somewhat different results. Unlike at DP-208, the minimum 50% detection time was at the higher ventilation rate (8 AC/h) in all cases for the four LPCs (Table IV). This contradiction may be explained by the LPCs being farther from the release source in TA-55-208, thus increasing the lag time and decreasing initial cloud concentration at the LPC.

For the final six releases in TA-55-208, eight LPCs were located in the laboratory as indicated in Fig. 6. The LPCs labeled as P5 and P7 were at 92 cm above the floor (the same as LPCs 1 through 4), but LPCs P6 and P8 were 366 cm above the floor (62 cm below the supply diffusers). In all six releases, either LPC 7 or 8 was the first to indicate the 50% detection level (Table IV, last line). This suggests not only that the room air circulation is

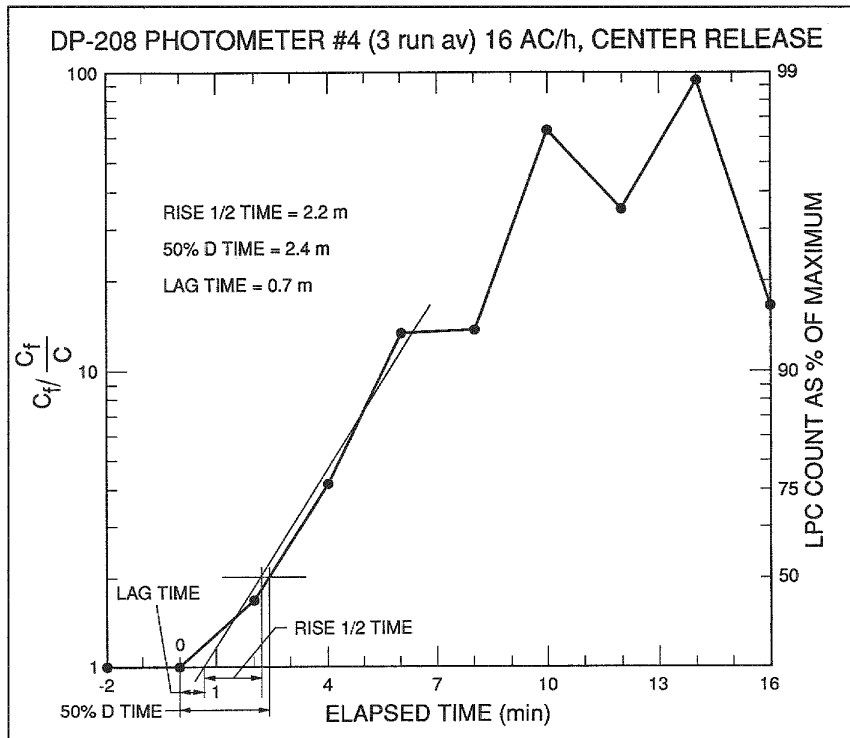


Figure 4. Example of LPC rise analysis. Lag time, 50% detection time, and rise 1/2 time graphical solutions are shown.

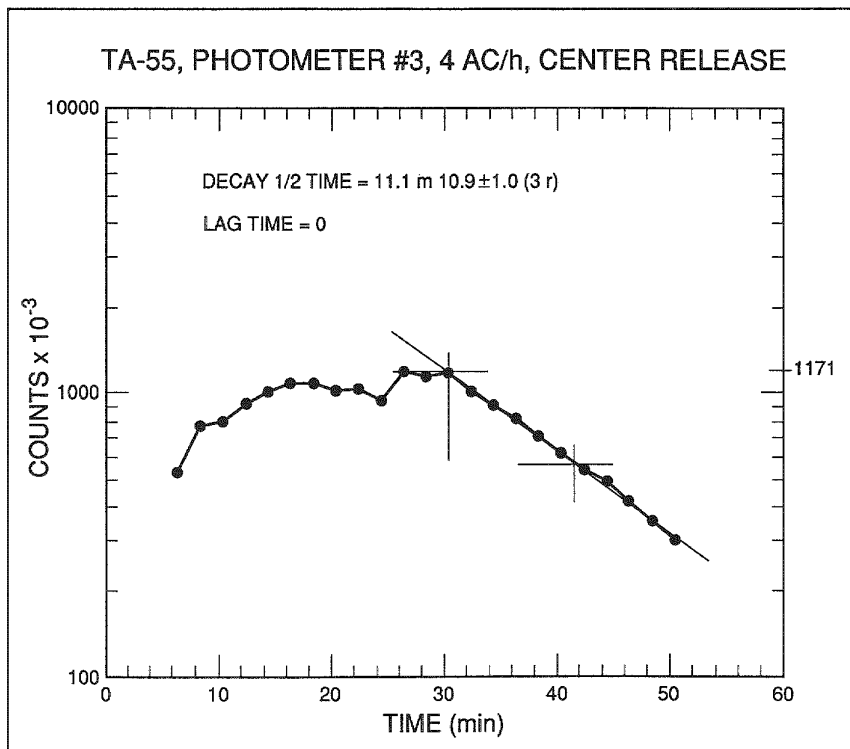


Figure 5. Example of LPC decay analysis. Graphical solutions for lag time and decay 1/2 time are shown. Aerosol generation time was 30 min.

Table II. DP-208 Ventilation Clearance Rates.					
	Clearance 1/2 Times (Minutes)				
Nominal AC/h Rates	Ideal IP Mix	Release Location			
		Center	NW	SE	Avg
4	9.5	9.4±0.6	9.0±1.4	9.4±1.2	9.3
8	4.9	7.3±0.5	7.1±0.3	6.9±0.9	7.1
16	2.4	2.3±0.5	2.6±0.5	2.1±0.6	2.3

Table III. DP-208 Laser Particle Counter Rise Times.						
	LPC Rise 1/2 Times (Minutes)					
Nominal AC/h Rates	Theory IP Mix	Release Location				
		Center	NW	SE	Avg	Avg SD
4	9.5	1.9±1.0	1.1±0.7	1.1±0.6	1.4	0.8
8	4.9	1.6±0.6	1.4±0.4	1.9±0.7	1.6	0.6
16	2.4	1.9±0.3	1.8±0.3	1.8±1.0	1.8	0.5

Table IV. Laser Particle Counter Minimum 50% Detection Times.					
	LPC 50% Detection Time				
Nominal AC/h Rates	Ideal IP Mix	Release Location			
		Center	NW	SE	
Total of 4 LPCs:					
DP-208:					
4	9.5	<u>1.9 / P4</u> (2) <sup>a</sup>	<u>0.6 / P2</u> (2)	<u>0.3 / P4</u> (1)	
8	4.9	<u>2.4 / P4</u> (2)	<u>1.2 / P2</u> (3)	<u>0.9 / P4</u> (2)	
16	2.4	<u>2.4 / P4</u> (3)	<u>1.3 / P3</u> (2)	<u>1.6 / P4</u> (2)	
TA-55-208:					
4		<u>2.9 / P3</u> (3) <sup>a</sup>	<u>4.9 / P3</u> (3)	<u>4.0 / P3</u> (3)	
8 (1 generator)		<u>2.0 / P3</u> (3)	<u>2.6 / P2</u> (3)	<u>1.7 / P3</u> (3)	
Total of 8 LPCs:					
8 (2 generators)		<u>1.6 / P8</u> (3)	<u>1.2 / P7, P8</u> (3)	No runs	
<sup>a</sup> Data is shown as: 50% Detection time in minutes / LPC number (no. of aerosol runs averaged). Time /LPC underlining indicates minimum 50% detection time.					

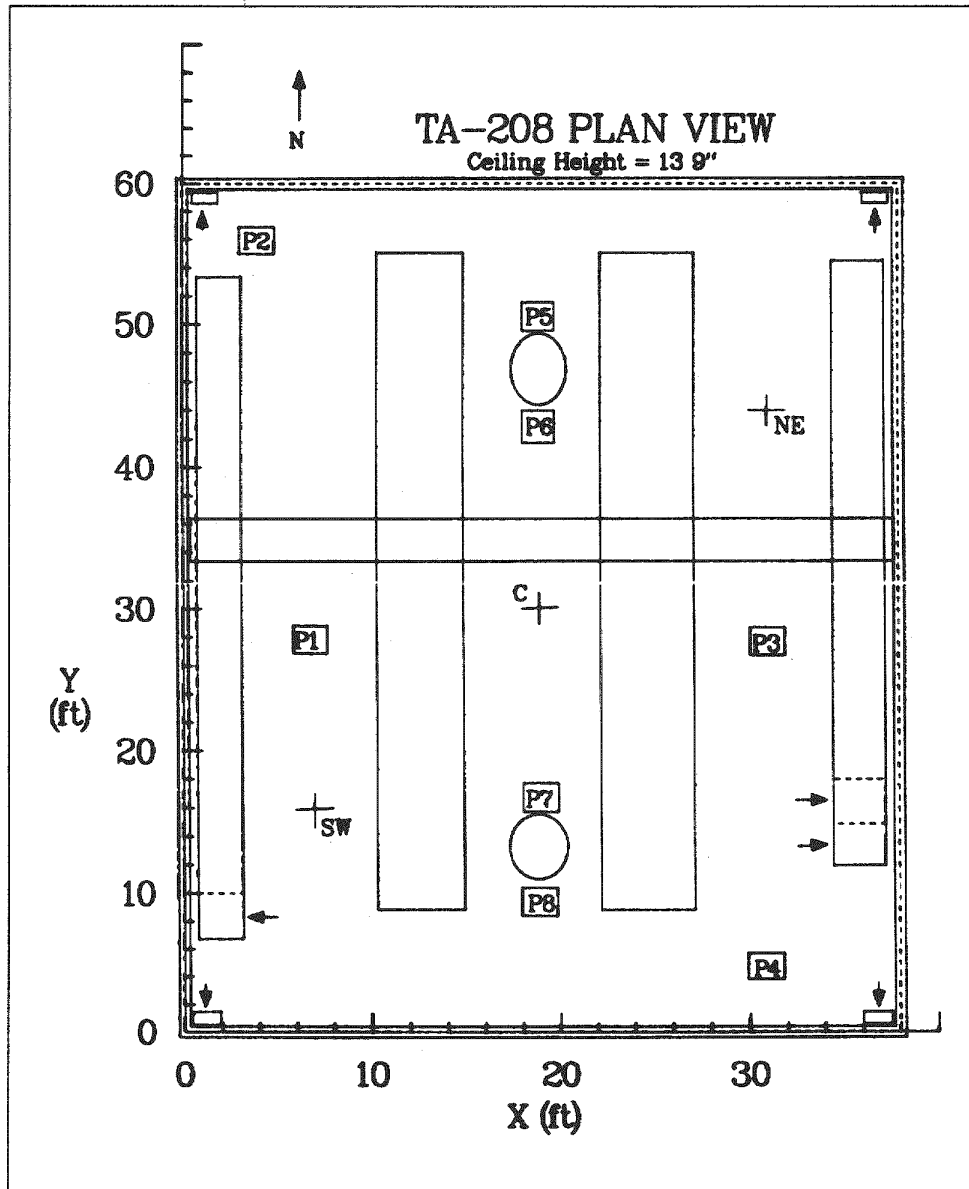


Figure 6. Plan view of TA-55, Room 208 laboratory showing glovebox lines (N-S rectangles) and transfer tunnel above gloveboxes (E-W rectangle). The two large circles are 3 ft diam ceiling supply air diffusers; arrows show positions of exhausts near floor and through hoods (H). Crosses labeled C, NE, or SW show aerosol release positions. Small rectangles, P1-P8, indicate laser particle counters; P5-P8 are directly beneath diffusers with P5 and P7 at 6 ft, and P6 and P8 at 12 ft above floor.

upward beneath the supply air diffusers, but that much of the room air is entrained in the air rising toward the diffuser. This is not surprising because the diffusers have been shown by others to have such a circulation pattern. However, this may be the first indication that directly beneath this type supply diffuser is an advantageous location for a detector.

**Modeling.** A computer code, SOLA-DM, is being used to simulate the air circulation of each of the test laboratories used in the present study. The code solves the three-dimensional, time-dependent, finite-difference, incompressible Navier-Stokes equations for fluid dynamics. Inflow is prescribed by the user for each supply inlet in terms of volume flux, and outflow is prescribed by distributing the volume flow according to output velocities. The computational domain is a mesh of parallelepiped cells in Cartesian geometry that need not be uniform in the three orthogonal directions. Flow obstructions such as gloveboxes, equipment, and such are modeled as solid obstacles. Particles may be injected into the problem at locations corresponding to aerosol release positions to trace major airflow patterns within the room. At present, small inertialess particles are simulated. Likewise, at present a constant turbulence factor and no turbulent or Brownian diffusion is included in the particle trajectory calculations.

As a major part of the application of SOLA-DM to modeling the three labs, validation at each stage of the aerosol testing has been conducted. Like the ventilation study itself, code validation progressed from a simple-geometry test chamber to increasingly complex laboratories. Initially, the

SOLA-DM code was selected from two codes that were evaluated against air flow measurements of the flow field in a 25 m<sup>3</sup> cubical chamber that circulated air at 16 AC/h. Subsequently, during and after each stage of validation, the code was improved to better predict the flow field in the laboratory tests. Validation was performed by two independent methods as described below.

1. The first method was a comparison of measured velocity vectors to code-predicted vectors at identical spatial positions. This method determined the agreement of the code with actual conditions at specific locations.
2. The second was to compare the results of an aerosol release within the laboratory. The code-predicted distribution of an injection of particles was compared with the experimentally determined distribution of an aerosol released in a similar location in the laboratory.

For the comparison of experimental versus code-predicted vectors, the velocity magnitudes were measured using a thermal anemometer with up to eight omnidirectional sensors at specific locations in the room. Direction of airflow was measured with sensitive wind vanes that provided three-dimensional directional data. Total velocity vectors could be measured down to about 5 cm/s, and the velocities generally observed were between 2 and 20 cm/s. A compilation of the comparison of velocity components shows (Table V) that the agreement was good in the small cubical chamber but was worse in the larger, more complex laboratories.

Table V. Comparison of Predicted vs. Measured Velocity Vectors.					
Room	AC/h	Magnitude	Direction		Total Vector Mag/Azi/Elev
			Asimuth	Elevation	
ETC	16	9 of 14	11 of 14	8 of 14	6 of 14
DP-208	16	29 of 39	10 of 36	23 of 38	8 of 38
DP-208	8	15 of 36	18 of 36	19 of 36	5 of 36
TA-55-208	8	16 of 22	14 of 22	11 of 22	9 of 22

Comparing the code-predicted and measured values of several velocity vectors determined in the foregoing manner provided an indication of the validity of the code. This comparison is summarized for each of the laboratories tested to date, including the original small environmental test chamber used for initial selection of the code. Criteria for agreement was that the numerical velocity magnitude be within a factor of two of the measured magnitude at velocities greater than 5 cm/s, and the numerical and measured azimuth and elevation angles agree within 45°. The columns of Table V indicate the number of vector components that agreed, of those that were available for comparison. Generally, greater than 50% of the magnitudes agreed within the criteria, whereas the directional agreement usually exceeded 50%. When total vectors were compared (a very stringent test), fewer of the vectors agreed, but TA-55-208 showed some improvement, probably because the code input values (velocities) were known relatively accurately.

The model was also validated using the concentration contour plots discussed earlier and three-dimensional plots of particle distribution calculated by SOLA-DM. Code-predicted particle distribution, plotted in a three-dimensional representation of the room, is displayed at a specific time after injection. Views of the room and distribution from different directions are available so that a perspective of the location of the particles may be obtained. In addition, a printout of the number of particles in selected calculational cells can be obtained.

The aerosol distributions from the releases in Room TA-55-208 may be compared with the particle distributions from code output for the same ventilation rate and release position. Results of aerosol dispersion during a center release at 8 AC/h (Fig. 3, left plots), discussed earlier, show contour plots of aerosol distribution at the 92- and 366-cm horizontal sampling planes. This clearly shows the areas of highest integrated concentration in each plane. The aerosol

concentration to the northeast of the release is higher than elsewhere, and the concentration south of the release is low at the sampling locations. The aerosol evidently drifts northeastward and upward, forming a high concentration area near the ceiling in the northeast quadrant.

Corresponding three-dimensional particle-distribution simulations are shown in SOLA-DM output (Figs. 7 and 8), depicting the same release position and room conditions as Fig. 3, left. The distribution of particles is shown from a 21,800-particle continuous injection starting after equilibrium flow is reached and ending 200 s later. The distribution is much like that indicated in the actual release, with the particles being mostly in the northwest quadrant of the room (Fig. 7, looking down), with little spreading of particles to the south. This is confirmed looking from a different view (Fig. 8, view looking north), which shows particles high and to the west of the release, with the highest concentration near the west wall. The model indicates a dense plume rising toward, and being dispersed by, the outflow of the south diffuser. This is probably why LPCs directly beneath the diffusers detect the aerosol first, as discussed earlier. The aerosol dispersion is minimized by the model because no turbulent or Brownian diffusion is included. As a consequence, the model emphasizes the main directional airflow rather than particle spread.

The final series of experiments for this study are being conducted at the Rocky Flats Plant, Building 371, Room 3305. At the conclusion of this series, a comprehensive analysis of the results from the three laboratories studied should provide information concerning the many effects of changes in ventilation rates in these and similar facilities. From the results of the experiments and modeling, recommendations will be made concerning the ventilation of such laboratories, and guidelines for the design of such facilities will be developed to supplement present DOE design criteria.

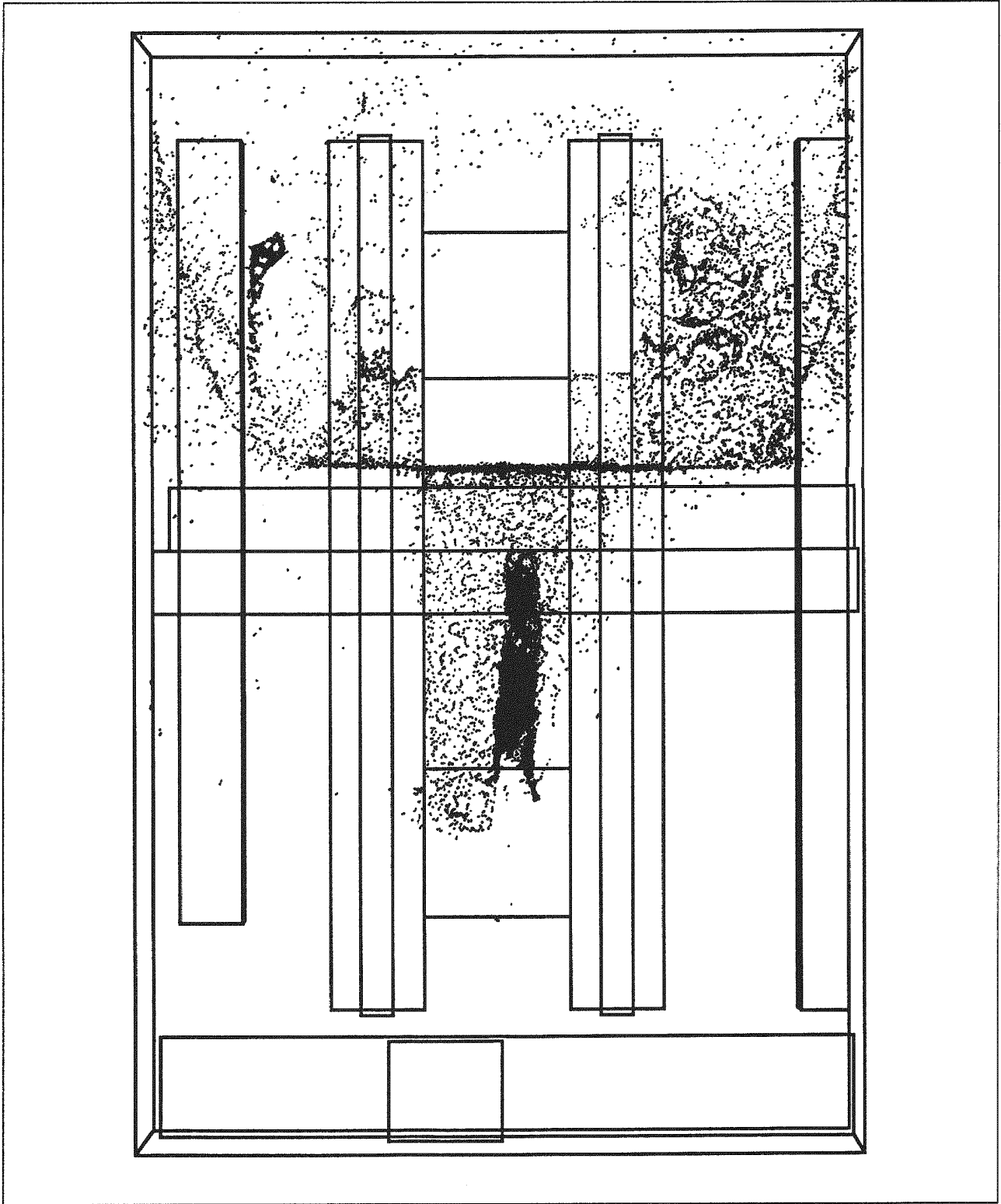


Figure 7. SOLA-DM numerical simulation of 20 000 particles injected over 440 sec. Source is at center of room and top of figure is north. Glove box lines and obstacles are outlined by rectangles. View is looking upward.

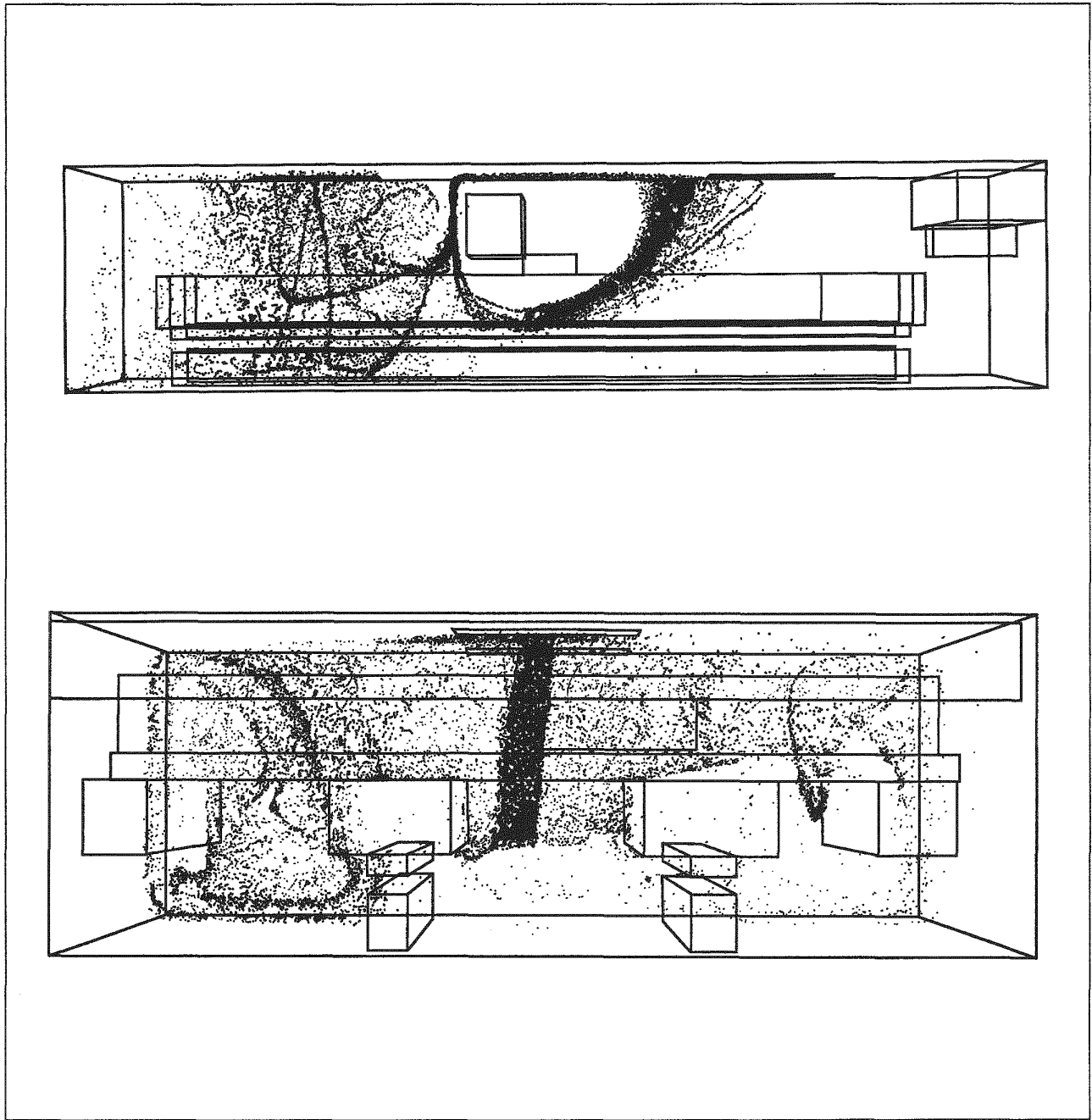


Figure 8. Two horizontal views of same simulation as Figure 7. Source is at center near bottom of glove boxes. Particles rise toward supply air diffusers before spreading. View (top) is looking east; view (bottom) is looking north.

## **TECHNICAL SUPPORT FOR FILTER TEST FACILITIES**

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Transportation Division, Interim Waste  
Operations*

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### **INTRODUCTION**

This program provides technical support to the DOE Filter Test Facilities (FTFs) both by carrying out scheduled activities and assisting in the solution of special problems. Continuing activities include: (1) initiating and reviewing Round-Robin Tests (RRTs), (2) summarizing yearly activities of the FTFs, and (3) providing technical consulting services to the FTFs. In addition to these scheduled activities, Los Alamos is available to provide technical assistance to the DOE and FTFs in addressing special problems. For 1988, special assistance was provided for the transfer of the High Flow Alternative Filter Test System (HFATS) to the FTFs and implementation of new flow calibration plates.

### **ROUND-ROBIN TEST PROGRAM**

The RRT program monitors the consistency of FTF measurements by circulating six high-efficiency particulate air (HEPA) filters among the three FTFs. One of the FTFs serves as a reference site, testing the filters before and after they are sent to the other FTFs. This reference testing documents that the test filters have not been affected by the test program and provides information on the internal consistency of the reference FTF. The goals of the RRT program are to (1) document measurement consistency among and within individual FTFs, (2) suggest areas where improvement of FTF measurement accuracy and precision may be possible, and (3) monitor

and document the effects on FTF measurements of implementing these proposed improvements.

In 1988, Los Alamos coordinated two RRTs, interpreted their results, and identified trends in the data collected over the history of the RRT program. This year's RRT results indicate certain improvements in measurement consistency among the FTFs that might be associated with actions suggested by earlier RRT results and implemented by the FTFs. Other results point to areas where FTF measurements might be improved.

### **FTF SEMIANNUAL REPORT SUMMARY**

In the fall and spring each FTF issues a report detailing numbers of filters accepted and rejected for each shipment during the previous six months. Los Alamos examines these data to assess the consistency of FTF tests and the quality of filters provided by individual manufacturers. With this evaluation, we hope to (1) provide a standard frame against which the performance statistics of the individual FTFs can be evaluated and (2) provide information on the performance of filters being used by DOE facilities for use in ventilation exhausts and other air-flow systems.

The first objective is met by monitoring the consistency of measurements among the FTFs. This analysis, in conjunction with the results of the ongoing DOE RRT program, provides DOE with documentation on the consistency of measurements among the FTFs.

The second objective is met by measuring the quality of filters presented to the DOE FTFs and the quality of filters installed in DOE facilities. The quality of filters received by the FTFs affects the FTFs' operating costs and potential delays in the mission of the DOE contractor purchasing the filters.

Analysis of FTF data from FY 1987 and FY 1988 is in progress.

## **SPECIAL TECHNICAL ASSISTANCE**

Los Alamos staff provides consultation on technical problems that affect individual FTFs and the FTFs collectively. These problems deal with compliance to FTF standards, potential improvements that are indicated by the RRT program or the review of FTF data, or special problems that the FTFs encounter during operation. Los Alamos involvement in solving the technical problems has included design of equipment, modifications, proof-testing of these modifications, analysis of data collected by the FTF, surveying and interpreting literature, and developing technical positions on FTF issues.

## **HFATS SUPPORT ACTIVITIES**

Los Alamos has developed an alternative to the current filter test system (Q107) used to test Size 4 (500 ft<sup>3</sup>/min rated flow) and larger nuclear-grade HEPA filters at DOE FTFs. This new test system, called the HFATS, consists of an air-operated nozzle aerosol generator and a laser aerosol spectrometer (LAS)/diluter monitoring system. Three major activities aided to further the transfer of the HFATS: (1) installation of a Los Alamos prototype HFATS at the Rocky Flats FTF, (2) support of the FTFs in the acquisition of their own HFATS, and (3) technical guidance in designing and conducting a HFATS RRT.

Los Alamos staff installed a prototype HFATS at the Rocky Flats FTF and trained Rocky Flats staff in its operation. While at Rocky Flats, the FTF staff practiced operating the HFATS and made drawings for use in fabricating their own system. Late in CY 1988, the prototype system was shipped to Oak Ridge FTF, where the drawings are to be completed and certain HFATS components are to be fabricated.

Los Alamos prepared blueprints and specifications to help support the FTFs in building their own HFATS. In addition, we provided computer software for operation of the laser spectrometer used with the HFATS. Finally,

spectrometers modified by the manufacturer specifically for HFATS use were calibrated before being sent to the FTFs. All the FTF laser spectrometers are being calibrated at Los Alamos.

An HFATS RRT is planned when each of the FTFs is operational with its own HFATS. The RRT is to be coordinated by Rocky Flats FTF staff. An initial RRT design suggested by Los Alamos is currently being finalized by Rocky Flats staff with technical assistance from Los Alamos. A set of 100 Size 5 (100 ft<sup>3</sup>/min rated flow) filters and 20 Size 6 (1250 ft<sup>3</sup>/min rated flow) filters was obtained by Rocky Flats FTF. Many of the filters were rejected because of penetration problems.

## **FLOW CALIBRATION PLATES**

Results of the FTF RRT program and the technical consultation visits made to the FTFs indicated the need for updated flow calibration plates at the FTF. New plates were designed at Los Alamos in FY 1986.<sup>1</sup> After fabrication in FY 1987, the plates were forwarded to Oak Ridge Gaseous Diffusion Facility for calibration. Calibration was completed in FY 1988 and the plates were distributed.

Los Alamos staff wrote a program for the HP-85 microcomputer to interpret results of the calibration for the FTFs. The program allows the FTFs to easily set test flows based on plate calibration data. Algorithms developed from regression analysis of calibration data are solved by the computer program. These solutions give plate differential pressure that corresponds to a desired test flow for a particular set of ambient conditions. Copies of the program along with an operation manual were distributed to the FTF in 1988.

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# PERMEATION OF CHEMICAL MIXTURES THROUGH POLYMERIC MATERIALS

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## INTRODUCTION

Chemical protective clothing (CPC) is often used to aid in the protection of employees from excessive chemical exposure. Because no item of CPC can provide complete protection indefinitely, there is a strong interest in trying to understand the barrier properties of CPC material against chemical permeation.

Historically, the two permeation measurements most reported are (1) breakthrough time, and (2) steady-state permeation rate. Breakthrough time estimates the immediate protection afforded by the CPC. The steady-state permeation rate estimates the amount of chemical that will potentially contact the skin through the CPC. These permeation measurements have usually involved challenging CPC materials with neat (pure) chemicals. In contrast, an employee in the industrial environment is very likely to encounter mixtures rather than single-component chemicals. Currently, there are no available methods for estimating the permeation behavior of mixtures from previous test results on single-component challenges.

Chemical mixtures exhibit a unique permeation behavior. It has been suggested<sup>1</sup> that for mixtures the most rapidly permeating component carries the slower permeating component through the CPC material at a faster rate than if the slow component were challenged in its neat form. In another study,<sup>2</sup> the authors state that binary mixtures may increase chemical exposure of employees over that of neat chemical components in three ways:

1. It may decrease the breakthrough time of the components.
2. A component that does not permeate in its pure form may be transported through the CPC by another component in the mixture.
3. The collective permeation rate for the mixture may be higher than the permeation of either pure component.

This research was directed toward learning more about how chemical binary mixtures permeate through CPC materials. The hypothesis examined is based on the fact that neat compounds that possess a high permeation rate in a CPC material also tend to swell the material during permeation. With a binary mixture composed of a high and a low permeation rate component, the low-permeation rate component may actually diffuse through the high-permeation rate component (which has swollen and partially filled the material matrix) rather than diffusing through the material itself. This principle was examined using both permeation and immersion experiments with binary mixtures.

## PERMEATION THEORY

Permeation consists of three processes: solution of the challenge in the upstream side of the membrane, molecular diffusion through the membrane, and desorption from the downstream side. By Fick's first law,

$$J = -D \frac{dc}{dx} \quad (1)$$

where

J = rate of permeation

D = diffusion coefficient

$\frac{dc}{dx}$  = concentration gradient of the challenge in the material.

Various methods are used for calculating the diffusion coefficient. A popular one uses the lag breakthrough time,  $T_L$ ,

$$D_L = \frac{2}{6T_L} \quad (2)$$

where  $T_L$  is the time determined by extrapolating the steady-state portion of a cumulative permeation curve to the time axis. When the membrane swells, the diffusion coefficient is considered concentration dependent. A suggested solution<sup>3</sup> is

$$D_V = \frac{D_L}{(\text{volume fraction of polymer})^3} \quad (3)$$

## EXPERIMENTAL

To examine the hypothesis, two types of challenge series were tested. A challenge series consists of two chemical components matched with a CPC material. The first type of challenge series (three series tested) was selected to conform to the following criteria:

1. One component (component B) of the series exhibits a predicted low permeation rate and the other (component A) exhibits a predicted high permeation rate when challenged neat against the specific material.
2. The material swells considerably when challenged against component A.

The second type of challenge series (one series tested) was selected to conform to the following criteria:

1. One component (component A) of the series exhibits a moderate to high predicted permeation rate and causes negligible swell in the material.
2. The second component (component B) exhibits a very low predicted permeation rate.

Both immersion and permeation tests were conducted in triplicate for all series. Five challenge concentrations were used with all series: (1) 100% component A, (2) 100% component B, (3) 25-75% by volume mixture of A and B, (4) 50-50% mixture,

and (5) 75-25% mixture. Three common generic CPC materials were chosen to avoid any complications associated with flocked, lined, or composite materials. The materials and chemical components for each series are listed in Table I.

Series	Material	Components
1	Nitrile	A) Benzene B) Carbon Tetrachloride
2	Butyl	A) n-Hexane B) Toluene
3	Neoprene	A) Carbon Tetrachloride B) Nitrobenzene
4	Neoprene	A) n-Hexane B) Isopropyl Alcohol

For the permeation tests, all material specimens with visual irregularities were discarded. Specimens were taken mainly from the gauntlet area of gloves, which yielded a plentiful source of consistent material. The mean thickness of each specimen was determined to the nearest 0.005 mm from five equally spaced points in the challenge area. Three material specimens with equal mean thicknesses were randomly assigned to challenge concentrations within a series.

Each specimen was mounted in a standard ASTM permeation cell with the normal outside surface of the specimen facing the challenge side. The cell was placed in a water bath to maintain a temperature of  $23^\circ \pm 1^\circ\text{C}$ . Once the cell was placed in line, the flow of the collection medium (filtered dry air) through the cell was adjusted to  $500 \text{ cm}^3/\text{min}$ . Standard concentrations of the challenge components were produced with two AID Model 309 standard generators with diffusion tubes and chromatographic-grade chemicals. Clean compressed air served as background. All analyses were conducted using a Dycor Model M200MDE quadrupole gas analyzer in an intensity-vs-time mode.

During the experiment, a timed sequence of standard, background, and cell concentrations were continuously monitored in an open-loop system. All cell runs were terminated after it appeared that steady-state permeation was achieved or 24 h had elapsed. The cell was then dismantled and a final thickness recorded.

For the immersion tests all specimens were produced using a 1-in.<sup>2</sup> punch. Specimens were washed in distilled water, then placed between filter paper and allowed to dry. The initial thickness, width, and mass for each specimen were recorded to the nearest 0.005 mm, 0.40 mm, and 0.0005 g, respectively. Specimens were then immersed in approximately 50 mL of challenge in a covered 150 mL beaker. All experiments were conducted in triplicate, at a temperature of 23° ± 2°C, and in the absence of direct light. Two test durations were examined. The first was 24 h with measurements at 1, 4, 8, 12, and 24 h. The second was 2 h with measurements every 10 min. All data were collected following the ASTM (D471) standard procedure for determining change in mass and volume.

## RESULTS AND DISCUSSION

### IMMERSION TESTS

In what follows, the following assumptions must be kept in mind: (1) there are no cavities in the material where the challenge could reside and yield an increase in mass with no corresponding increase in volume, (2) no chemical reactions are occurring that could cause an increase in material volume without a corresponding increase in mass, and (3) all mixtures are ideal solutions where no volume change occurs upon mixing.

For all pure challenges, measured volume changes ( $V_D$ ) were compared with volume changes calculated by dividing the measured mass changes by the densities of the pure challenges ( $V_M$ ). If assumptions 1 and 2 above are correct,  $V_D$  should equal  $V_M$ . This was true for series 1, 3, and 4 but not for series 2.

With the three assumptions above, the amount of both components A and B in a swollen membrane can be calculated:

$$M_{AC} = \frac{dV - (dM/\rho_B)}{1/\rho_A - 1/\rho_B} \quad (4)$$

and

$$M_{BC} = dM - M_{AC} \quad (5)$$

where

$M_{AC}$ ,  $M_{BC}$  = calculated masses of A and B, respectively

$dV$  = measured volume change

$dM$  = measured mass change

$\rho_A$ ,  $\rho_B$  = liquid densities of A and B, respectively.

To simplify comparisons, mass values were converted to moles.

If observed swelling is due to the even entry of the challenge into the material, the mole fractions of components A and B ( $X_{xm}$ ) in the material should equal that of the challenge ( $X_{xc}$ ). The calculated ratios,  $X_{xc}/X_{xm}$ , are shown in Table II.

These results show that with the exception of series 4, there is more of component A and less of component B in the material than in the challenge at steady-state. Also, both component ratio values increase as the percentage of component A increases in the challenge. This indicates that the material has a greater affinity for component A and as component A becomes more plentiful in the challenge, the material absorbed less of component B. Thus, the selectiveness of the material absorption was driven by the relative solubilities of the components for the material. The irregular ratio values of series 4 should be ignored and are probably due to the random errors associated with the small mass and volume measurements in the series.

Table II. Steady-State Mole Fraction Ratio Values ( $X_{xc}/X_{xm}$ ).				
Series	Component	Challenge Concentrations (A-B % by volume)		
		25-75%	50-50%	75-25%
1	A	0.863	0.839	0.916
	B	1.063	1.265	1.425
2	A	0.750	0.811	0.842
	B	1.100	1.234	1.928
3	A	0.691	0.759	0.868
	B	1.190	1.530	1.969
4	A	0.763	0.096	0.018
	B	0.625	-0.25	0.005

Permeation rate is usually reported in terms of unit area of membrane exposed. The area chosen is usually that at the beginning of the experiment. However, not only the thickness but the area of the test specimen usually increases during the test. Final thickness is much easier to measure than final challenge area. If area increase is well correlated with thickness increase, the final challenge area could possibly be calculated and used in the determination of permeation rates. This was examined by plotting the fractional changes in area against the corresponding fractional changes in thickness. Regression analysis revealed that the slopes for the four series varied from 0.52 to 1.06. These results displayed that this is not an adequate method of estimating the final challenge area.

In a similar vein, the final fractional increases in membrane thickness from the immersion tests were compared with those from permeation tests. In all cases, immersion produced significantly greater swelling than did permeation. This indicates that the two-sided immersion test overestimates the thickness increase in a one-sided (permeation) exposure.

### PERMEATION TESTS

A number of methods were used to calculate the diffusion coefficient in experiments with neat

challenges. First, initial and final thicknesses were used to calculate lag time diffusion coefficients ( $D_{LO}$  and  $D_{LF}$ , respectively) using Eq. (2). The second method used immersion weight-change data to calculate a D-value,  $D_M$ :

$$D_M = 0.04939 \ell_{1/2}^2 / T_{1/2} \quad (6)$$

where

$T_{1/2}$  = time for half the final amount of challenge to be absorbed

$\ell_{1/2}$  = thickness of specimen @  $T_{1/2}$ .

The third method,  $D_V$  from Eq. (3), was only applied to  $D_{LO}$  values. The results of these calculations are given in Table III.

In all cases, method 1, using initial thickness, gave the smallest coefficients,  $D_{LO}$ . Although this is the most commonly used calculation for the diffusion coefficient, it is most probably incorrect if the material swells. For series 1 and 3, the  $D_{LF}$  and  $D_M$  values are similar, and both of these compensate for thickness changes.

$D_V$  also compensates for thickness change but, in general, the values are considerably larger

Table III. Calculated Diffusion Coefficients (cm<sup>2</sup>/min).

Series	Component	D <sub>LO</sub>	D <sub>LF</sub>	D <sub>M</sub>	D <sub>V(DLO)</sub>
1	A	8.29 X 10 <sup>-6</sup>	1.34 X 10 <sup>-5</sup>	1.12 X 10 <sup>-5</sup>	1.82 X 10 <sup>-4</sup>
	B	7.14 X 10 <sup>-7</sup>	9.54 X 10 <sup>-7</sup>	9.11 X 10 <sup>-7</sup>	3.17 X 10 <sup>-6</sup>
2	A	2.75 X 10 <sup>-5</sup>	6.13 X 10 <sup>-5</sup>	3.68 X 10 <sup>-5</sup>	8.93 X 10 <sup>-4</sup>
	B	1.52 X 10 <sup>-5</sup>	3.32 X 10 <sup>-5</sup>	2.58 X 10 <sup>-5</sup>	4.53 X 10 <sup>-4</sup>
3	A	9.80 X 10 <sup>-6</sup>	1.95 X 10 <sup>-5</sup>	1.71 X 10 <sup>-5</sup>	3.41 X 10 <sup>-4</sup>
	B	4.16 X 10 <sup>-6</sup>	8.98 X 10 <sup>-6</sup>	9.86 X 10 <sup>-6</sup>	1.07 X 10 <sup>-4</sup>
4	A	4.39 X 10 <sup>-6</sup>	5.15 X 10 <sup>-6</sup>	1.08 X 10 <sup>-5</sup>	8.14 X 10 <sup>-6</sup>

Note: For component B (series 4), no breakthrough occurred during permeation testing and no change in volume was detected during immersion testing. Thus, no values for D could be calculated.

than either D<sub>LF</sub> and D<sub>M</sub>. The conclusion to be drawn from these results is that we are not sure how to calculate the diffusion coefficient of neat components through these materials, much less when mixtures are involved.

The lag breakthrough time (T<sub>L</sub>) of each component from all series was determined. These values were plotted against challenge composition for all series (Figs. 1-4). A significant decrease in T<sub>L</sub> for component B was observed as the concentration of component A in the challenge increased for all series except series 2. In series 2, only a slight decrease in T<sub>L</sub> for component B was observed. In many cases the T<sub>L</sub> of component B was essentially simultaneous with that of component A. In all series there seems to be a dilution effect which causes the mean T<sub>L</sub> for component A to increase as its concentration in the challenge decreases. We have not yet been able to quantify this effect.

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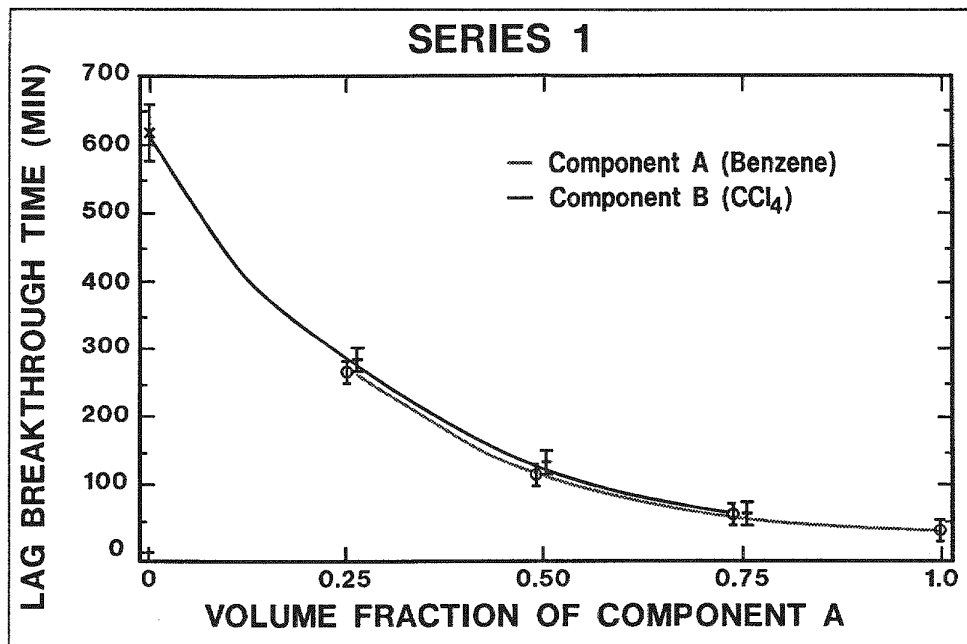


Figure 1. Mean lag breakthrough time vs volume fraction of component A in mixture challenged against nitrite. Bars represent one standard deviation ( $n = 3 - 5$ ).

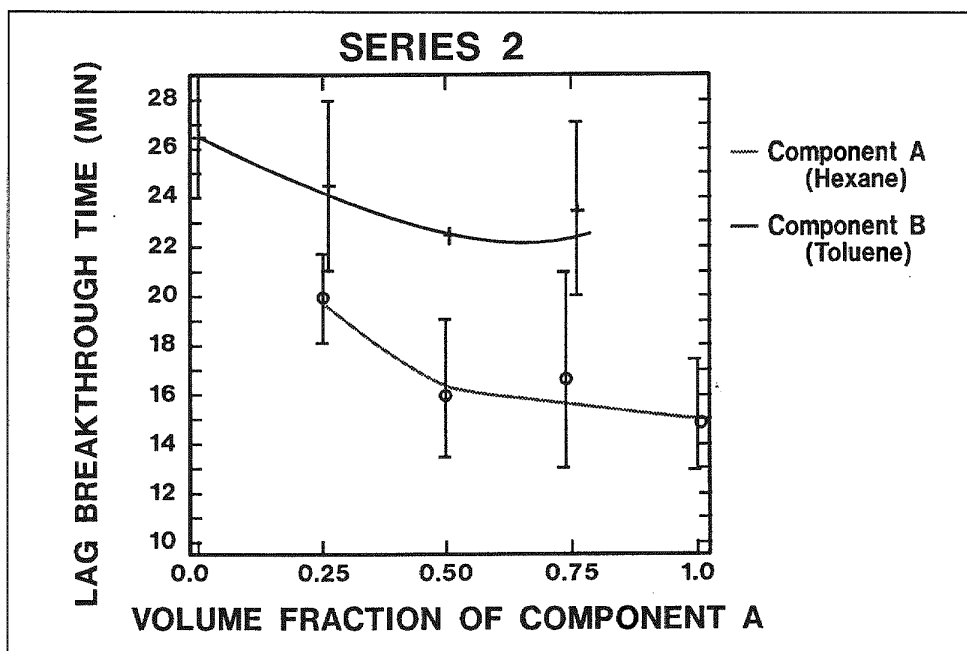


Figure 2. Mean lag breakthrough time vs volume fraction of component A in mixture challenged against butyl. Bars represent one standard deviation ( $n = 3 - 4$ ).

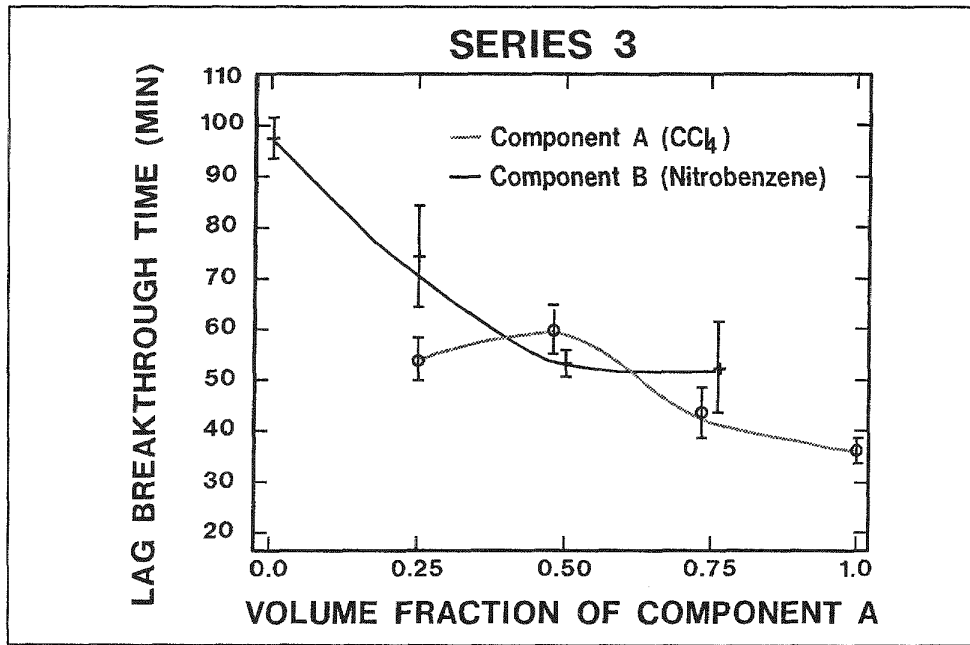


Figure 3. Mean lag breakthrough time vs volume fraction of component A in mixture challenged against neoprene. Bars represent one standard deviation ( $n = 3$ ).

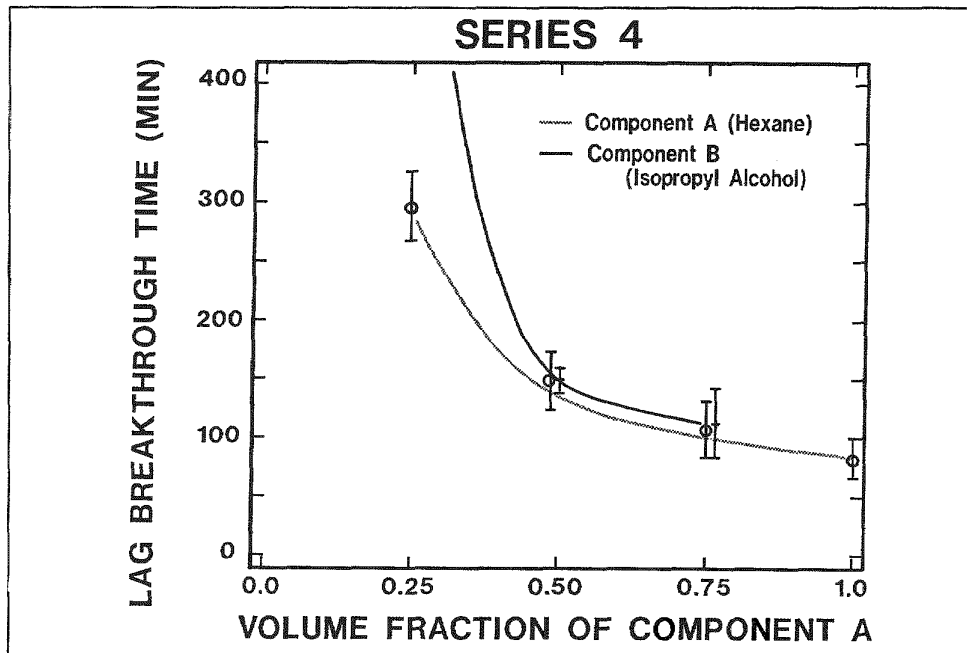


Figure 4. Mean lag breakthrough time vs volume fraction of component A in mixture challenged against neoprene. Bars represent one standard deviation ( $n = 3 - 5$ ).

# **EVALUATION OF DUST-RELATED HEALTH HAZARDS ASSOCIATED WITH AIR CORING AT G-TUNNEL, NEVADA TEST SITE**

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Personnel of the Yucca Mountain Project (YMP, formerly the Nevada Nuclear Waste Storage Investigations Project) investigated the technical feasibility of air coring in G-Tunnel at the Nevada Test Site. We completed an industrial hygiene evaluation and wrote the final report concerning the health hazards associated with the dust resulting from these tests. The study was divided into four tasks: Bulk Analysis, Evaluation of Dust Collection Equipment, Field Sampling, and Recommendations for the Exploratory Shaft Facility at Yucca Mountain.

## **TASK 1- BULK ANALYSIS**

We used scanning electron microscopy (SEM) and energy-dispersive x-ray (EDX) analysis to examine one erionite and one mordenite bulk mineral sample from Yucca Mountain and one commercial natural erionite sample. Two of the three samples contained significant quantities of microscopic fibers and fiber bundles. The third sample (the Yucca Mountain erionite) was amorphous with very few discernible fibers. EDX analysis of each sample indicated the common presence of aluminum, silicon, potassium, and calcium as major compositional elements, but the spectra were so similar that they were indistinguishable from one another. We concluded that x-ray diffraction (XRD) analysis is also necessary for conclusive identification/quantification of erionite and mordenite. SEM should be used for morphology, EDX for elemental composition, and XRD for positive crystal structure identification and quantification.

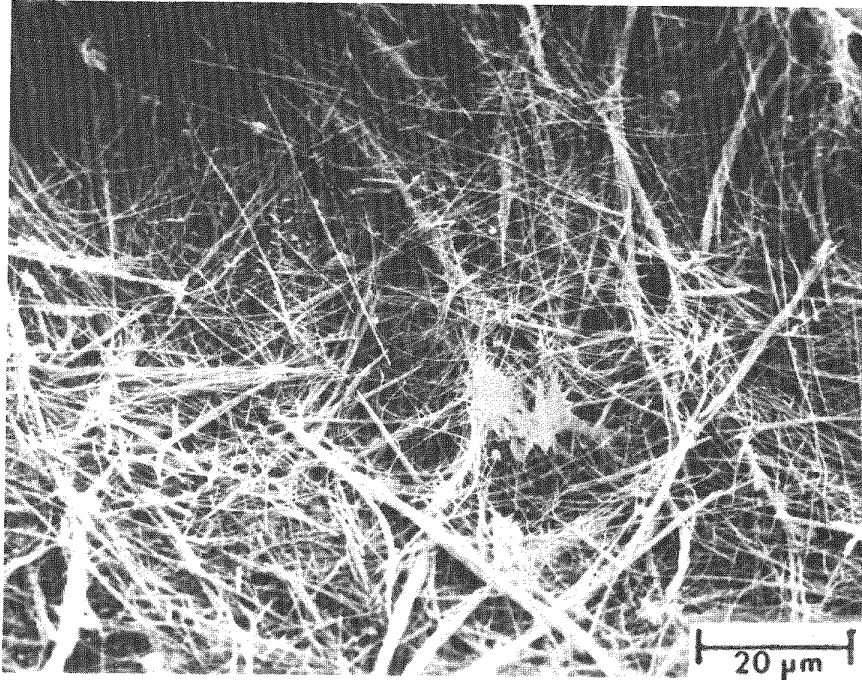
Figure 1 is a scanning electron micrograph illustrating the fibrous nature of the single Yucca Mountain mordenite sample examined. This reference mordenite, obtained from ESS-1 drill core material, contains many microscopic fibers of uniform diameter but various lengths. Fiber diameters measured in the sample ranged from ~0.08 to 3.0  $\mu\text{m}$ . The sample contained very few free (unattached) fibers.

## **TASK 2 - EVALUATION OF DUST COLLECTION EQUIPMENT**

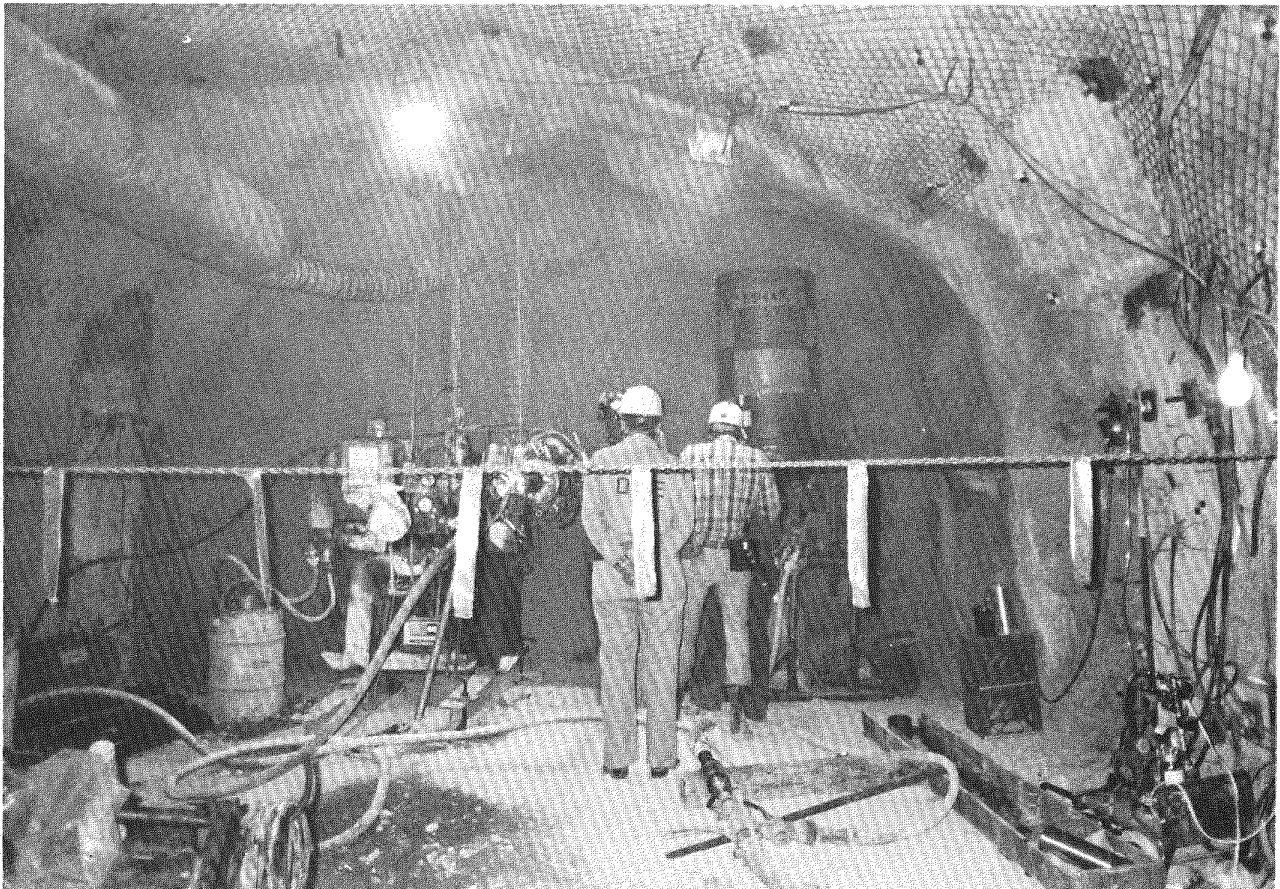
The proposed dust control equipment to be used in dry drilling was examined using the available manufacturer's technical literature, user experience, and regulatory agency experience. Based on available data on silica content, type, and particle size, and on proposed air coring (dry drilling) operations, we estimated that allowable exposures to silica-containing dust could range from 0.07 to 1.5  $\text{mg}/\text{m}^3$  (total dust) at the drilling sites. Our conclusion was that actual airborne concentrations of dust might approach or exceed these values during normal operations. As a result, the project management was prepared to control emissions and exposures during prototype air coring. Emission controls were provided where feasible, employees wore air-purifying respirators, and work practices were modified to minimize emissions and personal exposures. As an example of the emission controls applied, two exhaust ventilation ducts were added for fugitive dust control.

## **TASK 3 - FIELD SAMPLING**

During March and April 1988, as the prototype air coring of two holes, AC-1, cored in the Demonstration Drift (Fig. 2), and AC-2, cored in the Laser Drift, progressed, we collected personal and area air-particulate membrane filter samples and cascade impactor sets. Major dust collection system modifications were made during coring of AC-1.



*Figure 1. Mordenite from Yucca Mountain Core Hole ESS-1 USWG-4 1788.7-9.*



*Figure 2. Demonstration drift - location of Air Core Hole #1 (AC-1).*

Gravimetric analysis of the samples indicated that the total airborne dust concentrations for AC-1 ranged from 0.27 to 3.12 mg/m<sup>3</sup> (average = 1.36 mg/m<sup>3</sup>). Those for AC-2 ranged from 0.16 to 0.5 mg/m<sup>3</sup> (average = 0.35 mg/m<sup>3</sup>), a significant decrease. (The background for both drilling sites was 0.05 mg/m<sup>3</sup>.)

A contract laboratory conducted the respirable mass and x-ray diffraction analyses of quartz, cristobalite, and tridymite. Respirable dust at AC-1 ranged from 0.32 to 2.74 mg/m<sup>3</sup> (average = 1.02 mg/m<sup>3</sup>) and at AC-2 ranged from 0.17 to 0.43 mg/m<sup>3</sup> (average = 0.27 mg/m<sup>3</sup>). Once again, dust was reduced at AC-2 compared with AC-1.

No cristobalite or tridymite was detected on any sample, and only one sample contained a measurable amount of quartz (0.04 mg). The laboratory reported quartz on four other samples, but the quantity was below the lower limit of detection and quantification of the analytical instrumentation. Because of the short durations of the drilling operations, the calculated quartz time-weighted averages (TWAs) were below the ACGIH threshold limit value (TLV/TWA) for quartz (0.1 mg/m<sup>3</sup>). If these operations were performed for a full 8 h at the observed dust levels, the allowable 8-h TLVs could possibly be exceeded. Silica analysis of one bulk sample each from AC-1 and AC-2 indicated 6.8% quartz detected for the AC-1 sample and 11.0% for the AC-2 sample. (No cristobalite or tridymite was detected in these samples.)

Phase-contrast microscopic analysis of samples indicated the presence of very few fibers. No zeolite fibers were detected; however CaSO<sub>4</sub> fibers were detected. Sulfaset, a plaster-of-paris (CaSO<sub>4</sub>)-based packing mud was used to stabilize the collar/spider to the Laser Drift face. The collection/detection of these fibers (not considered a health hazard because of the low concentration) demonstrates that the monitoring/analytical techniques used can detect fibrous particles at very low concentrations (<0.05 fiber per cubic centimeters).

Photon-induced x-ray emission (PIXE) analysis determined the elemental content of the two bulk samples previously discussed and an

additional 10 samples. Comparison of the elemental analysis with published whole rock chemical analysis showed a similar distribution of elements. When the trace element compositions were compared with ACGIH TLVs for these elements, we concluded that no trace element is present that could pose a health hazard to area personnel.

The membrane filter air samples were examined by SEM/EDX. All particles observed were either nonfibrous particles or loosely bound agglomerates of nonfibrous particles; no fibers were detected. The bulk samples were also examined by SEM/EDX, and again no fibers were detected.

Andersen cascade impactors were used for the collection and quantification of the particle size distribution of the airborne material and total dust concentration in the range of 0.4-10 µm aerodynamic diameter. The particle size distribution at AC-1 ranged from 1.9 to 3.8 µm Mass Median Aerodynamic Diameter (MMAD) with a standard deviation (SD) of 2.5-3.8. The gross concentration average was 3.37 mg/m<sup>3</sup>. The particle size distribution for AC-2 ranged from 0.97 to 1.47 µm MMAD (SD = 6.06-9.8) with a gross concentration average of 0.57 mg/m<sup>3</sup>.

A significant number of the particles were smaller than 0.4 µm as shown by the high percentages of total amount of the aerosol collected on the final filters. A large portion of the aerosol was identified as diesel exhaust particles, normally in the 0.25- to 0.26-µm size range, which would not impact on any of the Andersen impactor stages. The AC-2 samples showed a smaller particle size than those from AC-1.

The diesel mucker (without an exhaust scrubber) operated more frequently in the vicinity of AC-2 than AC-1. The lower dust levels observed in AC-2 are attributed to the progressively implemented dust control measures designed into the dust-collection system. Approximately 90% of the gross concentration was within the respirable size range. Respiratory protection should be available in case of an emission excursion.

## Results and Conclusions for the Evaluation of the G-Tunnel Prototype Air Coring Project

1. SEM/EDX and x-ray diffraction analyses are required to make specific identification/quantification of fibrous zeolites.
2. A supplementary exhaust system is required.
3. Crystalline silica analysis detected no cristobalite or tridymite. Quartz was detected in one sample ( $0.04 \text{ mg/m}^3$ ), and four borderline quantities were within the level of detection and quantification for the analytical instrumentation. Calculated TWAs were lower than the ACGIH quartz TLV/TWA ( $0.1 \text{ mg/m}^3$ ).
4. Zeolite fibers were not detected in AC-1 and AC-2 airborne particulate samples collected during air coring. The G-Tunnel geologist confirmed that zeolite fibers have not been identified.
5. The dust-control system was effective in the configuration sampled.
6. Diesel exhaust particulates originating from vehicles working in the G-Tunnel area were detected.

## TASK 4 - RECOMMENDATIONS FOR THE EXPLORATORY SHAFT FACILITY AT YUCCA MOUNTAIN

Based on the findings, we recommended that the following be implemented in future air coring or dry drilling activities:

1. General dilution ventilation and local exhaust ventilation must be provided to the drilling site.
2. Air supplied to the drill hole must be balanced at all times with the exhaust capacity of dust-collection equipment.
3. Area employees (workers and investigators) must be fitted, trained, and provided with air-purifying respirators for high-emission situations.
4. Dust-control equipment should be operated and properly maintained during any air or dry drilling activity.
5. All potential emission sources should be identified before drilling, and emission control strategies should be developed.
6. Vehicle exhaust emissions should be kept as low as reasonably achievable.
7. Safe Operation Procedures should be developed and implemented.
8. Drilling operations should adhere to existing health and safety requirements applicable to the site.
9. The potential hazard (based on geologic mapping) to the personnel working in the Yucca Mountain area mandates that similar sampling and analysis for fibrous zeolites, especially mordenite, be performed during drilling at Yucca Mountain to determine the hazards and appropriate protection for the area personnel.
10. Animal inhalation studies with mordenite (one of the primary zeolites in the Yucca Mountain tuffs) should be conducted to determine the biological effects of this fibrous zeolite.

## **RESPIRATOR STUDIES FOR THE DEPARTMENT OF ENERGY**

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*Group: Industrial Hygiene, HSE-5*

*Funding Organization: Department of Energy,  
Quality Verification Division*

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This program helps to insure that DOE contractor personnel are adequately protected by personal respiratory protection equipment. Areas of support for this program include:

1. Acceptance testing of special respiratory protective equipment - devices that cannot be approved by the existing National Institute for Occupational Safety and Health (NIOSH) and Mine Safety and Health Administration (MSHA).
2. Technical consultation to DOE and DOE contractors.
3. Special short-term respirator studies as directed by the DOE Quality Verification Division (QVD).

### **ACCEPTANCE TESTING OF UNAPPROVED DEVICES**

Work with the Monsanto Research Corporation (Mound Facility) has continued. Mound requested assistance to prepare and submit a request for the acceptance of their supplied-air suit. The schedule of events are as follows:

1. Mound submitted its request to the local DOE field office.
2. The local DOE field office submitted the request to DOE/QVD.
3. DOE/QVD levied a task on the Industrial Hygiene Group at Los Alamos to conduct the scheduled testing.

Los Alamos and Mound personnel completed these steps in early FY 1988. Work then began in conjunction with Mound to ensure that the testing would be comparable to the way the equipment is used in operational conditions at Mound. Mound wrote a Standard Operating Procedure (SOP) with help from Los Alamos personnel. The first draft was received at Los Alamos in June 1988. After review, we determined that it needed revisions to make it more understandable to the worker and acceptable to the Respirator Advisory Committee (RAC).

We visited the Mound plant in August 1988 to more accurately make these revisions and ensure that the SOP would still fulfill the requirements of Mound operations. During this visit, Mound personnel demonstrated the donning procedures, operational use, and contaminated suit removal procedures. A revised draft of the SOP was made. This draft was then incorporated into the complete SOP and reviewed by Los Alamos and Mound personnel during and after these demonstrations.

Members of Los Alamos' Industrial Hygiene Group requested shipment of the entire supplied-air suit and all of its related equipment (air distribution systems, supplied-air hoses and fittings, hard hats, gloves, booties, and so forth). Other requirements delayed the actual testing until late December 1988; it will be completed in early 1989. Testing completed to date indicates that this is an excellent device that will provide comfort and respiratory and full-body protection to the workers at Mound.

Sandia National Laboratories at Livermore (SNLL) requested acceptance of the Savannah-River Plant's supplied-air suit for use in their plant. SNLL stated that they will use the same SOP, all related equipment, air flows, and safety procedures that are used at the Savannah River Plant. The DOE RAC charter allows for this type of technology transfer; however, the RAC Chairman must review the requesting organization's SOP to insure that there are no changes. No additional testing is required unless requested by the RAC Chairman.

SNLL was in compliance with the DOE/RAC charter, so the chairman recommended that DOE/QVD grant SNLL approval to use the Savannah River Plant's supplied-air suit in their plant.

## SPECIAL STUDIES

Sandia National Laboratories at Albuquerque (SNLA) submitted a request for the evaluation of eight respirators from the American Optical Company. This request was approved by the DOE/QVD. SNLA incorporates these devices into their inventory and requested independent testing to get data for their records. This testing began in November 1987 and was completed in February 1988. The results of this testing were provided to SNLA by telephone and in a written report.

The respirators tested consisted of two full-face and six half-mask. Of the two full-face respirators (same size), one was made of natural rubber, the other of silicon rubber. The half-masks came in three sizes, small, medium, and large. One of each size was natural rubber and one of silicon rubber.

### Test Subjects

Initially, 10 test subjects were chosen for this program (7 male and 3 female), but 2 female subjects dropped out of the program before enough data could be obtained. The subjects were chosen not to fit an anthropometric panel but to allow all sizes of the test respirators to be used. The subjects ranged in age from 21 to 56 years.

### Exercises

The exercises used in this experiment were 7 of 9 suggested by NIOSH for respirator certification testing.<sup>1</sup> The exercises lasted for 1 min each and were used in the following order:

1. Normal breathing
2. Deep breathing
3. Side-to-side head movement
4. Up and down head movement
5. Talk (reading the rainbow passage)

6. Frowning (full-face)/smiling (half-mask)
7. Normal breathing.

### Protocol

The test subjects all had backgrounds in the field of respiratory protection, but their jobs did not require them to use a respirator extensively. Although subjects were not chosen on the basis of anthropometric measurements, the total test population roughly covered the facial dimension cells as described by the Los Alamos panel structure<sup>2</sup> and included at least one subject for each size of respirator tested.

### Equipment Used

Relative aerosol concentrations were measured with a LANL forward light-scattering photometer to give a direct real-time measurement.

The instrument was calibrated before each test to provide accurate 0% and 100% settings. A strip chart recorded the average per cent penetration of each exercise. A printout for each test indicated the per cent penetration for seven exercises, the resulting fit factor, and a graph of the per cent penetration during each exercise.

### Observations

The subjects were briefed on the testing procedures. Each subject was tested wearing all mask sizes and materials. These donnings and tests were observed by at least two test facility workers.

The following observations were noted during the testing:

1. These respirators are designed with a stiff shell to the facepiece. This design makes the facepiece feel very secure and comfortable on the face, but it makes the sizing of the device more critical since it does not mold very well to unusual face structures.
2. The head-harness buckles, on the half-mask, cut into the back of the wearer's neck and need to be redesigned to prevent this. This is a major problem because the respirator soon becomes so uncomfortable to wear that

something must be placed between the buckle and the back of the neck, which could affect the protection provided.

3. As previously stated, the full-face respirators are made of natural rubber and silicon rubber. They are exactly the same color, however, with no obvious means to tell them apart, which creates a problem for inventory and device identification.
4. The exhalation valve covers on the half-masks are very poorly mounted and tend to be dislodged with the slightest contact of the hand or other object. This leaves the exhalation valve with no clean air chamber and exposed to foreign objects, that could cause damage or leakage.

### Conclusions

The test data show that if the AO half-mask is sized properly and donned according to the manufacturer's directions, most subjects can get an adequate fit (Table I). There is also some indication that the rubber facepiece gives better protection than the silicone facepiece. Two subjects (numbers 5 and 6) have participated in several respirator studies at Los Alamos in the past. As a rule, both are very difficult to fit with a half-mask (see fit factors in Table I). The data for these tests show that these masks are still not able to give subjects 5 and 6 as high a fit factor as the other subjects, indicating that the data are normal.

The data also show that the AO full-face respirator (Table II) gives the general population very good fit factors. These data are listed as inside the nose cup and outside the nose cup to indicate the location where the sample was taken. Data for inside the nose cup are taken directly in the nose-mouth breathing zone; data for outside the nose cup are taken from a probe located through the mask lens between the eyes. The data most applicable depend on whether there is an eye or respiratory threat. Again, these data show that the rubber facepiece provides higher fit factors than the silicone facepiece.

### Respirator Advisory Committee

The tenth annual meeting (1988) of the RAC has not been scheduled because of the delay in the testing of the Mound supplied-air suit.

In early 1988, the committee members reviewed the reports containing the retesting data for the Savannah River Plant supplied-air suits. The result of this review was the approval of all devices concerned. Action was taken by the RAC members and the Secretary to change the format of the reports for supplied-air suits. These changes will make the reports easier to understand by persons that are not familiar with the workings of the RAC and its suit testing procedures.

### TECHNICAL CONSULTATION

Consultations were conducted with several contractors, field office personnel, and DOE/QVD staff. These consultations involved such subjects as the selection of respirators, maintenance, training programs, methods of determining respirator fit, committees for the advancement of test methods, and so forth. Consultations in 1988 included the following:

1. A site visit to the Mound Facility to discuss the new supplied-air device that was submitted for acceptance.
2. Telephone consultations with SNLL to obtain the acceptance of the Savannah River Plant's supplied-air suit for their use.
3. Telephone consultations with Sandia National Laboratories at Albuquerque for the evaluation of respirators to be included in their inventory.
4. Telephone consultations with Oak Ridge National Laboratory about the modification of the Savannah River Plant's supplied-air suit for use at Oak Ridge).
5. A continuation of representing DOE interests on the ANSI Z88.2 and Z88.10 Consensus Standards Committees.

6. Participation in meetings having special interests in respiratory protection, such as the ASTM F-23-50 Protective Clothing Committee.

## REFERENCES

1. NIOSH Revision of Tests and Requirements for Certification of Permeability of Respirator Protective Devices Used in Mines and Mining. 42 CFR Part 84.232F.
2. "Selection of Respirator Test Panels Representative of U.S. Adult Facial Sizes," Los Alamos National Laboratory report LA-5488 Alan Hack (March 1974).

Table I. American Optical Half-Mask Respirators.				
Subject	Material	Large Fit Factor	Medium Fit Factor	Small Fit Factor
1	Rubber	>20K	11K	b
1	Silicon	>20K	>20K	b
2	Rubber	11K	1.8K	b
2	Silicon	>20K	70	b
3	Rubber	>20K	1K	b
3	Silicon	3K	2.5K	b
4	Rubber	6K	3.8K	b
4	Silicon	700	1.4K	b
5	Rubber	150	55	b
5	Silicon	88	39	b
6	Rubber	136	55	200
6	Silicon	38	<sup>a</sup>	200
7	Rubber	38	550	b
7	Silicon	118	1.2K	b
8	Rubber	>20K	85	b
8	Silicon	750	225	b

<sup>a</sup>Test aborted due to high penetration.  
<sup>b</sup>No test was run with this size mask.

**Table II. American Optical Full-Face Respirators.**

<b>Subject</b>	<b>Material</b>	<b>Fit Factor Inside Nose Cup<sup>a</sup></b>	<b>Fit Factor Outside Nose Cup<sup>b</sup></b>
1	Rubber	11.0K	12.5K
1	Silicon	100	32
2	Rubber	16.6K	5.5K
2	Silicon	130	25
3	Rubber	11.0K	>20K
3	Silicon	16.6K	11.0K
4	Rubber	50	37
4	Silicon	90	40
5	Rubber	3.3K	2.6K
5	Silicon	63	28
6	Rubber	7.0K	7.7K
6	Silicon	100	36
7	Rubber	16.0K	>20K
7	Silicon	100	27
8	Rubber	4.7K	5.5K
8	Silicon	172	28

<sup>a</sup>Inside Nose Cup = Sample taken from inside nose cup between nose and mouth.

<sup>b</sup>Outside Nose Cup = Sample taken from inside facepiece between the eyes.

## ***HIGH FLOW ALTERNATIVE FILTER TEST SYSTEM (HFATS)***

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*Group: Industrial Hygiene, HSE-5*

*Funding Organization: U.S. Army, Product  
Assurance Directorate*

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This program will provide the Army's Product Assurance Directorate (PAD) with a modern filter penetration measuring system for testing high efficiency particulate air (HEPA) filters. Filters with rated flows from 500 to 2000 cubic feet per minute (CFM) will be tested. The test system to be supplied is called a High Flow Alternative Filter Test System (HFATS). This system was designed and evaluated in a 5-year program at Los Alamos for the U.S. Department of Energy. The system uses di-(2-ethyhexyl) phthalate (DEHP) as the challenge aerosol material. HFATS has the potential to be converted for use with other test aerosol materials.

### **CONTRACT NEGOTIATIONS**

In FY 1987, a bid package was sent to five manufacturers. Of the five, only three responded with a bid. After the bids were received, it was discovered that funding would need to be increased. PAD was contacted, and the process of getting additional funding was begun. During the 6 months needed to get the increase, the time limits for the bids elapsed. Notification of the receipt of the funding was received in January 1988. The bid packages were again sent to the prospective manufacturers for reconfirmation of the bids. The packages were returned with only two of the previous three bidders responding in the positive. In March 1988, the contract was let to Chamberlin Manufacturing, MCI plant in Baltimore, Maryland.

### **SPECIFICATIONS CONSULTATIONS**

There have been several consultations on the details of the specification package. These have included a visit to Chamberlin, MCI by Ron Scripsick of Los Alamos and Jeff Taylor of PAD, Edgewood, Maryland. Discussions included the need for the following:

1. A speed controller for the main system blower.
2. The efficiency of the inlet filters for the blower.
3. The aerosol induction system.
4. The type of flow measurement instrument required to meet the specifications.
5. The time frame in which each of the phases of the contract should be accomplished.

Jeff Taylor visited Los Alamos to further discuss the specifications package and those exceptions that were being taken by MCI. The outcome of this visit was that MCI had accepted a contract to do the following:

1. Design the system.
2. Provide Los Alamos with preliminary drawings and calculations.
3. Build the HFATS system.
4. Provide a period of examination and evaluation of the system at their facility.
5. Deliver the system and set it up at PAD.
6. Provide the PAD with operating manuals and final drawings.

This means that if a blower speed controller, more efficient filters, or any other equipment is required to meet the specifications as outlined in the bid package, responsibility for the use of, or the purchase of, such items should be with the Chamberlin, MCI Company.

#### **PRELIMINARY DRAWINGS AND CALCULATIONS**

The preliminary drawings and calculations were delivered to Los Alamos in late November 1988, ahead of the planned schedule of events.

After receipt of the package, Los Alamos requested more data on the speed controller and the filters that MCI planned to use. This was received within a week. After review of this package, several exceptions, or questions, remained. Again, most of these exceptions were on the same items listed in 1 through 5 in specification consultations. A package was prepared and sent to MCI in December 1988 stating the exceptions and the opinion of the Los Alamos Research and Development Section.

# **SORBENT TESTING AND DEVELOPMENT OF MODELS FOR PREDICTING PERFORMANCE OF CARBON BEDS**

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*Groups: Industrial Hygiene, HSE-5; Statistics, A-1*  
*Funding Organizations: U.S. Department of  
Energy, U.S. Department of Defense*

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## **INTRODUCTION**

This ongoing program continues to (1) provide testing of respirator sorbents against selected gases at selected conditions, while (2) developing methods to predict carbon bed efficiencies and lifetimes without the necessity of such testing. The time dependence of sorbent bed efficiency is usually represented as a breakthrough curve, increasing in vapor penetration as the bed becomes saturated. The service life is the time required for bed penetration to reach an unacceptable level. Figure 1 shows the categories of information needed to describe breakthrough curves and to predict bed efficiencies and service lives. This program is addressing each of these categories so that, ultimately, known or easily measured or calculated properties of a carbon bed and an adsorbate will allow such predictions.

We are developing an extensive database using experimental measurements of breakthrough curves for many gases and vapors under a wide variety of conditions. The experimental systems, initial testing conditions, and initial set of compounds (hydrocarbons and fluorocarbons) have been described in previous reports.<sup>1,2</sup> Resulting breakthrough curves were fit to semiempirical equations to average the data, to allow interpolations and extrapolations of breakthrough times, and to separate kinetic and capacity parameters (Fig. 1).

Successful correlations have been found for (1) breakthrough times at different penetration fractions for the same curve, (2) capacities

calculated from curve midpoints as a function of boiling points, and (3) capacities and rate coefficients as functions of the number of carbon and fluorine atoms.<sup>2</sup> Correlative models of the effects of relative humidity and vapor concentration on capacities have also been developed using published sources of carbon bed breakthrough data.<sup>3,4</sup>

## **DATABASE DEVELOPMENT AND ANALYSIS**

The database was extended during 1988. It now includes data from over 300 experiments with various combinations of 30 challenge compounds, 2 charcoals, 6 challenge concentrations, 7 relative humidities, 6 bed depths, and 2 flow rates. In particular, a matrix of conditions (two bed depths, two vapor concentrations, two relative humidities, and two carbons) for butane and perfluoropropene was used to verify the significant and independent effects of these parameters. The statistical analysis of these results has not yet been completed.

Additional breakthrough curves were determined on ASC carbon at dry conditions to test the applicability of correlations found with hydrocarbons and fluorocarbons. Five compounds, each from a different chemical class but with similar boiling points, were selected (Table I). Only vapor concentration was varied, 340-1360 ppm. Table I lists average 50% breakthrough times for 1360 ppm, from which approximate bed capacities can be calculated. The equations developed previously for estimating one breakthrough time from another (above 10%) were also successful for these five compounds. However, values of  $\alpha$  (estimated breakthrough times) calculated from the boiling correlations<sup>2</sup> worked only for the diethyl ether data (Fig. 2). Not surprisingly, correlations of  $\alpha$  and Langmuir isotherm parameters with number of carbon atoms<sup>2</sup> also did not hold up for the different classes of compounds. More fundamental determining parameters, such as vapor properties, are being investigated.

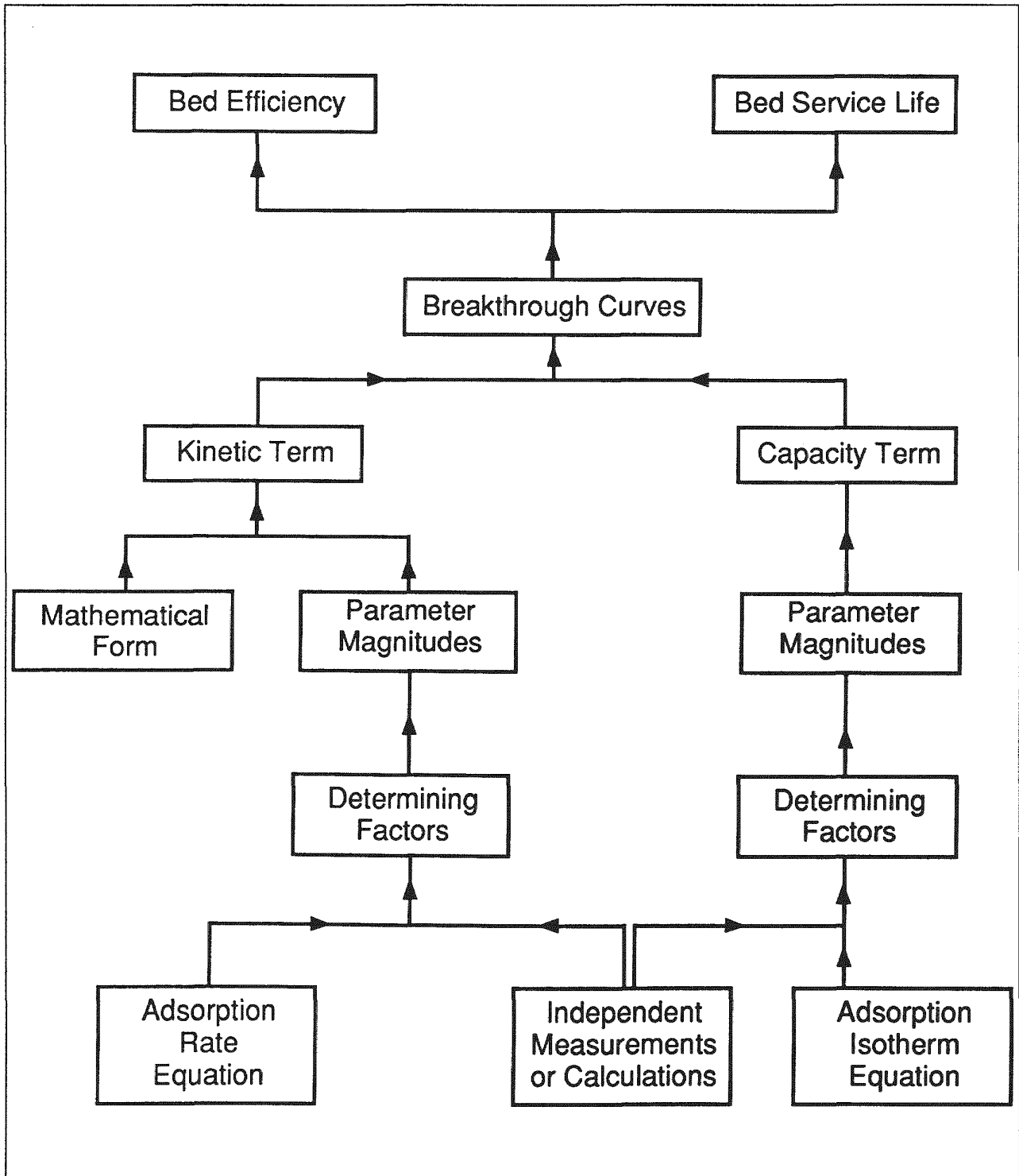


Figure 1. Sorbent testing and modeling program scope.

Table I. Capacity and Kinetic Parameters.

Compound	Boiling Point (K)	Number of Carbons	Breakthrough Time, $\alpha$ , at 1350 ppm (min)	Average Rate Coefficient ( $\text{min}^{-1}$ )
Acetone	329	3	74	2690
Chloroform	334	1	85	2290
Diethyl Ether	308	4	66	2910
Ethyl Acetate	350	3	101	2550
Benzene	353	6	161	3560

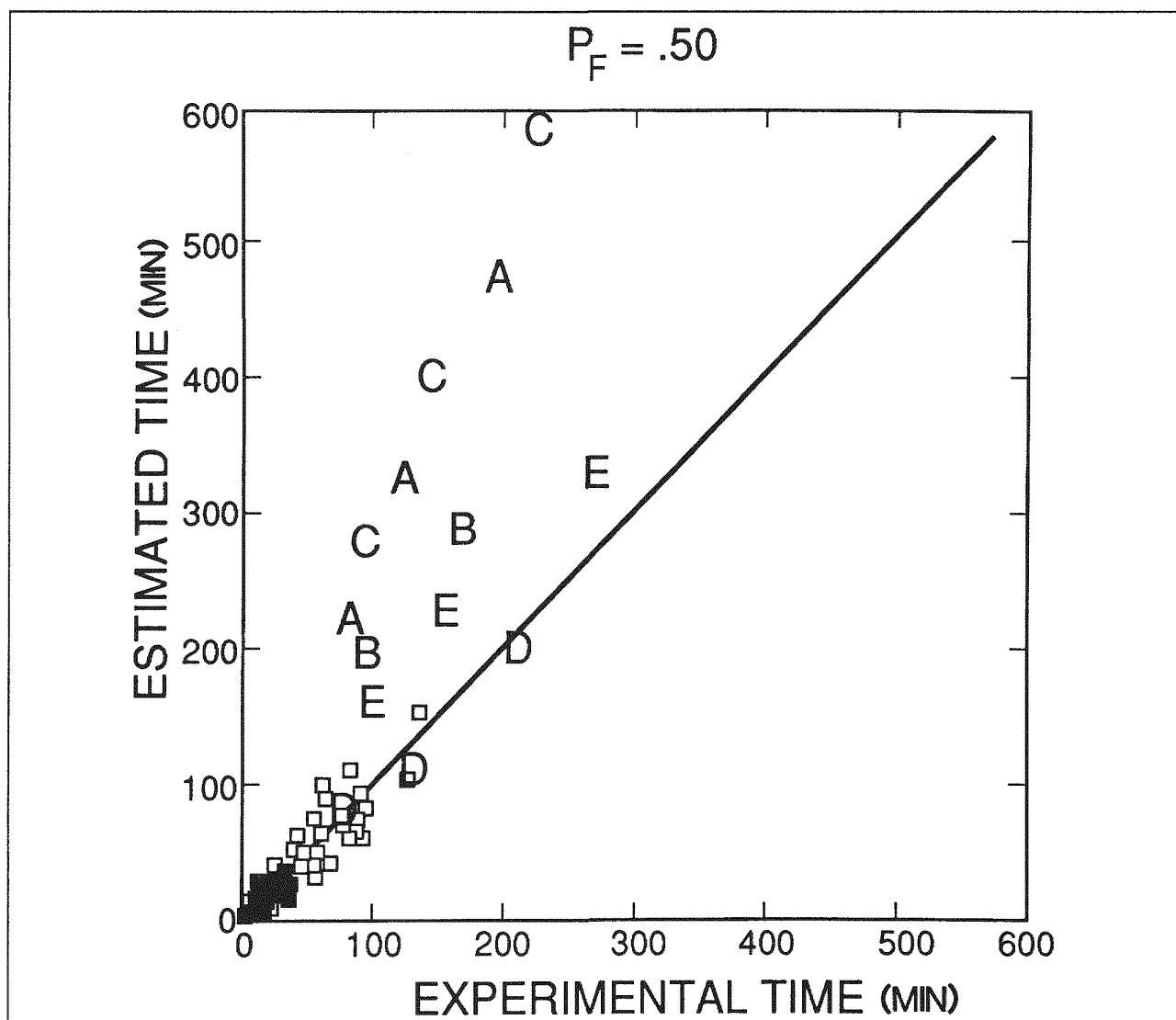


Figure 2. Comparison of experimental 50% breakthrough times with those calculated from correlations with boiling points. A = acetone, B = benzene, C = chloroform, D = diethyl ether, E = ethyl acetate, and squares = hydrocarbons and fluorocarbons.

Table I also lists average rate coefficients from fitting the breakthrough curves to the ideal Wheeler equation. As seen before, these did not appear to vary with vapor concentrations 340-1360 ppm. No trends among compounds have yet been discovered.

## RELATIVE HUMIDITY EFFECTS

Previously most of the experiments for the database were done with dry ASC carbon and dry air (about 3% relative humidity). The reason for this was to establish a fundamental understanding of the adsorption rate and equilibrium process for a variety of compounds. However, in the real world water vapor is present in air at various relative humidities. The significant influence of water vapor has been illustrated and modeled for various organic compounds on unimpregnated carbons.<sup>3</sup>

More recently this program has addressed effects of relative humidity on breakthroughs of some of the compounds previously studied dry (Table II). The test beds were preconditioned at the test relative humidity.

Another significant experimental change was to use an ASC carbon also impregnated with triethylenediamine, which will be called ASC-T. Bed depths of 2, 4, and 6 cm (for 2.3 cm diam) and airflow velocities of 740 and 370 cm/min were selected for the testing matrix.

The effects of relative humidity on breakthrough curves were quite pronounced, shifting them to shorter times and changing their shapes. Capacities calculated at 1360 ppm from 50% breakthrough times (Table II) decreased drastically, as expected.<sup>3</sup> More surprisingly, the rate coefficients, calculated from the differences in 10% and 50% breakthrough times, also decreased drastically with increasing relative humidity. The reason for this is not obvious and will be explored further.

## APPLICATIONS OF THE WHEELER EQUATION

Our modified Wheeler equation was studied using an extensive database of acetone breakthrough curves for several brands and many organic vapor respirator cartridges. This database was developed by Dr. Ernest Moyer of the National Institute for Occupational Safety and Health in Morgantown, West Virginia. The data

were analyzed using three applications of the Wheeler equation: (1) varying bed weight, (2) varying bed residence time, and (3) breakthrough curve fitting.

The results of this study are summarized in Table III. When the adsorption capacity values are compared, there is excellent agreement among the three methods of applying the Wheeler equation. Neither airflow rate or bed weight had any significant effects. The adsorption rate coefficients (Table III), however, differed with the various methods and was influenced by flow rate. The effect of airflow rate on the rate coefficient was approximately proportional to the square root, as would be expected for external mass transfer being the rate-limiting step.

One conclusion of this study is that the curve-fitting application of the Wheeler equation that we have been using is as valid as any other application of it. However, it must be used with caution since not all breakthrough curves fit this ideal equation. The kinetic parameters (rate coefficients) obtained must be recognized as empirical and dependent on the selected application and breakthrough curve equation. Until it can be better defined, such a rate coefficient has little usefulness for extrapolating to other situations and conditions.

## LOW-PENETRATION DATA FITTING

Breakthrough curves for a wide variety of compounds at dry conditions have been reasonably well fit by the modified Wheeler equation for penetrations at and above 10%. However, these fits often fail to provide good estimates of the breakthrough times at 1% penetration when compared with experimental times (Fig. 3). Deviations from the equivalence line are in the direction of longer experimental times than calculated. This can be due to a heterogeneous surface activity, not reflected in the Wheeler equation, that is based on a homogeneous surface assumption. Or it could be due to a change in mechanism, such as from surface adsorption at low loadings to pore condensation at high loadings. In either case, a better fit of breakthrough curves is needed at the lower breakthrough fractions, which are often the ones of most interest. Work on such an improved fit equation has begun.

Compound	Relative Humidity (%)	Average Rate Coefficient <sup>a</sup> (min) <sup>-1</sup>	Average Capacities <sup>b</sup> (g/g <sub>c</sub> )
Butane	3	2640	0.062
	50	1210	0.016
	80	540	0.003
Perfluoropropene	3	1560	0.066
	50	540	0.012
	80	-	0.001
Perfluorobutene-2	50	1860	0.031
	80	--	0.002
Perfluorobutane	50	--	0.031
	80	--	0.000
Trifluoromethylpropene	50	2100	0.037
	80	1740	0.007

<sup>a</sup>At 740 cm/min airflow velocity.  
<sup>b</sup>Calculated from 50% breakthrough times for 1360 ppm for ASC-T carbon.

Average Airflow Rate (L/min)	Adsorption Capacity, W <sub>e</sub> (g/g)			Adsorption Rate Coefficient, K <sub>v</sub> (min <sup>-1</sup> )		
	Bed Weight Variation	Residence Time Variation	Breakthrough Curve Fitting	Bed Weight Variation	Residence Time Variation	Breakthrough Curve Fitting
32.2	0.106	0.106	0.092	4730	5050	6580
40.4	0.110	0.108	0.107	4880	5920	6870
63.9	0.116	0.116	0.114	7940	7860	9160
83.0	0.115	0.113	0.109	7440	7860	10410
94.5	0.113	0.113	0.108	8670	8770	13090
112.5	0.109	0.110	0.107	9460	8730	12780
Averaged	0.112	0.111	0.108			

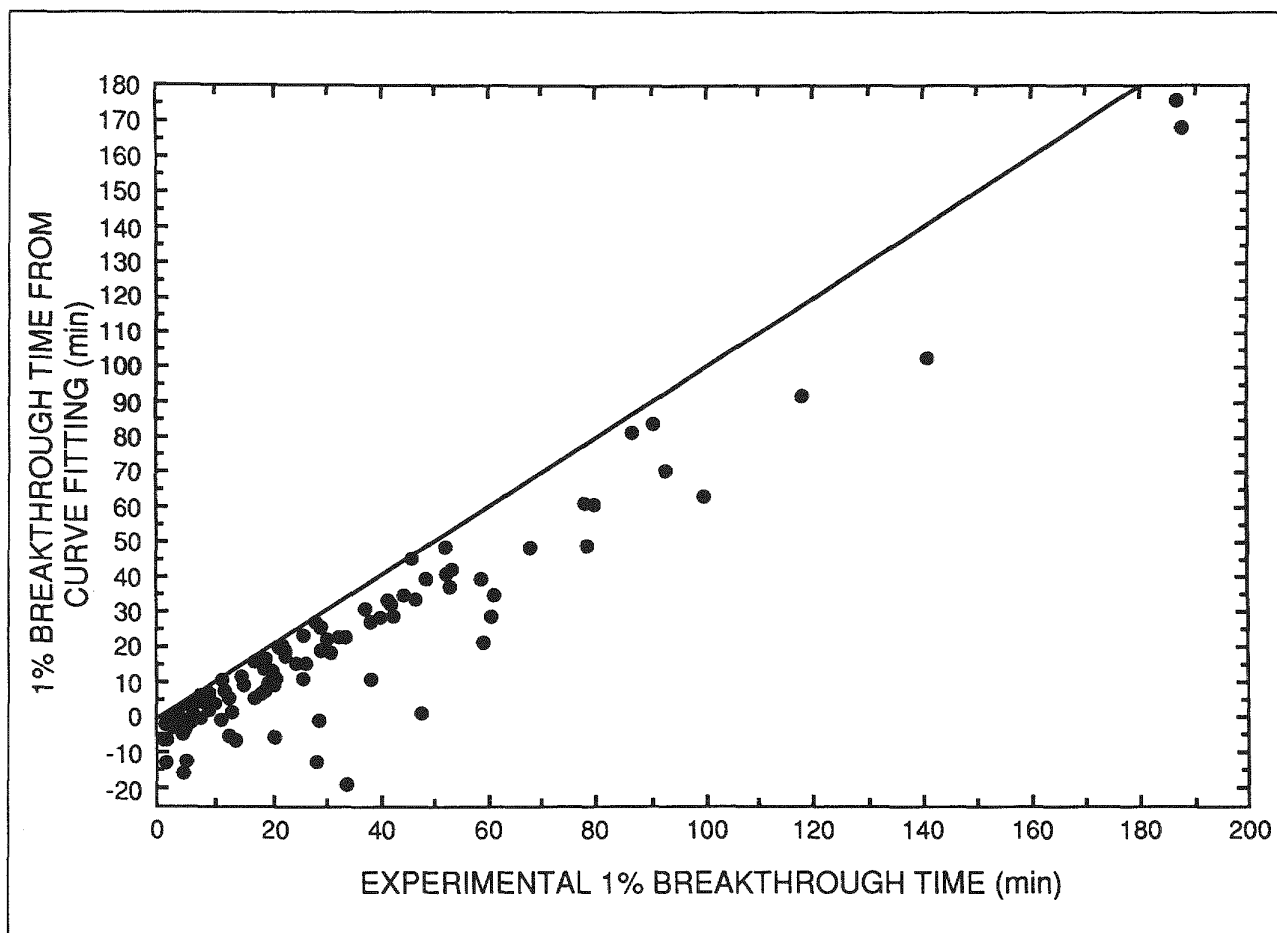


Figure 3. Comparison of experimental 1% breakthrough times with those calculated from fitting the ideal Wheeler equation to the entire breakthrough curve.

## ADSORPTION ISOTHERM EQUATIONS

Carbon capacity as a function of vapor concentration is called an adsorption isotherm. Many equations have been proposed to describe observed isotherms. Some of these have been used to correlate bed breakthrough times and parameters obtained from fits of the Wheeler equation. In another study using data from NIOSH, four two-adjustable-parameter adsorption isotherm equations were compared: (1) Freundlich, (2) Langmuir, (3) Dubinin-Radushkevich (D/R), and (4) HacsKaylo-LeVan (H/L). Five compounds of widely ranging polarities (Table IV) were included in the database used. The objective was to determine which of these equations best describes the canister breakthrough data. A nonlinear least squares fitting was used. A goodness-of-fit parameter was calculated, based

on the sum of the squares of the deviations and the number of data points fit.

Table IV summarizes the results obtained in this study. The conclusion is that this particular set of data fit all four equations equally well, except that the fit was significantly worse for ethanol and the Freundlich equation. Therefore, there is no preference for the Langmuir, D/R, or H/L isotherm equations on the basis of data fit. Other bases for preference do exist, but they will not be discussed here.

## A TESTING PROTOCOL

Performance data are required for initial design, improvements, and evaluations of organic vapor canisters, cartridges, and other activated carbon beds and for making decisions on using

Table IV. Fits of Adsorption Isotherm Equations to Canister Testing Data.					
	Goodness of Fit Parameter				
	Ethanol	Acetone	Chloroform	Carbon Tet	Hexane
<u>Equation</u>					
Freundlich	0.025	0.049	0.051	0.053	0.102
Langmuir	0.016	0.047	0.053	0.053	0.102
Dubinin/ Radushkevich	0.015	0.048	0.052	0.053	0.102
HacsKaylo/ LeVan	0.015	0.048	0.053	0.053	0.102

them in the field. A generic test plan has been developed for laboratory measurements of such performance against any selected vapor. It assumes that the test system is set up, the test beds are prepared, and the capability for interpreting the resulting data exists. Objectives in designing this protocol have been to (1) optimize the number of tests, (2) optimize the amount of information available, (3) have the results in a form that can be correlated by any one of a variety of mathematical models, (4) define data reproducibility, (5) allow scaling of results to other bed geometries for evaluation or design purposes, and (6) provide a visual representation of results for field use.

Portions of breakthrough curves are determined for a reference set of conditions and for ranges of bed depths, airflow rates, vapor concentrations, relative humidities, and temperatures. Twenty-five tests have been identified as the optimum number required to quantify effects of all the above parameters. A flow chart summarizing the specific recommendations for the initial phase of testing (nine tests) is shown in Fig. 4.

A two-level factorial experimental design is proposed for testing effects of three environmental conditions (vapor concentration, relative humidity, and temperature) on breakthrough times. Two levels of each parameter in all combinations form the corners of a cube when plotted on the three axes of a Cartesian coordinate system (Fig. 5, for

example). Values of the dependent variable (breakthrough time or service life) are determined at the corners and repeated randomly (16 tests). This method allows summary of service life test results for the selected testing conditions in a cube representation (Fig. 5). The numbers shown for service lives in the circles within the cube (reference conditions) and at the corners (combinations of two levels for three parameters) are for illustration only.

#### SUMMARY

In conclusion, significant progress is being made toward understanding the factors that determine carbon bed capacities and service lives. The efforts reported here are being supplemented by work elsewhere at Los Alamos to experimentally measure and theoretically model activated carbon and its interactions with adsorbates.

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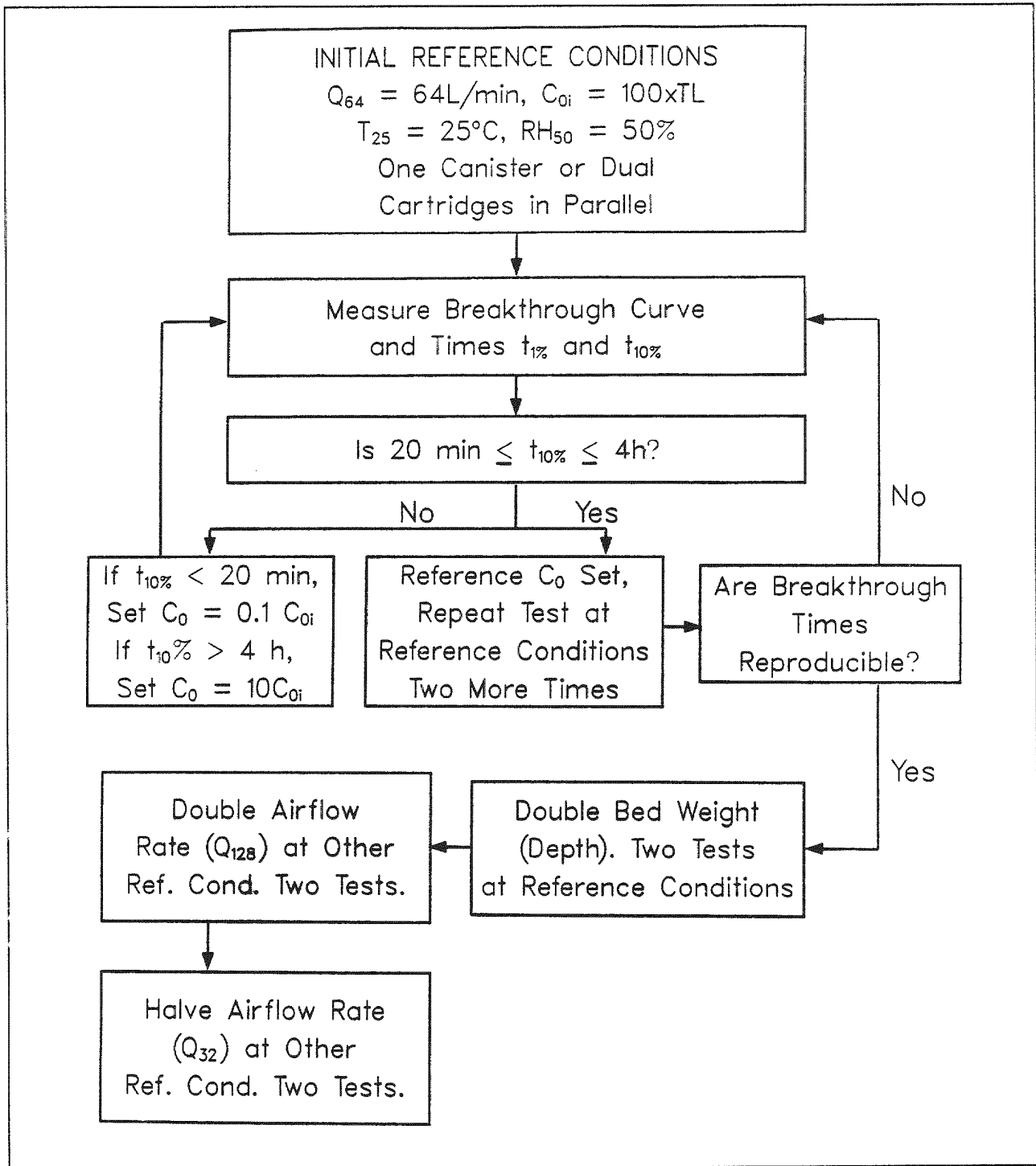


Figure 4. A testing protocol for organic vapor respirator canisters: Phase I.

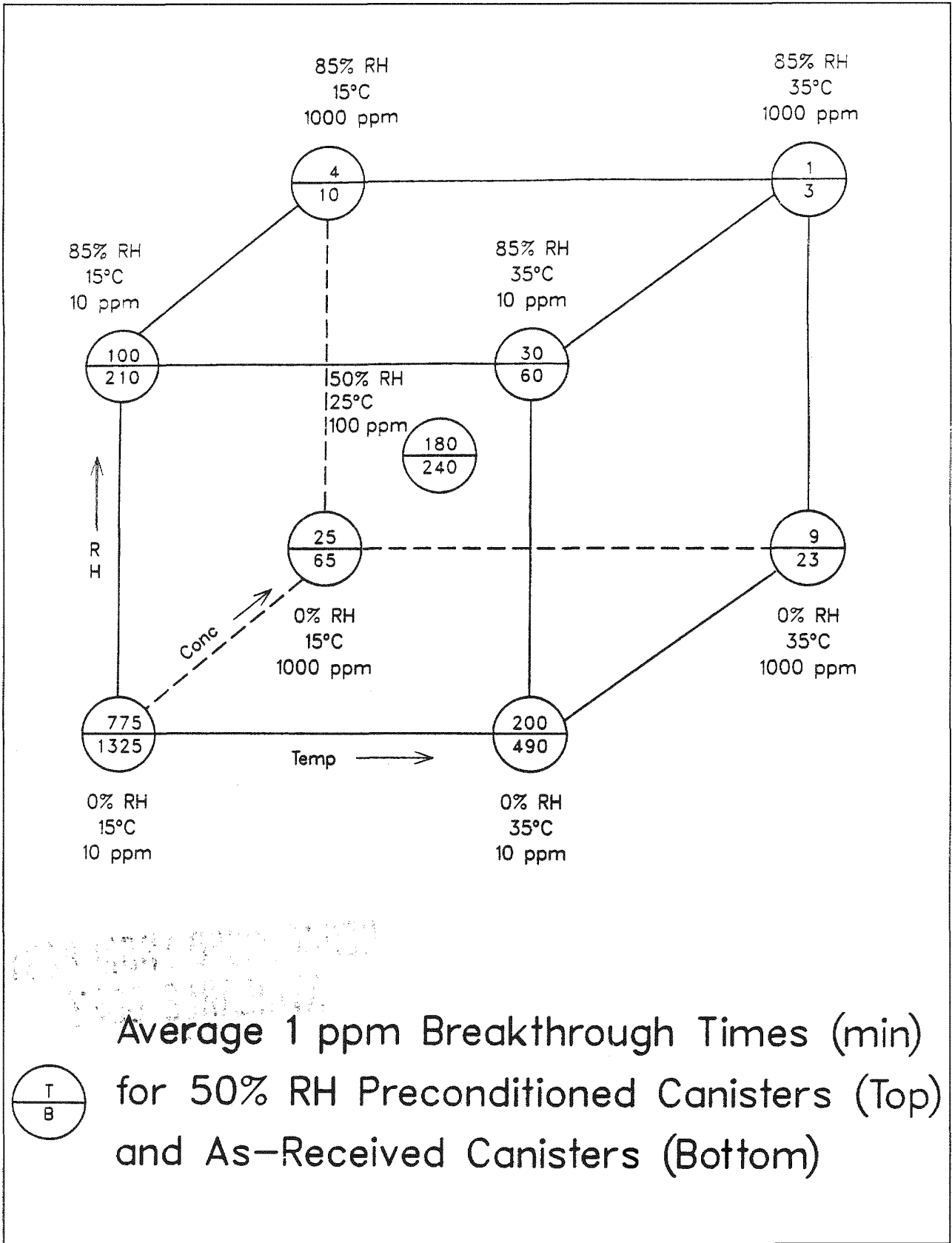


Figure 5. A sample of Phase II test matrix for environmental effects on canister service life.

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## ENVIRONMENTAL CHEMISTRY FOR THE NATIONAL PARK SERVICE

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Group: Health and Environmental Chemistry,  
HSE-9*

*Funding Organization: Air Quality Division,  
National Park Service*

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The Los Alamos National Laboratory has a continuing Interagency Agreement with the National Park Service to assess the effects of air pollution at selected national parks and monuments. Studies, conducted by the Health and Environmental Chemistry Group, are under way at Arches National Park, Bandelier National Monument, Canyonlands National Park, Cascades National Park, Chaco Culture Natural History Park, Chiricahua National Monument, Everglades National Park, Mesa Verde National Park, Petrified Forest National Park, Saguaro National Monument, and Walnut Canyon National Monument. The following are short descriptions of these studies.

1. Sulfur Dioxide Fumigation of Pinon Pine Seeds and Seedlings - These studies are aimed at determining the effects of sulfur dioxide, a common pollutant from power plants and smelters, on pinon pine, a common and ubiquitous tree species in Southwestern parks and monuments.
2. Baseline Sulfur Concentrations around Four Corners - The sulfur content of foliage and soil samples from parks and monuments around the Four Corners power plant is being determined to provide a baseline against

which to assess future impacts from power plant emissions.

3. Saguaro National Monument - Tree cores and samples of saguaro tissue and soils are being analyzed in an attempt to determine the cause of the "saguaro decline."
4. Everglades National Park - Pine needles are being analyzed to evaluate the effects of air pollution on the park.
5. Cascades National Park - Subalpine fir needles, lichens, and soil and duff samples are being analyzed to provide baseline data and to assess whether smelter emissions from the Seattle-Tacoma area are affecting the park.
6. Rock Pool Study - Water chemistry of samples from natural rock pools in Canyonlands and Arches National Parks is being characterized to determine if these pools are being affected by pollutant emissions.
7. Cryptogamic Soil Study - Samples of cryptogamic soils from about ten national parks and monuments on the Colorado Plateau will be analyzed to determine if air pollution is affecting these soils.

The first six of these studies are ongoing, although some of them are nearing completion. The cryptogamic soil study will begin in 1989.

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## MORTALITY AMONG MOUND EMPLOYEES

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*Funding Agency: DOE, Office of Health and  
Environmental Research, Human Health and  
Assessments Division*

We have completed a cohort mortality study of 4182 white males employed by the Mound Facility near Dayton, Ohio, from 1947 through 1979. Mortality did not exceed expected mortality based on U.S. death rates for the overall cohort or for the subcohort monitored for external ionizing radiation. Some increase in mortality from nonmalignant digestive diseases was observed for workers never monitored for external ionizing radiation.

Mortality for workers with cumulative radiation doses of at least 10 mSv was not significantly increased when compared with mortality for workers with cumulative radiation doses of less than 10 mSv (Table I). A significant dose-response

was observed for all leukemias (ICDA 204-207) based on two cases with cumulative external radiation doses of at least 50 mSv (5 rem). No leukemia cases were observed in the 10- to 49.9-mSv category. Further examination revealed that one of the cases in the 50+ mSv category was a chronic lymphatic leukemia, a type of leukemia generally not regarded as radiogenic. When this case was removed from the analyses, the strength of the dose-response was reduced.

In addition to the analyses reported above, preliminary analyses of mortality among Mound workers monitored for polonium internal contamination are currently under way. During this year, additional data entry of 29,036 polonium urinalysis records (607 people) was completed.

Follow-up has been extended for the Mound cohort through the end of 1983. This was accomplished through a combination of methods including a Social Security search of vital status, a submission to the Ohio Department of Motor Vehicles, and active tracing. Future analyses, including the analyses of workers monitored for polonium, will consider vital status through the end of 1983.

Table I. Maximum Likelihood Estimates of the Rate Ratio for Mound Workers Exposed to External Radiation (10 mSv or more) Compared With Unexposed Workers (<10 mSv) Using a 10-year Induction Period.				
	Observed Cases <sup>a</sup>			
	Exposed (8824 PY)	Unexposed (45326 PY)	RR	CI <sup>b</sup>
<b>Cause of Death</b>				
All Causes	62	241 <sup>c</sup>	0.96	0.75, 1.23
All Cancers	17	49	1.18	0.72, 1.93
Digestive Organs	4	10	1.26	0.34, 4.07
Lung	7	15	1.46	0.65, 3.27
Prostate	0	6	0.00	0.00, 2.40
Lymphopoietic/ Hematopoietic	3	7	1.47	0.30, 5.80

<sup>a</sup>Includes all categories, exposed or unexposed, in which 6 or more cases are recorded.  
<sup>b</sup>90% confidence index limits.  
<sup>c</sup>One death, cause unknown, omitted.

## ***A NESTED CASE-CONTROL STUDY OF SUICIDE AMONG WOMEN EMPLOYED BY THE LOS ALAMOS NATIONAL LABORATORY***

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***Funding Agency:*** DOE, Office of Health and  
Environmental Research, Human Health and  
Assessments Division

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A cohort study of women employed by the Los Alamos National Laboratory reported elevated rates of suicide among the subcohort of women monitored for external ionizing radiation. As a result, we conducted a nested case-control study to determine whether factors other than a history of radiation monitoring might also be associated with suicide mortality among members of this cohort. Persons dying from suicide (cases) were compared with two comparison groups (controls). One of the control groups consisted of persons dying from other types of injuries and the other consisted of a random sample of persons dying from causes of death not including injuries.

Compared with both control groups, a higher proportion of the suicide deaths were monitored for external ionizing radiation. This finding remained when education, a potentially confounding factor, was controlled, although the relationship was much stronger among women with a college degree. Job classification in a technical or professional job was also associated with suicide mortality. This variable is closely related to whether an individual had been monitored for radiation exposure. Note that 9 of the 19 cases were monitored for external ionizing

radiation, but only 2 of the cases had cumulative doses of at least 10 mSv (1 rem).

We observed no association between suicide mortality and a history of monitoring for plutonium internal deposition. Only 1 of the 19 suicide cases had been monitored for plutonium body burden. The low frequency of plutonium monitoring may be a result of the small number of women working with plutonium until recent years.

Additional variables were examined. Marital status, which has been related in other studies of suicide, was not associated with suicide mortality among members of this cohort. All of the suicide cases had been married at some time in their lives. At the time of death, 11 were married, 6 were divorced, and 2 were widowed.

Duration of employment was also examined. This variable was not significantly associated with suicide mortality. Furthermore, it is interesting that only two of the cases were employed by the Laboratory at the time of their deaths.

Education was also not associated with suicide mortality. Five of the cases were college graduates, of which one was a medical doctor.

From this study it is not possible to explain the strong association between suicide mortality and a history of monitoring for external ionizing radiation. This relationship is most likely tied to a yet unidentified selection factor that selects individuals for both jobs in which they are monitored for radiation exposure and an increased risk of suicide mortality.

## UPDATE ON EPIDEMIOLOGICAL DATABASES OF LOS ALAMOS EMPLOYEES

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*Group: Occupational Medicine, HSE-2*

*Funding Agency: DOE, Office of Health and  
Environmental Research, Human Health and  
Assessments Division*

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Collection of epidemiologic data on all past and present employees at Los Alamos has involved untangling many years of records for tens of thousands of persons. Especially difficult have been the records generated in the early years, during and just after World War II. The goal is to obtain a complete roster of employees, accurate personal identifiers, employment history, radiation exposure history, vital status, and if deceased, cause of death. Among the several ongoing studies at DOE facilities, the Los Alamos data have been the most difficult to collect because of the Laboratory's complex history.

Databases are maintained for the two major employers, the Laboratory and the former Zia Company. The data have been collected independently for each database, so the following synopses describe them separately.

### LOS ALAMOS NATIONAL LABORATORY DATABASE

The first study from this database, a cohort mortality analysis of 6790 females, was published last year.<sup>1,2</sup> A cohort of 16 424 males employed from 1943 through 1979 has also been identified. Analyses will focus on the 16 101 (98%) workers who are white males. In 1988, the analytic data file was compiled and preliminary data preparation completed. After some additional quality assurance checking, analysis of the male cohort will begin in 1989.

During 1988, follow-up was updated for these males to include information on vital status through the end of 1983. The follow-up rate for white males

employed at least 6 months is 95%. Death certificates have been obtained for 96.8% of the 2182 white males identified as deceased. For the subcohort of white males employed at least 6 months, death certificates have been obtained for 98.1% of those known to be deceased.

Radiation monitoring status has been determined for members of the cohort. Of the 16 101 white males, 9469 have been monitored for exposure to external ionizing radiation, 3787 have been monitored for internal deposition of plutonium, and 3221 have been monitored for both plutonium and external ionizing radiation.

### ZIA COMPANY DATABASE

Data have been collected on 14,692 Zia Company employees thus far. Work on this cohort is not as far along as for the Laboratory employees, but during the past 2 years about 5000 more names have been added and much progress has been made to determine vital status of members of this cohort.

Of this cohort, 5278 persons have been monitored for external radiation and 2802 have been monitored for plutonium. A total of 5518 persons have been monitored. Because we plan to restrict analysis initially to persons monitored for radiation exposure, priority is being given to completing the data collection on this subcohort.

Of the monitored persons, 1434 possible deaths have been identified. Death certificates have been received on 1338 persons thus far for a 93.3% ascertainment rate. Vital status is known for over 88% of the monitored subcohort through 1985. Further work is in progress to decrease the number of persons lost to follow-up. The database should be sufficiently complete by late 1989 or early 1990 to begin analyses.

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## COLLABORATIVE EPIDEMIOLOGY STUDIES

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*Group: Occupational Medicine, HSE-2*

*Funding Organization: DOE, Office of Health and Environmental Research, Human Health and Assessment Division*

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Collaborative DOE epidemiology studies of occupational radiation exposures have become increasingly important during the past 2 years. These studies require data collected by several groups of investigators for different worker populations. Combined studies increase the sensitivity and power of analyses. The three participating DOE research groups in the U.S. are Los Alamos, Oak Ridge Associated Universities (ORAU), and Hanford (Battelle Pacific Northwest Laboratory and Hanford Environmental Health Foundation). Because these collaborative efforts are recent, all studies described below are still in progress.

- o The plutonium workers health studies conducted by us include a combined analysis of data from six DOE facilities. Data on plutonium-exposed workers and control subjects from Savannah River, Oak Ridge, and Hanford will be contributed by the responsible collaborators and analyzed together with the Los Alamos studies of the Rocky Flats, Mound, and Los Alamos cohorts.
- o We have been collaborating with the ORAU group, which is conducting the "Five Rem Study." This study involves interviewing all past DOE subcontractor workers exposed to 5 rem or more in any calendar year and comparing their health experience with that of control subjects from the same facilities. The study thus far involves 2529 exposed workers and over 5000 controls. We are contributing data on Los Alamos, Rocky Flats, and Mound workers.
- o We provided data of the Rocky Flats cohort to the Hanford statistical research staff for a combined analysis of Hanford, Oak Ridge National Laboratory, and Rocky Flats workers. A draft report entitled "Analyzing Combined Data from Studies of Workers Exposed to Low Levels of Radiation" has been prepared and reviewed. An important part of this study is the evaluation of the advantages and disadvantages of methods that may be used for such analyses.
- o In June 1988, G. Voelz and L. Wiggs participated in an international workshop of epidemiologists involved in studies of radiation workers. The meeting was hosted by the International Agency for Research on Cancer (IARC) in Lyon, France. A working committee of researchers of United States, United Kingdom, and Canada was created to develop a study protocol for combining data at an international level. A first draft of the protocol was reviewed and is now being revised. Submission of data will be made to IARC beginning in 1989.
- o During 1988, planning for multisite case control studies began. A protocol prepared by the Hanford HEHF group for a case control study of multiple myeloma in radiation workers was reviewed and approved. The ORAU group also began preliminary plans for a case control study of leukemia in radiation workers. Data were prepared and sent to the investigators for both these studies.



*Environmental Surveillance*

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## ENVIRONMENTAL SURVEILLANCE AT LOS ALAMOS

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*Authors: Environmental Surveillance Group  
Group: Environmental Surveillance, HSE-8  
Funding Organization: Indirect*

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### MONITORING OPERATIONS

The Laboratory maintains an ongoing environmental surveillance program as required by U.S. Department of Energy (DOE) Orders 5480.1A ("Environmental Protection, Safety, and Health Protection Programs," August 1981) and 5484.1 ("Environmental Protection, Safety, and Health Protection Information Reporting Requirements," February 1981). The surveillance program maintains routine monitoring for radiation, radioactive materials, and chemical substances on the Laboratory site and in the surrounding region. These activities document compliance with appropriate standards, identify trends, provide information for the public, and contribute to general environmental knowledge. More detailed, supplemental environmental studies are carried out to determine the extent of the potential problems, to provide the basis for any remedial actions, and to provide further information on surrounding environments.

The monitoring program also supports the Laboratory's policy to protect the public, employees, and environment from harm that could be caused by Laboratory activities and to reduce environmental impacts to the greatest degree practicable. Environmental monitoring information complements data on specific releases, such as those from radioactive liquid-waste treatment plants and stacks at nuclear research facilities.

Monitoring and sampling locations for various types of measurements are organized into three groups: (1) Regional stations are located within the five counties surrounding Los Alamos County (Fig. 1) at distances up to 80 km (50 mi) from the Laboratory. They provide a basis for determining conditions beyond the range of potential influence from normal Laboratory operations. (2) Perimeter

stations are located within about 4 km (2.5 mi) of the Laboratory boundary, and many are in residential and community areas. They document conditions in areas regularly occupied by the public and potentially affected by Laboratory operations. (3) Onsite stations are within the Laboratory boundary, and most are in areas accessible only to employees during normal working hours. They document environmental conditions at the Laboratory, where the public has limited access.

Samples of air particulates and gases, waters, soils, sediments, and foodstuffs are routinely collected at these stations for subsequent analyses (Table I). External penetrating radiation from cosmic, terrestrial, and Laboratory sources is also measured.

Additional samples are collected and analyzed to gain information about particular events, such as major surface runoff events, nonroutine releases, or special studies. More than 25 000 analyses for chemical and radiochemical constituents were carried out for environmental surveillance during 1987. Resulting data were used for dose calculations, for comparisons with standards and background levels, and for interpretation of the relative risks associated with Laboratory operations.

### ESTIMATED DOSES AND RISKS FROM RADIATION EXPOSURE

**Radiation Doses.** Estimated individual radiation doses to the public attributable to Laboratory operations are compared with applicable standards in this report. Doses are expressed as a percentage of DOE's Radiation Protection Standard (RPS). The RPS is for doses from exposures excluding contributions from natural background, fallout, and radioactive consumer products. Estimated doses are those believed to be potential doses to individuals under realistic conditions of exposure.

Historically, estimated doses from Laboratory operations have been less than 7% of the 500-mrem/year standard that was in effect before 1985

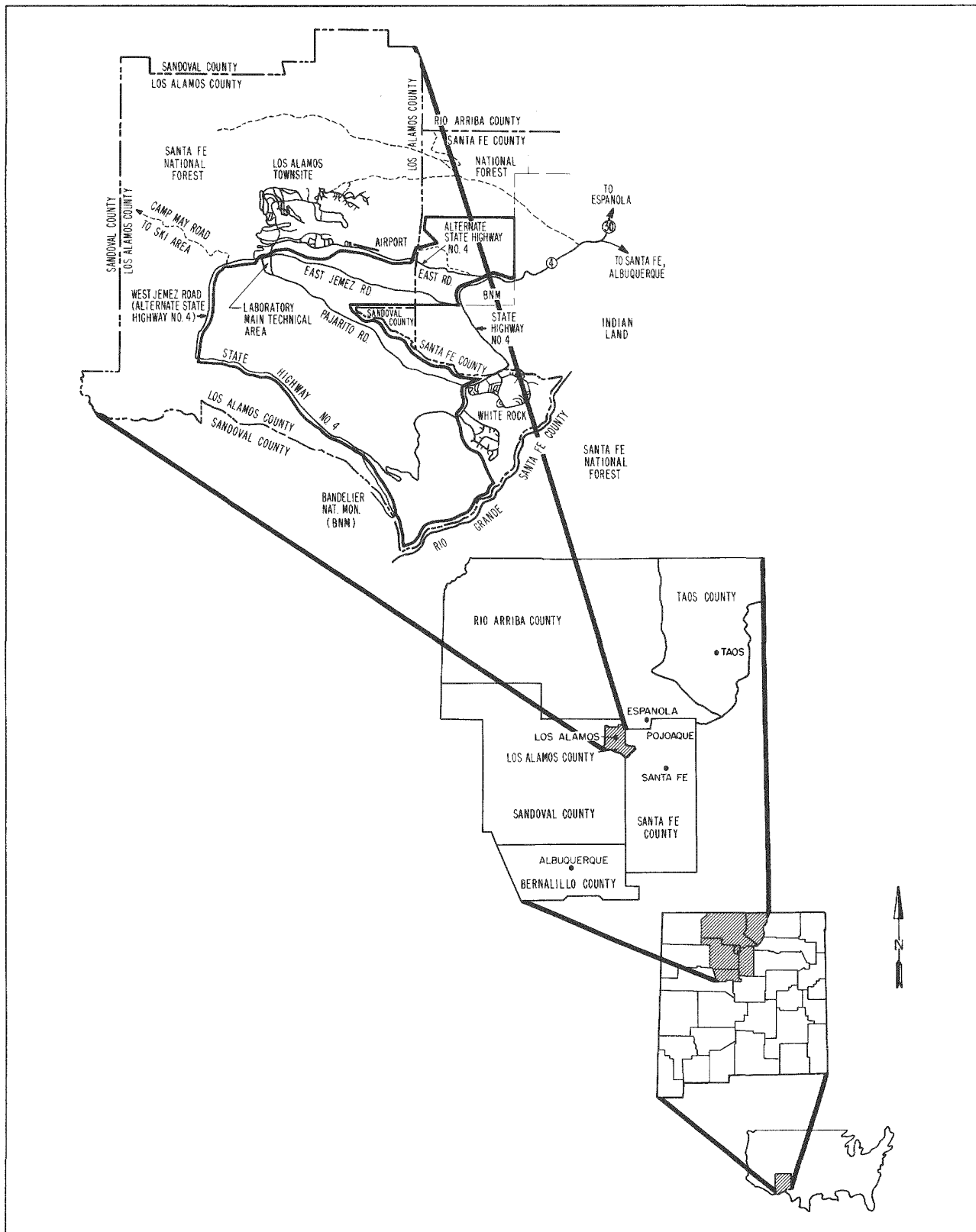


Figure 1. Regional location of Los Alamos.

Table I. Number of Sampling Locations.			
Type of Monitoring	Regional	Perimeter	Onsite
External Radiation	4	12	139
Air	3	11	12
Surface and ground waters <sup>a</sup>	6	32	37
Soils and sediments	16	16	34
Foodstuffs	10	8	11

<sup>a</sup>An additional 22 stations for the water supply and 33 special surface and ground water stations related to the Fenton Hill Geothermal Program were also sampled and analyzed as part of the monitoring program.

(Fig. 2). These doses have principally resulted from external radiation from the Laboratory's airborne releases. In 1985, DOE issued interim guidelines that lowered its RPS to 100 mrem/year (effective dose equivalent) from all exposure pathways. In addition, exposure via the air pathway is further limited to 25 mrem/year (whole body) in accordance with requirements of the U.S. Environmental Protection Agency (EPA). In 1987 the estimated maximum individual dose was 6.1 mrem, 24% of EPA's 25-mrem standard. This dose resulted mostly from external radiation from short-lived airborne emissions from a linear particle accelerator, the Los Alamos Meson Physics Facility (LAMPF).

Another perspective is gained by comparing these estimated doses with the estimated effective dose attributable to background radiation. The highest estimated dose caused from Laboratory operations was about 2% of the 327 mrem received from background radioactivity in Los Alamos during 1987.

**Risk Estimates.** Estimates of the added risk of cancer were calculated to provide a perspective for comparing the significance of radiation exposures. Incremental cancer risk to residents of Los Alamos townsite resulting from 1987 Laboratory operations was estimated to be 1 chance in 50 000 000 (Table II). This risk is <0.5% of the 1 chance in 31 000 cancer risk from natural

background radiation and the 1 chance in 190 000 risk from medical radiation.

The Laboratory's potential contribution to cancer risk is small when compared with overall cancer risks. The overall lifetime risk in the United States of contracting some form of cancer is 1 chance in 4. The lifetime risk of cancer mortality is 1 chance in 5.

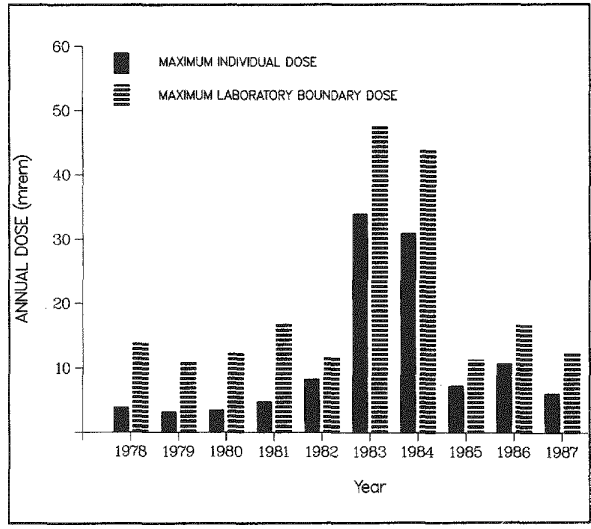


Figure 2. Summary of estimated maximum individual and maximum Laboratory boundary doses (excluding contributions from cosmic, terrestrial, and medical diagnostic sources) from Laboratory operations.

**Table II. Added Individual Lifetime Cancer Mortality Risks Attributable to 1987 Radiation Exposure.**

Exposure Source	Incremental Effective Dose Equivalent (mrem) Used in Risk Estimate	Added Risk (Chance) to an Individual of Cancer Mortality
Average exposure from Laboratory Operations		
Los Alamos Townsite	0.21	1 in 50 000 000
White Rock	0.17	1 in 60 000 000
Natural Radiation Cosmic, Terrestrial, Self-Irradiation, and Radon Exposure <sup>a</sup>		
Los Alamos and White Rock	327 <sup>a</sup>	1 in 31 000 <sup>b</sup>
Medical X-Rays (Diagnostic Procedures) Average Whole Body Exposure	53	1 in 190 000

<sup>a</sup>An effective dose equivalent of 200 mrem was used to estimate the risk from inhaling <sup>222</sup>Rn and its transformation products.

<sup>b</sup>The risks from natural radiation from non-radon sources were estimated to be 1 chance in 79 000 in Los Alamos and in White Rock. The risk of lung cancer from radon exposure was estimated to be 1 chance in 50 000 for both locations. Risk estimates are derived from ICRP Publication 26 and NCRP Report 93.

**EXTERNAL PENETRATING RADIATION**

Levels of external penetrating radiation (including x and gamma rays and charged-particle contributions from cosmic, terrestrial, and manmade sources) in the Los Alamos area are monitored with thermoluminescent dosimeters (TLDs) at 147 locations.

The TLD network monitoring radiation from airborne activation products released by the LAMPF measured about 12 ± 5 mrem/year (excludes background radiation from cosmic and terrestrial sources). This measured external radiation level was used to calculate radiation dose by taking into account shielding by buildings and self-shielding by the body. The value measured in 1987 is lower than the measured 18 ± 2 mrem/year obtained in 1986 (Fig. 2). The difference is probably caused by variation in weather patterns between the two years rather than differences in LAMPF operations because the estimate of

airborne activity emitted from LAMPF increased in 1987 (Table III).

Radiation levels (including natural background radiation from cosmic and terrestrial sources) are also measured at regional, perimeter, and onsite locations in the environmental TLD network. Some measurements at onsite stations were above background levels, as expected, reflecting ongoing research activities at or historical releases from Laboratory facilities.

**AIR MONITORING**

Airborne radioactive emissions were monitored at 87 release points at the Laboratory. Total airborne emissions of activity increased about 30 000 Ci from 1986 emissions (Table III). This was principally due to an increase in estimated releases of activation products from LAMPF.

**Table III. Comparison of 1986 and 1987 Radioactive Releases from the Laboratory.**

<u>Airborne Emissions</u>				
Radionuclide	Units	<u>Activity Released</u>		Ratio 1987:1986
		1986	1987	
<sup>3</sup> H	Ci	10 700	3 180	0.3
<sup>32</sup> P	Ci	70	48	0.7
<sup>41</sup> Ar	Ci	276	232	0.8
<sup>131</sup> I	Ci	38	0	0
Uranium	MCi	847	1 080	1.3
Plutonium	mCi	207	73	0.4
Gaseous Mixed Activation Products	Ci	112 000	150 000	1.3
Mixed Fission Products	M Ci	2 570	1 290	0.5
Particulate/Vapor Activation Products	Ci	<u>0.1</u>	<u>0.2</u>	<u>2.0</u>
<b>Total</b>	<b>Ci</b>	<b>122 976</b>	<b>153 412</b>	<b>1.2</b>
<u>Liquid Effluents</u>				
Radionuclide	<u>Activity Released(mCi)</u>		Ratio 1987:1986	
	1986	1987		
<sup>3</sup> H	89 710	110 000	1.2	
<sup>89,90</sup> Sr	9.9	65	6.6	
<sup>137</sup> Sr	18	8.1	0.4	
<sup>234</sup> U	2.4	1.6	0.7	
<sup>238,239,240</sup> Pu	5.1	4.6	0.9	
<sup>241</sup> Am	3.2	3.6	1.1	
Other	<u>1 166.7</u>	<u>610.5</u>	<u>0.5</u>	
<b>Total</b>	<b>90 915.3</b>	<b>110 693.4</b>	<b>1.2</b>	

Ambient air is routinely sampled for tritium, uranium, plutonium, americium, and gross beta activity at 26 sampling stations. Measurements of radioactivity in the air are compared with concentration guides based upon DOE's RPS. These guides are concentrations of radioactivity in air breathed continuously throughout the year that result in effective doses equal to DOE's RPS of 100 mrem/year for offsite areas (Derived Concentration Guides for Uncontrolled Areas) and to the occupational RPS for onsite areas (Concentration Guides for Controlled Areas).

Hereafter, they are called guides for onsite and offsite areas.

Only the onsite tritium air concentrations showed any measurable impact resulting from Laboratory operations. Annual average concentrations of tritium remained <0.1% of DOE's guides at all stations and posed no environmental or health problems in 1987. Annual average concentrations of longer lived radionuclides in air were also <0.1% of the guides during 1987.

## WATER, SOIL, AND SEDIMENT MONITORING

Liquid effluents containing low levels of radioactivity were routinely released from one waste treatment plant and one sanitary sewage lagoon system. Concentrations at all discharge points were well below DOE's concentration guides for onsite areas. The dominant change was an increase in tritium discharge from TA-50's radioactive liquid-waste treatment facility caused by increased concentrations in the released waters (Table III).

Surface and ground waters are monitored to detect potential dispersion of radionuclides from Laboratory operations. Only the surface and shallow ground waters in onsite liquid effluent release areas contained radioactivity in concentrations that are above natural terrestrial and worldwide fallout levels. These concentrations are minute fractions (<0.1%) of DOE's guides for onsite areas. These onsite waters are not a source of industrial, agricultural, or municipal water supplies. The radiochemical quality of water from regional, perimeter, and onsite areas that have received no direct discharge showed no significant effects from Laboratory releases.

The potable water supply met all applicable EPA radiochemical and chemical standards. Lack of a hydrologic connection to the deep aquifer was confirmed by lack of radioactive or chemical contamination in municipal water supply sources.

Measurements of radioactivity in samples of soils and sediments provide data on less direct pathways of exposure. These measurements are useful for understanding hydrological transport of radioactivity in intermittent stream channels near low-level radioactive waste management areas. Onsite areas within Pueblo, Los Alamos, and Mortandad Canyons all had concentrations of radioactivity on sediments at levels slightly higher than attributable to natural terrestrial sources or worldwide fallout. The low levels of cesium, plutonium, and strontium in Mortandad Canyon are due to liquid effluents from a waste treatment plant. No above-background radioactivity on sediments or in water has been measured in locations beyond the Laboratory boundary in Mortandad Canyon. However, small amounts of radioactivity on sediments in Pueblo Canyon (from pre-1964 effluents) and Los Alamos Canyon (from

1952 to current treated effluents) have been transported to the Rio Grande. Theoretical estimates, confirmed by measurements, show that the incremental effect on Rio Grande sediments is insignificant when compared with background concentrations in soils and sediments.

Environmental monitoring is done at 1 active and 11 inactive waste management areas at the Laboratory. The general public is excluded from these controlled-access sites. Surface runoff has transported some low-level contamination from the active disposal area and several of the inactive areas into controlled-access canyons. Leachate extracts (following EPA guidelines) from the surface contamination indicate the presence of no constituents in excess of EPA criteria for hazardous waste determination.

## FOODSTUFFS MONITORING

Most fruit, vegetable, fish, bee, and honey samples from regional and perimeter locations showed no radioactivity distinguishable from that attributable to natural sources or worldwide fallout. Some produce samples from onsite locations had slightly elevated tritium concentrations, <2% of DOE's guides for tritium in water (no concentration guides exist for produce).

## UNPLANNED RELEASES

During 1987, there were two unplanned releases of radioactive or hazardous materials. Both involved release of tritium. The quantities of tritium released were small and resulted in radiation doses that were small fractions of the RPS.

***March 18 Tritium Release at the Van de Graaff Facility at TA-3.*** On March 18, 1987, 375 Ci of tritium (as elemental tritium gas) were released from the Van de Graaff facility at TA-3. Air samples collected from four downwind air samplers were within normal ranges for tritium at these locations. All measured concentrations were <0.1% of DOE's Derived Concentration Guide for tritium in offsite areas. Calculations from meteorological modeling estimated a dose to the maximum exposed individual of 0.003 mrem to the lung, <0.1% of EPA's air emission standard of 75 mrem/year (any organ) to a member of the public.

#### ***December 11-12 Tritium Release at TA-33.***

Approximately 165 Ci of elemental tritium gas were inadvertently released from TA-33 on December 11-12, 1987. Air samples were collected at five downwind locations. All measured air concentrations were within their normal range and <0.1% of DOE's Derived Concentration Guide for tritium. The highest estimated dose to a member of the public was 0.001 mrem to the lung, <0.1% of the 75 mrem/year EPA air emission standard.

### **ENVIRONMENTAL COMPLIANCE ACTIVITIES**

#### ***Resource Conservation and Recovery Act.***

The Resource Conservation and Recovery Act (RCRA) regulates hazardous wastes from generation to ultimate disposal. The EPA has transferred full authority (with the exception of the Hazardous and Solid Waste Amendment of 1984) for administering RCRA to New Mexico's Environmental Improvement Division (EID). In 1987, the Laboratory had numerous interactions with EID and prepared documentation to comply with RCRA requirements. The Laboratory has revised RCRA Parts A and B permit applications, originally submitted in 1985. The latest revisions were submitted in November 1987.

***Clean Water Act.*** Regulations under the Clean Water Act set water quality standards and effluent limitations. The two primary programs at the Laboratory to comply with the Clean Water Act are the National Pollutant Discharge Elimination System (NPDES) and the Spill Prevention Control and Countermeasure (SPCC) Program.

The NPDES requires permits for nonradioactive constituents at all point-source discharges. A single NPDES permit for the Laboratory authorizes liquid effluent discharges from 98 industrial outfalls and 10 sanitary sewage treatment outfalls; the permit expires in March 1991. The Laboratory was in compliance with the NPDES permit in about 96% and 99% of the analyses done on samples collected for compliance monitoring at sanitary and industrial waste discharges, respectively. Chronically noncompliant discharges are being upgraded under an EPA/DOE Federal Facility Compliance Agreement.

Another NPDES permit authorizes liquid effluent discharge from the Fenton Hill Geothermal

Project. The permit for a single outfall was issued to regulate the discharge of mineral-laden water from the recycle loop of the geothermal wells.

The SPCC program provides for cleanup of spills and requires preparation of an SPCC plan. The Laboratory has many elements that are required in an SPCC plan and has assembled a Laboratory-wide formal SPCC plan that was implemented in 1987.

***National Environmental Policy Act.*** The Laboratory Environmental Review Committee reviews environmental documentation required by National Environmental Policy Act legislation as well as identifying other environmental items of concern to the Laboratory. An Environmental Evaluations Coordinator helps prepare required DOE documentation and identify other items requiring committee attention. Documentation usually consists of Action Description Memorandums (brief environmental evaluations) or Environmental Assessments (more detailed evaluations). During 1987, the committee approved 20 Action Description Memorandums and 1 Environmental Assessment and forwarded this documentation to DOE.

***Clean Air Act.*** During 1987, the Laboratory's operations remained in compliance with all federal and state air quality regulations. State regulations are required to be as stringent as federal regulations, and many state standards are more stringent. Over 180 asbestos removal jobs involved the disposal of 270 m<sup>3</sup> (9500 ft<sup>3</sup>) of asbestos. All beryllium shops met emissions performance requirements. The Laboratory applied to EPA for approval to construct the Independent Management Activity facility. This program will emit depleted uranium similar to other dynamic testing projects at the Laboratory. Approval was obtained from EPA in January 1988.

***Safe Drinking Water Act.*** The municipal and industrial water supply for the Laboratory and community come from 16 deep wells and 1 gallery (collection system fed by springs). The wells range in depth from 265 to 942 m (869 to 3090 ft). The chemical quality of the water easily met EPA's National Interim Primary Drinking Water Standards (40 CFR 141) in 1987.

**Federal Insecticide, Fungicide, and Rodenticide Act.** The Federal Insecticide, Fungicide, and Rodenticide Act (FIFRA) requires registration of all pesticides, restricts use of certain pesticides, recommends standards for pesticide applicators, and regulates disposal and transportation of pesticides. The Laboratory stores, uses, and discards pesticides in compliance with this act.

**Archaeological and Historical Protection.** The Laboratory's Environmental Evaluation Coordination and Quality Assurance programs provide protection as mandated by law for the hundreds of archaeological and historical resources on Laboratory land. Pursuant to federal regulations implementing Section 106 of the National Historic Preservation Act of 1966, as amended, clearance for construction where no resource will be affected and mitigation of unavoidable adverse effects from Laboratory activity is determined in consultation with New Mexico's State Historical Preservation Office. During 1987, archaeologists performed 28 cultural resource surveys, monitored 7 projects, fenced 1 site, and undertook adverse impact mitigation at 2 sites.

**Threatened/Endangered Species and Floodplains/Wetlands Protection.** The DOE and Laboratory must comply with the Endangered Species Act of 1973, as amended, and with Executive Orders 11988, Floodplain Management, and 11990, Protection of Wetlands Environmental Review Requirements. Three Floodplains/Wetlands notifications were prepared for publication in the Federal Register. Laboratory biologists surveyed 17 proposed construction sites for potential impact. They identified no endangered or rare species at these sites.

**Comprehensive Environmental Response, Compensation, and Liability Act.** The Comprehensive Environmental Response, Compensation, and Liability Act (CERCLA) of 1980 mandated cleanup of toxic and hazardous contaminants at closed and abandoned hazardous waste sites. The Superfund Amendments and

Reauthorization Act (SARA) of 1986 extensively amended CERCLA. Laboratory compliance activities at hazardous waste sites are part of DOE's Albuquerque Operations Office's Comprehensive Environmental Restoration Program (CERP). The program is evaluating all areas at the Laboratory for possible contamination.

**Toxic Substances Control Act.** The Toxic Substances Control Act (TSCA) regulates the manufacture, processing, distribution, use, storage, and labeling of chemical substances, including polychlorinated biphenyls (PCBs). The Laboratory has EPA authorization to dispose of PCB wastes at its chemical waste landfill (Area L) and burn PCB-contaminated wastes at its Controlled Air Incinerator (99.9999% combustion efficiency). The Laboratory is in compliance with EPA's permit conditions for authorizing onsite disposal of PCB-contaminated wastes.

**Emergency Planning and Community Right-to-Know Act.** Title III of SARA, also known as the Emergency Planning and Community Right-to-Know Act (EPCRA), is the centerpiece of federal policy on chemical disaster prevention and response. In response to this legislation, the state of New Mexico has established an Emergency Response Commission (ERC) within the State Police Department's Hazardous Materials Emergency Response Division. The commission has designated Los Alamos County as the local Emergency Planning District, and the Laboratory's Emergency Management Office will continue to develop and coordinate a comprehensive laboratory-wide, all-hazards emergency response plan that is compatible with the county's overall plan.

The Industrial Hygiene (HSE-5) and Environmental Surveillance (HSE-8) groups provided to the Emergency Management Office a preliminary list of 137 chemical substances used onsite. In addition, individual Materials Safety Data Sheets for each of these 137 chemicals have also been provided to the Emergency Management Office to facilitate county planning.

## ENVIRONMENTAL SUPPORT ACTIVITIES

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*Authors: Environmental Surveillance Group  
Group: Environmental Surveillance, HSE-8  
Funding Organization: U.S. DOE and Indirect*

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### **VADOSE ZONE CHARACTERIZATION AT AREAS L AND G (Alice Barr, Anthony Grieggs, and David McInroy)**

The Resource Conservation and Recovery Act (RCRA) requires that hazardous waste disposal facilities such as the Laboratory either (1) perform ground water monitoring, or (2) obtain a waiver of ground water monitoring requirements, provided a low potential exists for migration of hazardous waste or constituents from the disposal areas to water supply wells via the uppermost aquifer. A vadose zone (unsaturated zone above the main aquifer) characterization program was initiated to substantiate the Laboratory's request for a ground water monitoring waiver.

At Areas L and G (TA-54), the uppermost aquifer is approximately 300 m (1000 ft) below the surface. The zone above the aquifer (the vadose zone) was studied to characterize its hydrogeology and evaluate the potential for contaminant migration. Data were collected to determine intrinsic permeability, moisture characteristic curves, unsaturated hydraulic conductivity, pore gas distribution, and actual contaminant presence in the vadose zone. Several conclusions reached from this study including the following: (1) The dominant mechanism of subsurface transport is through vapor-phase migration; aqueous transport of contaminants is highly unlikely. (2) Perched water is confined to the alluvium in the adjacent canyon and does not extend beneath the mesa or connect hydraulically to the main aquifer. (3) Some metal contamination exists at shallow depths in Area L. (4) Organic vapor contamination exists in Areas L and G. (5) No contamination is evident in the perched canyon water. (6) Vertical cooling fractures are present in the disposal areas but their ability to transport contaminants and water has not been determined. A final report presenting these

findings and the data collected was submitted to the EID's Hazardous Waste Program on March 31, 1987.

The analytical results of this study indicated the presence of organic vapor contamination at depths up to 30 m (100 ft). As a result, the Laboratory has initiated a program to determine the vertical and horizontal extent of this contamination and appropriate remediation, if deemed necessary. The program consists of four phases: (1) an initial experimental effort to determine the most effective method for monitoring hole completion and sampling; (2) an expanded sampling and analytical program to delineate the extent of contamination; (3) interpretation of results and proposal of any necessary remedial action; and (4) the remediation itself.

The first phase is now in progress. Four different (one existing, three new) borehole completions have been sampled. Initial analytical results indicate the new sampling technique is more effective in determining a concentration gradient. Also, the three new completion methods surpass the existing method in sensitivity, ease of installation, and cost. Additional sampling will be performed to substantiate these findings before we proceed to the second phase.

### **REMEDIAL INVESTIGATIONS AT THE PROPOSED WHITE ROCK Y INTERCHANGE (Lars Sohlt, Richard Romero, Eddie Lujan, John Salazar, and Thomas Buhl)**

The state of New Mexico is proposing to construct an interchange to improve the intersection of State Road 502 (SR 502) and the Main Hill Road (Alternate SR 502) into the Los Alamos townsite. The DOE intends to grant an easement to the state for construction and maintenance of the interchange on DOE-managed lands. The easement area would include parts of Los Alamos and Lower Pueblo Canyons that are known to have residual radioactivity at levels above background. This residual radioactivity is the result of liquid discharges from TA-2, TA-21, and TA-45. As part of DOE's ER Program, the

Environmental Surveillance Group (HSE-8) carried out an investigation to determine if the lands were suitable for release to the state without remedial action to lower levels of residual radioactivity. The results of this remedial investigation indicate that the lands in Los Alamos and Lower Pueblo Canyons are suitable for construction of the White Rock Y interchange without need for remedial action.

Above-background, residual radioactivity in Los Alamos Canyon is dominated by  $^{137}\text{Cs}$  (up to 50 pCi/g) and  $^{90}\text{Sr}$  (up to 13 pCi/g). Uranium and transuranics are also present at above-background levels, but activity concentrations are lower. These radionuclides have deposited in the alluvial accumulation of sediment where the canyon's stream intersects SR 502. Within Lower Pueblo Canyon,  $^{239}\text{Pu}$  is the dominant residual radionuclide (up to 15 pCi/g), and uranium is also present at above-background levels. These radionuclides have deposited where the canyon's stream widens, upstream from its confluence with Los Alamos Canyon.

Transport pathways analyses were carried out using conservative scenarios to determine if the levels of residual radioactivity indicated that remedial action was necessary before an easement is granted to the state. The two scenarios that were considered are listed below.

- o Construction activity in Los Alamos and Lower Pueblo Canyons.
- o Removal of soil material for use in a home garden.

Potential pathways of exposure within the construction scenario include worker inhalation of dust suspended during earth-moving activities and direct exposure to gamma radiation from residual  $^{137}\text{Cs}$ . Within the home garden scenario, it was assumed that material was removed from the construction site for use as garden soil. Potential pathways for exposure of a home gardener include direct gamma radiation from  $^{137}\text{Cs}$ , inhalation of dust suspended during gardening activities, ingestion of produce grown in the garden, and ingestion of water from a nearby well that has received radionuclides leached from garden soils.

For residual radioactivity in Los Alamos Canyon, the pathways analyses resulted in a

calculated commitment of 9 mrem/year effective dose equivalent within the construction scenario and 29 mrem/year effective dose equivalent within the home garden scenario. For residual radioactivity in Lower Pueblo Canyon, the pathways analyses resulted in a calculated commitment of 4 mrem/year effective dose equivalent within the construction scenario and 9 mrem/year effective dose equivalent within the home garden scenario. All of these doses are less than the 100 mrem/year effective dose equivalent commitment that serves as DOE's RPS for protection of the general public. Maximum concentrations of airborne radionuclides during construction would be less than 15% of DOE's limits for exposure of the general public.

#### **ENVIRONMENTAL MONITORING AT THE FENTON HILL SITE (William Purtymun and Max Maes, HSE-8; Roger Ferenbaugh and Mary Williams, HSE-9)**

The Laboratory is currently evaluating the feasibility of extracting thermal energy from the hot dry rock geothermal reservoir at the Fenton Hill Geothermal Site (TA-57). The site is about 45 km (28 mi) west of Los Alamos on the southern edge of the Valles Caldera. The hot dry rock energy concept involves drilling two deep holes, connecting these holes by hydraulic fracturing, and bringing thermal energy to the surface by circulating water through the system. Environmental monitoring is performed adjacent to the site to assess any impacts from the geothermal operations.

The chemical quality of surface and ground waters in the vicinity of TA-57 (Fig. 3) has been determined for use in geohydrologic and environmental studies. These water quality studies began before construction and testing of the hot dry rock system.<sup>1</sup> The most recent samples were collected in November 1987.

Surface water stations (13 on the Jemez River, the Rio Guadalupe, and their tributaries) are divided into four general groups based on the predominant ions and total dissolved salts (TDS) (Table IV). The predominant ions are (1) sodium and chloride, (2) calcium and bicarbonate, (3) calcium and sulfate, and (4) sodium and bicarbonate. Ground water stations (five mineral and hot springs, one well, and five springs) are also grouped according to predominant ions.

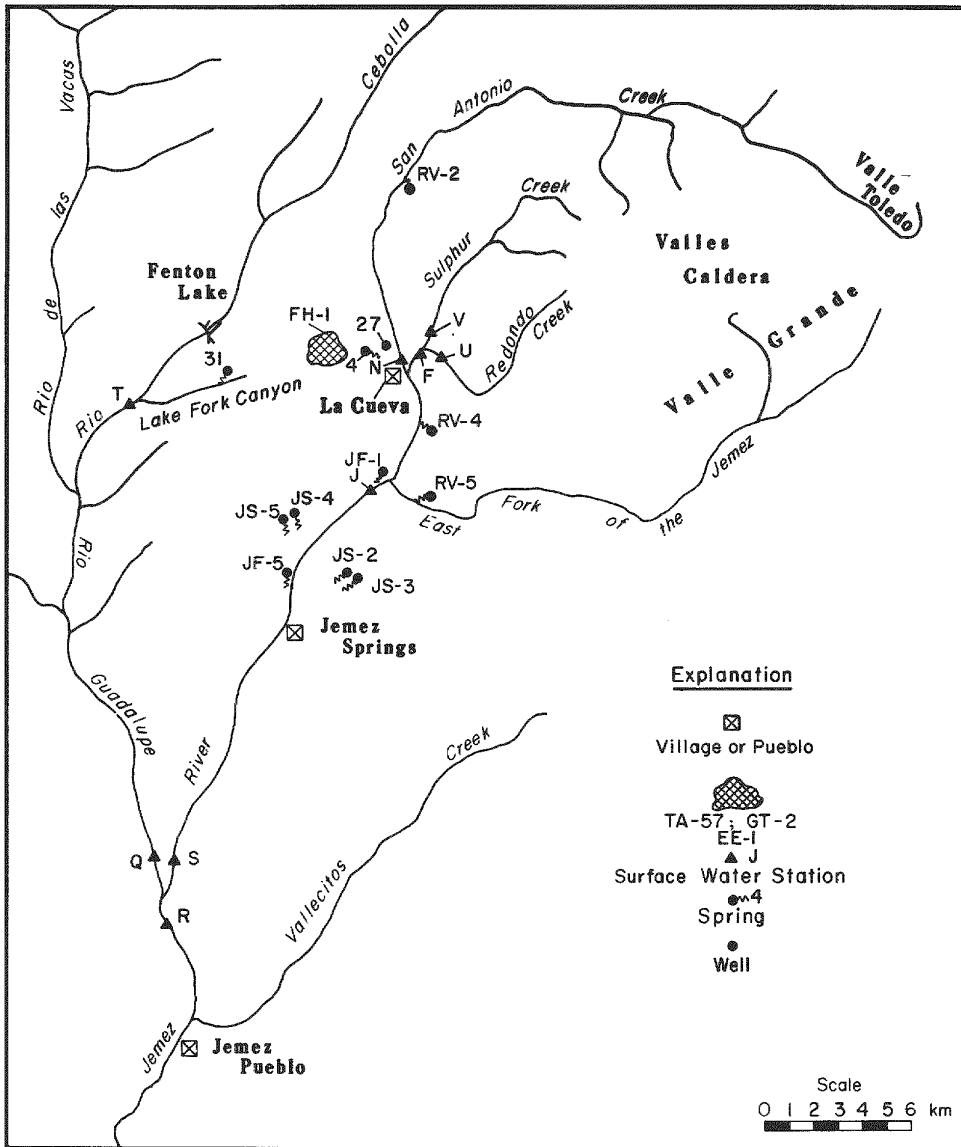


Figure 3. Sampling stations for surface and ground water near the Fenton Hill Site.

These ions are (1) sodium and chloride, (2) calcium and bicarbonate, and (3) sodium and bicarbonate (Table IV).

The chemical quality of surface and ground water at the individual stations has not changed significantly from previous years.<sup>2</sup>

#### REFERENCES

1. W. D. Purtymun, F. G. West, and W. H. Adams, "Preliminary Study of Quality of Water in the Drainage Area of the Jemez River and Rio Guadalupe," Los Alamos Scientific Laboratory report LA-5595-MS (April 1974).
2. W. D. Purtymun, R. W. Ferenbaugh, M. C. Williams, and M. N. Maes, "Water Quality in the Vicinity of Fenton Hill, 1985 and 1986," Los Alamos National Laboratory report LA-11210-PR (March 1988).

<b>Table IV. Quality of Surface and Groundwaters at Fenton Hill Geothermal Site (Concentrations in mg/L) - November 1987.<sup>a</sup></b>			
<b><u>Surface Water</u></b>			
	<b>Na</b>	<b>Cl</b>	<b>TDS</b>
<b>Sodium Chloride</b>			
Redondo Creek (U)	10	15	126
Jemez River (R)	75	97	436
Jemez River (S)	85	132	388
	<b>Na</b>	<b>HCO<sub>3</sub></b>	<b>TDS</b>
<b>Calcium Bicarbonate</b>			
San Antonio Creek (N)	16	62	141
Rio Cebolla (T)	10	88	118
Rio Guadalupe (Q)	15	170	228
Lake Fork 1 (LF-1)	10	54	132
Lake Fork 2 (LF-2)	17	71	168
Lake Fork 3 (LF-3)	13	50	220
Lake Fork 4 (LF-4)	15	72	284
	<b>Ca</b>	<b>SO<sub>4</sub></b>	<b>TDS</b>
<b>Calcium Sulfate</b>			
Sulphur Creek (V)	52	305	456
Sulphur Creek (F)	28	66	150
	<b>Na</b>	<b>HCO<sub>3</sub></b>	<b>TDS</b>
<b>Sodium Bicarbonate</b>			
Jemez River (J)	16	59	104
<b><u>Groundwater</u></b>			
	<b>Na</b>	<b>Cl</b>	<b>TDS</b>
<b>Sodium Chloride</b>			
Loc. JF-1 (Hot Spr)	460	1000	1940
Loc. JF-5 (Hot Spr)	960	2300	3830
	<b>Ca</b>	<b>HCO<sub>3</sub></b>	<b>TDS</b>
<b>Calcium Bicarbonate</b>			
FH-1 (Supply Well)	48	117	280
Loc. 39 (Spr)	13	41	52
	<b>Na</b>	<b>HCO<sub>3</sub></b>	<b>TDS</b>
<b>Sodium Bicarbonate</b>			
JS-2, 3 (Spr)	17	80	160
JS-4, 5 (Spr)	17	72	154
Loc. 4 (Spr)	16	55	92
Loc. 31 (Spr)	11	62	110
RV-2 (Hot Spr)	22	45	114
RV-4 (Hot Spr)	52	123	168
RV-5 (Hot Spr)	20	83	78
<sup>a</sup> See Fig. 3 for sampling locations. One sample taken at each location.			

## **DISTRIBUTION OF RADIONUCLIDES IN CHANNEL ALLUVIUM OF MORTANDAD CANYON (Donald VanEtten, William Purtymun, and Max Maes, HSE-8; Richard Peters, HSE-9)**

Trace amounts of radionuclides remaining in effluent are released from the treatment plant at TA-50 into adjacent Mortandad Canyon (Table V). The effluent recharges a shallow body of ground water in the alluvium. The radionuclides in the effluent are adsorbed or bound to the sediments in the channel, reducing the amount found in the water of the shallow aquifer. This shallow aquifer is of limited extent and lies within the Laboratory boundary.

The sediments and radionuclides in the stream channel alluvium are subject to transport by additional releases of effluent or by storm runoff. The small drainage area of the canyon and the ability of the thick section of unsaturated alluvium to store the runoff has prevented transport to the Laboratory boundary. A series of sediment traps have been installed in the canyon since early 1970 to confine the surface runoff and contaminants within the Laboratory. The traps range from gravel-filled galleries to stilling basins that contain suspended solids as well as bed sediment (alluvium).

A storm on June 7, 1987, produced a record 50-year, 2-h rainfall of 5.5 cm (2.16 in.). The rainfall resulted in the largest runoff event in Mortandad Canyon since hydrologic studies began in 1960. The peak discharge in the upper canyon at gaging station GS-1 was estimated to be 4.5 m<sup>3</sup>/s [160 ft<sup>3</sup>/s (cfs)]. Two other large runoff events occurred in August 1968 [3.2 m<sup>3</sup>/s (115 cfs)] and November 1987 [2.9 m<sup>3</sup>/s (102 cfs)]. The peak discharge at the sediment traps of the June 1987 event was about 3 m<sup>3</sup>/s (100 cfs). The runoff filled the two sediment traps and overflowed into the third. The estimated volume of runoff was 3500 m<sup>3</sup> (930 000 gal.).

A set of sediment samples collected in the canyon on June 16, 1987, was analyzed for transuranics and gamma-emitting radionuclides (Fig. 4). The concentrations of plutonium and americium above the effluent outfall from TA-50 (stations 1, 2, and A) were background (Table VI). The <sup>238</sup>Pu concentrations between gaging station GS-1 and station 7, just above the sediment trap,

ranged from 3.3 to 11.7 pCi/g, and the <sup>239, 240</sup>Pu in the same reach of canyon ranged from 12.2 to 39.3 pCi/g. The <sup>241</sup>Am for this same sampling area ranged from 11.72 to 33.81 pCi/g. The largest concentrations of transuranics in sediments were in sediment trap 1: 18 pCi/g of <sup>238</sup>Pu, 58.9 pCi/g of <sup>239, 240</sup>Pu, and 79.50 pCi/g of <sup>241</sup>Am. The concentrations decreased in trap 2 and decreased further in trap 3. All of the bed sediments and most of the suspended sediments were retained in trap 1, resulting in higher concentrations here than in traps 2 and 3.

Gamma-emitting radionuclides followed the same trends as transuranics (Table VII). The concentrations above the effluent outfall were background. The highest concentrations were for <sup>137</sup>Cs with a range from 15.3 to 62.9 pCi/g in the channel above the traps and 96.7 pCi/g in trap 1. Trace amounts of <sup>134</sup>Cs, <sup>59, 60</sup>Co, and <sup>75</sup>Se were found in the channel sediment samples with the highest concentrations in the sediment traps.

The sediments from traps 1 and 2 were analyzed using EPA's toxic characteristic leach procedure (TCLP) to identify hazardous wastes. Analyses were carried out for pesticides (8 compounds), extractable organics (15 compounds), volatile organics (18 compounds), and metal (8 elements). None of these were detected.

Previous runoff events have not been contained in the area of the sediment traps. The analyses of sediments below the traps indicated that runoff events had carried radionuclides to station 10. Below station 10 and still within the Laboratory, the concentrations of radionuclides were at or below background. The maintenance of the sediment traps is essential to contain residual radioactivity within the Laboratory boundaries.

## **BIOMONITORING OF THE LABORATORY'S LIQUID EFFLUENTS (Roy Bohn and Charles Nylander)**

Group HSE-8 has initiated a biomonitoring program at the Laboratory in support of its NPDES program. Biomonitoring is used as a strategy to evaluate the overall toxic impact of effluents without specifically identifying individual contaminants.

Table V. Quality of Effluent from the TA-50 Liquid Radioactive Waste Treatment Plant for 1987.<sup>a</sup>

Radionuclide	Activity Released (mCi)	Mean Concentration (Ci/mL)	Mean as % DOE's CG <sup>b</sup>
<sup>3</sup> H	100,000	3.8 x 10 <sup>-3</sup>	3.8
<sup>89</sup> Sr	64	2.4 x 10 <sup>-6</sup>	0.8
<sup>90</sup> Sr	1.0	3.9 x 10 <sup>-8</sup>	0.4
<sup>137</sup> Cs	8.1	3.0 x 10 <sup>-7</sup>	0.1
<sup>234</sup> U	1.6	6.0 x 10 <sup>-8</sup>	<0.1
<sup>238</sup> Pu	1.4	5.3 x 10 <sup>-8</sup>	<0.1
<sup>239,230</sup> Pu	3.2	1.2 x 10 <sup>-7</sup>	0.1
<sup>241</sup> Am	3.6	1.3 x 10 <sup>-7</sup>	0.1
Nonradioactive Constituents			Mean Concentration (mg/L)
Cd <sup>c</sup>			1.1 x 10 <sup>-3</sup>
Ca			170
Cl			150
Total Cr <sup>c</sup>			2.4 x 10 <sup>-2</sup>
Cu <sup>c</sup>			0.33
F			12
Hg <sup>c</sup>			4.9 x 10 <sup>-4</sup>
Mg			1.1
Na			920
Pb <sup>c</sup>			5.1 x 10 <sup>-2</sup>
Zn <sup>c</sup>			0.32
CN			0.3
COD			100
NO <sub>3</sub> -N			476
PO <sub>4</sub>			1.5
TDS			4150
pH <sup>c</sup>			6.98 - 7.77
Total Effluent Volume - 2.66 x 10 <sup>7</sup> L			
<sup>a</sup> As reported on DOE forms F-5821.1.			
<sup>b</sup> Department of Energy's Concentration Guide for Controlled Areas.			
<sup>c</sup> Constituents regulated by National Pollutant Discharge Elimination System permit.			

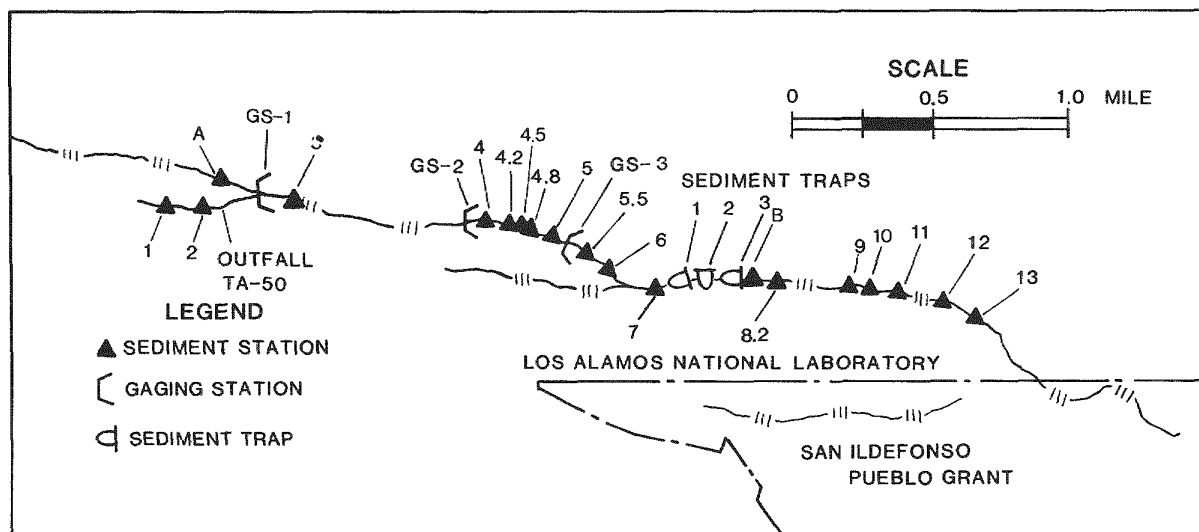


Figure 4. Sediment sampling stations in Mortandad Canyon.

Table VI. Transuranics in Mortandad Canyon Channel Alluvium, June 16, 1987.<sup>a</sup>

Station	<sup>238</sup> Pu (pCi/g)	<sup>239,240</sup> Pu (pCi/g)	<sup>241</sup> Am (pCi/g)
<b>Effluent Canyon</b>			
1	0.012 (0.007)	0.024 (0.011)	-0.008 (0.002)
2	0.011 (0.006)	0.033 (0.011)	0.130 (0.050)
TA-50 Outfall	0.712 (0.058)	1.81 (0.107)	0.970 (0.050)
<b>Mortandad Canyon</b>			
A	0.005 (0.009)	0.033 (0.010)	0.02 (0.002)
GS-1	3.91 (0.330)	16.2 (1.20)	11.72 (0.14)
3	7.69 (0.580)	17.6 (1.30)	16.79 (0.17)
GS-2	7.00 (0.560)	24.6 (1.80)	31.96 (0.24)
4	7.27 (0.550)	26.2 (1.80)	33.81 (0.24)
4.2	11.7 (0.070)	39.3 (2.30)	22.08 (0.20)
4.5	4.69 (0.37)	18.9 (1.30)	19.85 (0.20)
4.8	4.26 (0.390)	18.1 (1.40)	28.76 (0.22)
5	7.11 (0.690)	29.7 (2.20)	20.51 (0.19)
GS-3	4.67 (0.450)	20.1 (1.60)	22.60 (0.19)
5.5	5.67 (0.430)	24.5 (1.60)	27.71 (0.22)
6	6.29 (0.610)	16.2 (1.40)	14.60 (0.16)
7	3.30 (0.171)	12.2 (0.519)	16.53 (0.17)
Sediment Trap 1	18.2 (1.30)	58.9 (3.90)	79.50 (0.40)
Sediment Trap 2	9.71 (0.750)	35.8 (2.60)	51.42 (0.31)
Sediment Trap 3	2.06 (0.126)	7.06 (0.329)	10.11 (0.13)
3	2.13 (0.24)	7.17 (0.64)	12.53 (0.15)
8.2	0.105 (0.018)	0.399 (0.037)	0.56 (0.03)
10	0.09 (0.02)	0.33 (0.04)	0.31 (0.03)
11	0.095 (0.024)	0.330 (0.042)	0.01 (0.002)
12	-0.025 (0.017)	0.010 (0.012)	0.1 (0.01)
13	-0.010 (0.011)	0.089 (0.020)	0.8 (0.002)

<sup>a</sup>Location of sediment stations shown on Fig. 4; counting uncertainty in parentheses.

Table VII. Radionuclides in Mortandad Canyon Channel Alluvium, June 16, 1987.<sup>a</sup>

Station	<sup>137</sup> Cs (pCi/g)	<sup>134</sup> Cs (pCi/g)	<sup>60</sup> Co (pCi/g)	<sup>57</sup> Co (pCi/g)	<sup>75</sup> Se (pCi/g)
<b>Effluent Canyon</b>					
1	0.24 (0.10)	0.11 (0.09)	0.07 (0.14)	0.39 (0.21)	-0.11 (0.14)
2	0.005 (0.07)	-0.12 (0.09)	-0.07 (0.12)	0.02 (0.14)	-0.03 (0.09)
TA-50 Outfall	0.69 (0.14)	0.28 (0.12)	0.16 (0.13)	1.56 (0.33)	0.71 (0.17)
<b>Mortandad Canyon</b>					
A	0.13 (0.09)	0.03 (0.08)	-0.10 (0.11)	0.09 (0.014)	0.003 (0.008)
GS-1	15.30 (2.31)	0.83 (0.17)	1.73 (0.30)	15.60 (2.36)	2.82 (0.44)
3	30.20 (4.55)	0.25 (0.18)	0.73 (0.20)	8.23 (1.26)	2.13 (0.36)
GS-2	48.90 (7.36)	0.10 (0.09)	0.40 (0.16)	4.30 (0.69)	2.00 (0.34)
4	62.90 (9.46)	0.97 (0.18)	0.35 (0.14)	3.52 (0.55)	1.92 (0.31)
4.2	31.60 (4.76)	0.34 (0.11)	0.23 (0.17)	3.00 (0.53)	1.27 (0.24)
4.5	37.30 (5.63)	0.56 (0.16)	0.18 (0.17)	1.80 (0.35)	0.73 (0.17)
4.8	43.20 (6.50)	0.78 (0.15)	0.12 (0.12)	1.99 (0.33)	1.33 (0.23)
5	23.50 (3.55)	0.29 (0.12)	0.02 (0.15)	1.75 (0.36)	0.79 (0.18)
GS-3	39.50 (5.95)	0.84 (0.16)	0.20 (0.12)	1.45 (0.25)	0.76 (0.15)
5.5	55.40 (8.33)	0.98 (0.19)	0.24 (0.12)	2.08 (0.35)	1.24 (0.23)
6	53.30 (8.01)	0.16 (0.11)	0.11 (0.16)	1.02 (0.29)	0.21 (0.12)
7	29.70 (4.48)	0.34 (0.10)	-0.18 (0.12)	0.85 (0.21)	0.34 (0.12)
Sediment Trap 1	96.70 (19.5)	0.24 (0.12)	1.36 (0.26)	11.70 (1.79)	7.64 (1.17)
Sediment Trap 2	96.50 (14.5)	1.68 (0.28)	0.71 (0.18)	5.52 (0.85)	3.41 (0.52)
Sediment Trap 3	15.10 (2.29)	0.16 (0.10)	-0.19 (0.14)	1.37 (0.36)	0.61 (0.17)
B	27.40 (4.13)	0.37 (0.16)	0.21 (0.19)	0.70 (0.32)	0.38 (0.19)
8.2	3.12 (0.48)	0.29 (0.10)	-0.14 (0.13)	0.48 (0.21)	0.07 (0.14)
10	1.60 (0.26)	0.23 (0.11)	-0.09 (0.14)	0.66 (0.22)	0.16 (0.14)
11	0.46 (0.13)	0.02 (0.08)	-0.27 (0.14)	-0.06 (0.12)	0.09 (0.09)
12	0.63 (0.15)	0.19 (0.11)	-0.20 (0.14)	0.66 (0.24)	0.01 (0.12)
13	0.23 (0.08)	-0.08 (0.09)	-0.05 (0.12)	-0.16 (0.14)	0.09 (0.09)

<sup>a</sup>Location of sediment stations shown on Fig. 4; counting uncertainty in parentheses.

With over 100 NPDES-permitted outfalls at the Laboratory, consistent monitoring of each effluent is not feasible. Outfalls were segregated into nine basic categories according to wastewater source. Biomonitoring samples are collected from one representative outfall of each category. Biomonitoring assays using *Daphnia pulex* as a test organism are conducted for each representative effluent and LC<sub>50</sub> values are calculated. To date each outfall has been sampled three times and preliminary results indicate that overall water quality of effluents is good. Biomonitoring sampling will continue in 1988.

#### **NATIONAL ATMOSPHERIC DEPOSITION PROGRAM (NADP) NETWORK STATION (David Nochumson and Michael Trujillo)**

Group HSE-8 operates at Bandelier National Monument a wet deposition station that is part of the NADP Network. Composite precipitation samples are collected on a weekly basis. The samples are initially weighed and analyzed for pH and conductivity before being sent to Colorado State University for analysis of ionic species. Summary statistics of the data for the four latest completed quarters are presented in Table VIII.

The magnitude of the ionic species deposition was generally highest in the third quarter of 1987 and lowest during the second quarter. The amount of precipitation was also lowest during the second quarter of 1987. The amount of deposition is quite variable, reflecting the variability in the cleanliness of the atmosphere that the storm clouds have contacted. The ions in the rainwater are both manmade and natural, coming from both nearby and distant locations. High nitrate and sulfate deposition is most likely from manmade sources (motor vehicles, copper smelters, and power plants).

The natural pH of local rainfall, without manmade contribution, is unknown. It is most likely higher than 5.6, however, because of the contribution from alkaline soils. For the latest four quarters, all but two of the weekly samples had pHs below 5.6, which indicates contributions from acidic species other than carbon dioxide.

#### **VADOSE ZONE CHARACTERIZATION AT TA-16, AREA P (Steven McLin, David McInroy, and Anthony Grieggs)**

The hydrologic transmitting characteristics of the vadose zone in Area P are under detailed investigation. These efforts will support the ground water monitoring waiver that was requested in December 1987 as required under 40 CFR 265, Subpart F. This waiver must demonstrate a low potential for migration of hazardous wastes or their components from the landfill via the uppermost aquifer to water supply wells or to surface water in Canon de Valle.

Based on available information, major potential migratory pathways from the landfill include (in decreasing order of importance) (1) surface erosion into Canon de Valle waters and subsequent sediment transport; (2) shallow percolation into the underlying unsaturated tuff with hydraulic interconnection to the surface stream; and (3) deep percolation to the major freshwater aquifer. Soluble barium nitrate is the major contaminant of concern, although other substances may also be present in the landfill. During a December 1987 survey of locations adjacent to the landfill, barium concentrations did not exceed 3 mg/L; in the past, barium concentrations have occasionally exceeded 100 mg/L.

Five neutron moisture access wells and nine ground water monitoring wells were installed around the landfill in 1987. Additional boreholes were also drilled approximately 240 m (800 ft) south to verify suspected stratigraphic unit correlations within the unsaturated Bandelier Tuff and to obtain continuous core samples from a third borehole for laboratory testing. A thin veneer of alluvium [locally less than 1.5 m (5 ft)], has been deposited on the floor of Canyon de Valle; however, the entire landfill site is underlain by the Bandelier Tuff.

Two major lithologic subunits were identified at Area P, based on degree of welding. The uppermost subunit varies in thickness from about 40 to 60 m (140 to over 200 ft) and consists of unsaturated, friable to moderately welded,

Table VIII. Wet Deposition Measurements (eq/m <sup>2</sup> unless specified). <sup>a</sup>				
Parameter	Fourth Quarter 1986	First Quarter 1987	Second Quarter 1987	Third Quarter 1987
Precipitation (in.)	0.45 (0.0-2.0)	0.27 (0.0-.83)	0.17 (0.0-.77)	0.40 (0.0-1.49)
Field pH	4.8 (4.6-5.1)	4.8 (4.0-5.8)	4.7 (4.6-4.9)	4.7 (4.1-5.5)
Calcium	68 (0.5-470)	92 (1.5-290)	66 (0.5-200)	180 (1.0-580)
Magnesium	9.4 (0.0-48)	14 (1.6-29)	12 (0.0-31)	29 (0.0-86)
Potassium	4.2 (0.0-25)	3.4 (0.0-6.6)	7.8 (0.0-37)	9.8 (0.0-32)
Sodium	31 (0.0-90)	34 (1.7-90)	15 (0.0-43.9)	39 (1.3-84)
Ammonium	110 (1.1-47)	63 (1.1-270)	79 (0.55-270)	110 (4.4-300)
Nitrate	130 (0.8-450)	133 (16-310)	120 (0.58-300)	300 (32-60)
Chloride	19 (3.4-51)	26 (2.0-64)	15 (0.84-63)	41 (1.1-100)
Sulfate	280 (2.1-1000)	150 (21-430)	140 (1.5-390)	280 (31-590)
Phosphate	-- --	-- --	6.9 --	2.2 --

<sup>a</sup>Mean; range in parentheses.

yellowish-brown tuff. The lower subunit, also unsaturated, consists of a densely welded gray tuff. The top of the major freshwater aquifer is estimated to be between 240 and 370 m (800 and 1200 ft) below the surface of Area P.

Continuous core samples were recovered from well P-16A, immediately south of the western portion of the Area P landfill. Total borehole depth was about 25 m (80 ft); this test hole was converted to a neutron moisture access well when 2.5-in. aluminum casing was set.

Laboratory testing on selected core segments included analysis for a number of parameters: saturated and unsaturated hydraulic conductivity, moisture retention characteristics, initial gravimetric and volumetric moisture contents, bulk density, and porosity. This information will be used in a numerical simulation of potential barium migration from the landfill through these upper Bandelier Tuff units in an attempt to evaluate the likelihood of deep ground water contamination. A detailed water balance computation and sediment erosion characterization study for Canon de Valle will complete the efforts required under the waiver request.

#### **ENVIRONMENTAL STUDIES OF TA-49 (William Purtymun, Alan Stoker, and Max Maes)**

From 1959 to 1961, hydronuclear experiments were conducted in underground shafts at TA-49, on Frijoles Mesa in the southwest corner of the Laboratory between TA-28 and TA-33 (Fig. 5). These experiments involved a combination of conventional (chemical) high explosives, usually in a nuclear weapon configuration, and fissile material in quantities far below the amount required for a nuclear explosion. A total of 35 hydronuclear experiments and 9 related equation-of-state and criticality experiments, all involving some fissile material, were conducted. Other experiments involving high explosives but no fissile materials were conducted during the same period.

A total of about 41 kg (90 lb) of plutonium, 93 kg (200 lb) of enriched uranium, 82 kg (180 lb) of depleted uranium, and 15 kg (33 lb) of beryllium was used. These materials were dispersed in the bottoms of the shafts by detonation of the conventional (chemical) high explosives.

Some surface plutonium contamination measured in one experimental area in December 1960 was traced to cuttings from a shaft drilled during October and November. Plutonium had apparently been dispersed through fractures in the tuff by the detonation of an experiment in an adjacent, experimental shaft. All surface soil contamination ascertainable by standard procedures and instruments of the time was cleaned up and placed back in the shaft from which it originated.<sup>1</sup>

Three deep test wells (DT-5A, DT-9, and DT-10) extend from the surface of the mesa at TA-49 into the main aquifer of the Los Alamos area (Fig. 6). The depth to the main aquifer is about 360 m (1200 ft). No water is perched in beds between the surface of the mesa and the top of the main aquifer. The chemical and radiochemical quality of water from these wells indicated no contamination from activities at TA-49.

Eleven sediment surface stations were established in 1972 in natural drainage from the experimental areas. A twelfth station was added in 1981 because the drainage had changed (Fig. 6). Samples collected in 1986 and 1987 indicated that sediments at Station A-3 contained plutonium concentrations in excess of background (Table IX). The concentrations are below cleanup levels (100 pCi/g) and are from the chemistry building (removed) at Area 11. Analytical results indicated that <sup>3</sup>H, <sup>137</sup>Cs, total uranium, and gross gamma were at or near background levels.

Sediments from the 12 stations were analyzed for chemical constituents extracted from sediments downgradient from the experimental area (Fig. 6). The results of the analyses indicated that the constituents were below limits of detection or EP toxic criteria concentrations where applicable (Table X).

Storm runoff samples were taken from four stations in late August and early September 1987. The <sup>137</sup>Cs, plutonium in solution, and plutonium in suspended sediments were at or below background, indicating no detectable transport in storm runoff (Table XI).

Analysis of the chemical quality of the runoff showed that the water contained only naturally occurring constituents (Table XII).

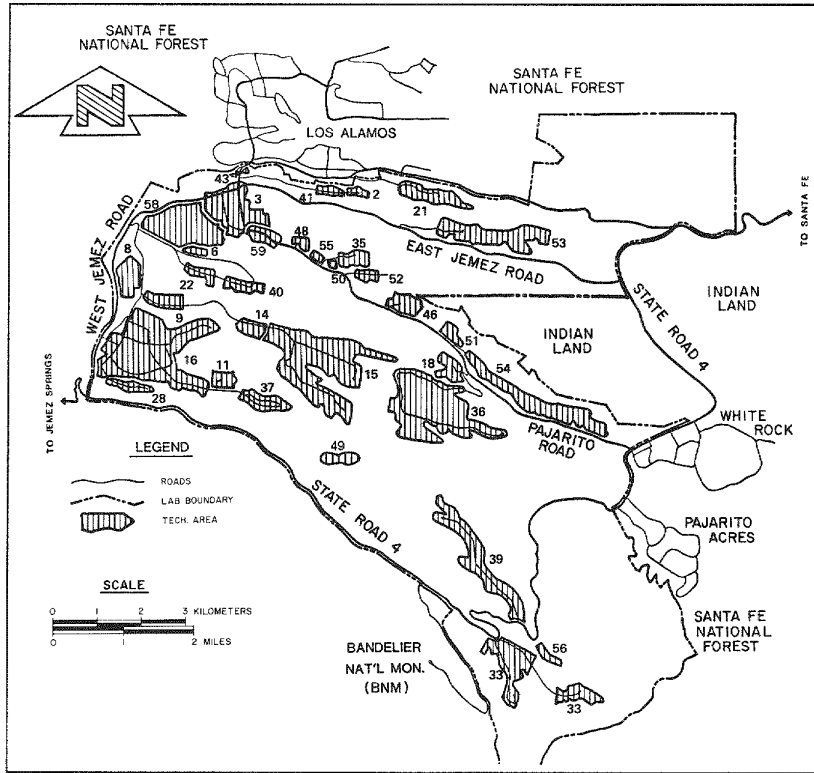


Figure 5. Technical Areas (TAs) of Los Alamos National Laboratory.

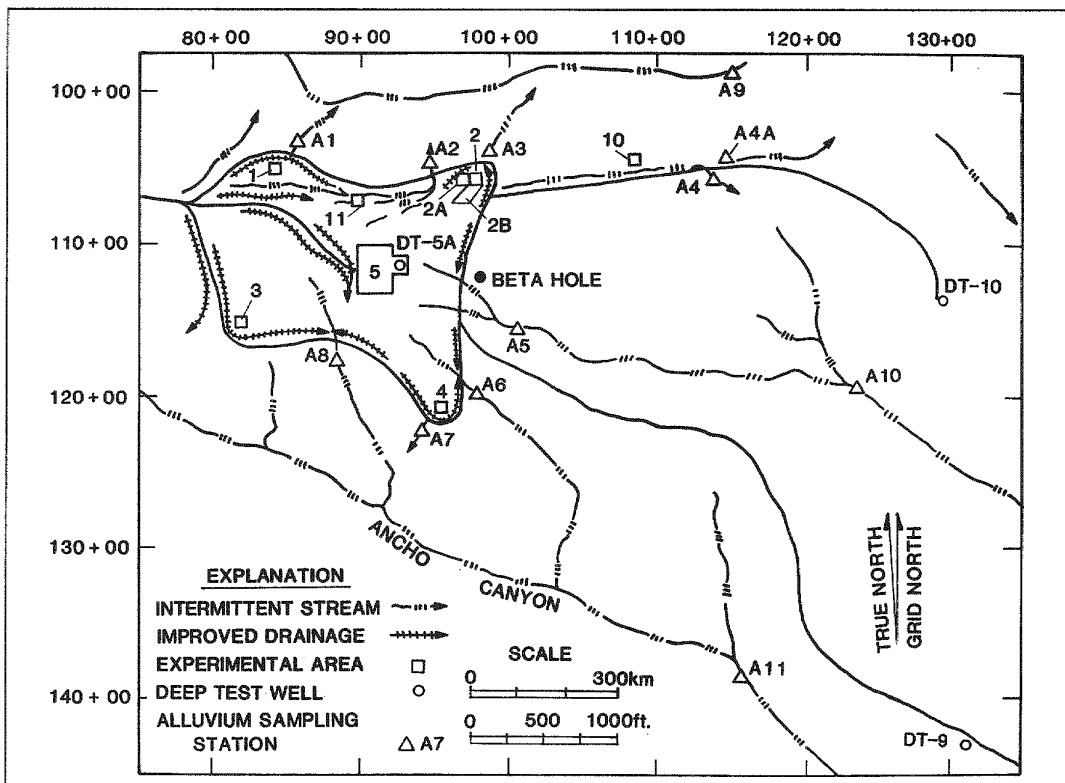


Figure 6. Location of experimental areas and test wells at TA-49.

Table IX. Radiochemical Analyses of Sediments at TA-49.

Station No.	Date	$^3\text{H}$ ( $10^{-6}\text{pCi/g}$ )	$^{137}\text{Cs}$ ( $10^{-9}\text{pCi/g}$ )	Total U (g/g)	$^{238}\text{Pu}$ ( $10^{-9}\text{pCi/g}$ )	$^{239,240}\text{Pu}$ ( $10^{-9}\text{pCi/g}$ )	Gross Gamma (Counts/Min/g)
A-1	8/6/86	2.4 (0.5)	0.11 (0.05)	3.0 (0.3)	0.000 (0.001)	0.004 (0.002)	4.0 (0.7)
A-1	4/13/87	-- --	0.08 (0.07)	2.3 (0.2)	0.004 (0.002)	0.003 (0.002)	7.3 (0.9)
A-2	8/6/86	5.4 (0.7)	0.35 (0.10)	4.2 (0.4)	0.005 (0.002)	0.022 (0.004)	5.2 (0.7)
A-2	4/13/87	-- --	0.22 (0.08)	3.7 (0.4)	0.001 (0.001)	0.004 (0.005)	9.0 (1.0)
A-3	8/6/86	3.4 (0.5)	1.2 (0.21)	5.3 (0.5)	0.216 (0.013)	10.7 (0.425)	6.5 (0.7)
A-3	4/13/87	-- --	0.29 (0.08)	4.7 (0.5)	0.001 (0.000)	0.083 (0.010)	10 (1.0)
A-4	8/6/87	5.7 (0.7)	-0.20 (0.05)	4.2 (0.4)	0.000 (0.001)	0.004 (0.002)	5.5 (0.8)
A-4	4/13/87	-- --	0.16 (0.09)	3.4 (0.3)	0.001 (0.002)	0.006 (0.002)	9.0 (1.0)
A-4A	8/6/86	3.1 (0.5)	0.70 (0.15)	4.0 (0.4)	0.001 (0.001)	0.000 (0.001)	6.5 (0.8)
A-4A	4/13/87	-- --	0.23 (0.09)	3.4 (0.3)	0.001 (0.001)	0.008 (0.002)	9.0 (1.0)
A-5	8/6/87	3.0 (0.5)	-0.08 (0.02)	3.8 (0.4)	0.001 (0.002)	0.042 (0.006)	5.8 (0.8)
A-5	4/13/87	-- --	0.39 (0.11)	3.4 (0.3)	0.000 (0.001)	0.016 (0.004)	9.0 (1.0)
A-6	8/6/86	4.7 (0.6)	0.49 (0.10)	4.2 (0.4)	0.002 (0.001)	0.012 (0.002)	6.3 (0.8)
A-6	4/13/87	-- --	0.14 (0.08)	3.5 (0.4)	0.000 (0.001)	0.001 (0.001)	9.0 (1.0)
A-7	8/6/86	4.1 (0.6)	0.48 (0.10)	4.1 (0.4)	0.003 (0.002)	0.016 (0.003)	6.3 (0.8)
A-7	4/13/87	-- --	0.37 (0.11)	3.9 (0.4)	0.002 (0.001)	0.016 (0.003)	8.0 (1.0)
A-8	8/6/86	3.8 (0.6)	0.20 (0.06)	2.6 (0.3)	0.001 (0.001)	0.001 (0.002)	4.4 (0.7)
A-8	4/13/87	-- --	0.17 (0.09)	3.4 (0.3)	-0.001 (0.002)	0.004 (0.002)	7.8 (0.9)
A-9	8/8/86	6.4 (0.8)	0.10 (0.06)	3.9 (0.4)	0.000 (0.001)	0.003 (0.002)	6.1 (0.8)
A-9	4/13/87	-- --	0.09 (0.09)	2.9 (0.3)	0.000 (0.001)	0.002 (0.002)	6.3 (0.8)
A-10	8/6/86	8.0 (0.9)	0.10 (0.07)	2.6 (0.3)	0.000 (0.001)	0.004 (0.002)	4.8 (0.7)
A-10	4/13/87	-- --	0.45 (0.11)	3.7 (0.4)	0.005 (0.001)	0.014 (0.003)	9.0 (1.0)
A-11	8/6/86	2.9 (0.5)	0.03 (0.06)	3.2 (0.3)	0.000 (0.001)	0.003 (0.002)	5.9 (0.8)
A-11	4/13/87	-- --	0.57 (0.13)	2.6 (0.3)	0.002 (0.002)	0.010 (0.002)	7.0 (0.9)

Table IX. Radiochemical Analyses of Sediments at TA-49 (Cont.).							
Station No.	Date	$^3\text{H}$ ( $10^{-6}$ pCi/g)	$^{137}\text{Cs}$ ( $10^{-9}$ pCi/g)	Total U (g/g)	$^{238}\text{Pu}$ ( $10^{-9}$ pCi/g)	$^{239,240}\text{Pu}$ ( $10^{-9}$ pCi/g)	Gross Gamma (Counts/Min/g)
A Back-ground	4/13/87	--	0.20 (0.09)	3.4 (0.3)	0.000 (0.001)	0.005 (0.002)	5.7 (0.8)
B Back-ground	4/13/87	--	0.05 (0.08)	2.9 (0.3)	0.004 (0.003)	0.001 (0.003)	3.1 (0.6)
Limits of Detection		0.3	0.1	0.1	0.002	0.002	0.1
Maximum Concentration		8.0	1.2 (0.21)	5.3 (0.5)	0.216 (0.013)	10.7 (0.425)	10 (1.0)
Regional Background		7.2	0.44	4.4	0.006	0.023	7.9
Maximum as % of Regional Background		111	272	120	3600	4652	126

Note: Station number shown in Fig. 6; counting uncertainty in parentheses.

**Table X. Chemical Concentrations in Solution Extracted from Sediments Downgradient from Experimental Areas at TA-49.**

Chemical <sup>b</sup>	Max. EP Toxic Concentration <sup>c</sup>	Limit of Detection	Station Numbers <sup>a</sup>											Background <sup>a</sup>		
			A-1	A-2	A-3	A-4	A-4A	A-5	A-6	A-7	A-8	A-9	A-10	A-11	A	B
Arsenic	5.0	0.05	BLD	BLD	BLD	BLD	BLD	BLD	BLD	BLD	BLD	BLD	BLD	BLD	BLD	BLD
Barium	100	0.5	0.6	BLD	0.6	BLD	0.8	0.5	0.7	BLD	0.6	BLD	BLD	BLD	0.5	1.3
Cadmium	1.0	0.01	BLD	BLD	BLD	BLD	BLD	BLD	BLD	BLD	BLD	BLD	BLD	BLD	BLD	BLD
Chromium	5.0	0.05	BLD	BLD	BLD	BLD	BLD	BLD	BLD	BLD	BLD	BLD	BLD	BLD	BLD	BLD
Lead	5.0	0.05	BLD	0.06	BLD	BLD	BLD	BLD	BLD	BLD	BLD	BLD	BLD	BLD	BLD	BLD
Mercury	2.0	0.005	BLD	BLD	BLD	BLD	BLD	BLD	BLD	BLD	BLD	BLD	BLD	BLD	BLD	BLD
Selenium	1.0	0.01	BLD	BLD	BLD	BLD	BLD	BLD	BLD	BLD	BLD	BLD	BLD	BLD	BLD	BLD
Silver	---	0.05	BLD	BLD	BLD	BLD	BLD	BLD	BLD	BLD	BLD	BLD	BLD	BLD	BLD	BLD
Nickel	---	0.01	BLD	BLD	BLD	BLD	BLD	BLD	BLD	BLD	BLD	BLD	BLD	BLD	BLD	BLD
Beryllium <sup>d</sup>	---	0.001	BLD	BLD	BLD	BLD	BLD	BLD	BLD	BLD	BLD	BLD	BLD	BLD	BLD	BLD
Sulfate	---	0.2	BLD	BLD	BLD	0.4	BLD	BLD	0.3	BLD	1.1	BLD	BLD	BLD	BLD	0.5
Nitrate	---	0.2	BLD	BLD	BLD	BLD	BLD	BLD	BLD	BLD	BLD	BLD	BLD	BLD	BLD	BLD
Uranium <sup>d</sup>	---	1.0	BLD	BLD	BLD	BLD	BLD	BLD	BLD	BLD	BLD	BLD	BLD	BLD	BLD	7.9

<sup>a</sup>Station number shown in Fig. 6; background stations are Bandelier National Monument, entrance (A) and small canyon north of supply well PM-1 (B).

<sup>b</sup>Concentrations in mg/L except as noted; BLD = Below Limit of Detection.

<sup>c</sup>New Mexico Hazardous Waste Management Regulations (HWMR) 201 B.5.; extraction procedure.

<sup>d</sup>Units are µg/g.

Table XI. Storm Run-Off from TA-49.

Sta.	Date 1987	Solution			Suspended Sediments	
		$^{137}\text{Cs}$ ( $10^{-9}\text{Ci/mL}$ )	$^{238}\text{Pu}$ ( $10^{-9}\text{Ci/mL}$ )	$^{239,240}\text{Pu}$ ( $10^{-9}\text{Ci/mL}$ )	$^{238}\text{Pu}$ (pCi/g)	$^{239,240}\text{Pu}$ (pCi/g)
A-1	8/24	152 (65)	0.007 (0.011)	0.010 (0.008)	0.000 (0.001)	0.027 (0.006)
A-1	8/24	15 (60)	-0.008 (0.011)	0.000 (0.010)	-0.004 (0.008)	0.048 (0.013)
A-2	8/24	38 (60)	-0.057 (0.033)	0.000 (0.010)	0.001 (0.001)	0.002 (0.001)
A-5	8/24	-13 (60)	-0.009 (0.015)	0.033 (0.018)	0.000 (0.001)	0.007 (0.033)
A-5	8/28	67 (61)	0.008 (0.008)	0.025 (0.010)	0.000 (0.001)	0.006 (0.002)
A-5	9/10	82 (61)	0.011 (0.014)	-0.004 (0.004)	0.004 (0.002)	0.024 (0.004)
A-8	8/24	45 (53)	0.000 (0.010)	0.000 (0.010)	0.001 (0.001)	0.001 (0.001)
A-8	8/28	-27 (60)	0.021 (0.017)	0.005 (0.014)	0.000 (0.001)	0.028 (0.004)

Table XII. Chemical Quality of Storm Run-Off from TA-49.						
Station	Date 1987	mg/L				
		As	Cr	N	SO <sub>4</sub>	pH
A-1	8/24	0.002	0.02	1.2	2	6.0
A-1	8/24	0.002	0.03	0.3	3	7.8
A-2	8/24	0.003	0.02	0.2	2	7.7
A-5	8/24	0.003	0.03	0.2	2	7.6
A-5	8/28	0.002	0.04	1.0	2	7.1
A-5	9/10	0.003	0.03	0.2	2	7.0
A-8	8/24	0.003	0.04	0.7	4	8.1
A-8	8/28	0.008	0.04	0.7	2	7.0
Standard <sup>a</sup>		0.05	0.05	10	250	
Constituent <sup>b</sup>		Concentration (mg/L)		Standards <sup>a</sup> (mg/L)		
Ag		<0.05		0.05		
Ba		<0.1		1.0		
Be		<0.01		--		
CN		<0.01		--		
Cd		<0.01		0.01		
Hg		<0.002		0.002		
Ni		<0.01		--		
Pb		<0.01		0.05		
Se		<0.02		0.01		
<sup>a</sup> Primary or secondary drinking water standards.						
<sup>b</sup> Analyzed from stations for each run-off event.						

## REFERENCE

1. William D. Purtymun and Alan K. Stoker, "Environmental Status of Technical Area 49, Los Alamos, New Mexico," Los Alamos National Laboratory report LA-11135-MS (November 1987).

## QUALITY OF SURFACE AND GROUND WATER ADJACENT TO THE LOS ALAMOS NATIONAL LABORATORY: ORGANIC COMPOUNDS (William Purtymun, Roger Ferenbaugh, and Max Maes)

Surface and ground water samples were collected from 43 stations representing the major

occurrences of natural and municipal water and industrial and sanitary effluents in the Los Alamos area (Fig. 7). The samples were analyzed for volatile organics (35 compounds), semivolatile organics (65 compounds), BNA fraction, pesticides (20 compounds), herbicides (3 compounds), polychlorinated biphenyls (7 compounds), and cyanides. These samples were analyzed to investigate possible areas of organic contamination for further study; however, the amount of organic contamination in surface and ground water is minimal. A limited program of organic monitoring will be incorporated into the annual surveillance of surface and ground water in and adjacent to the Laboratory.<sup>1</sup>



expanded to seven units during 1987. Three HPICs continued to monitor external radiation levels north, north-northeast, and northeast of LAMPF, across Los Alamos Canyon, during most of the LAMPF operating cycle, June through November. The other four units were placed at various locations for shorter periods. Monitors were placed at Kwage Mesa [2.0 km (1.2 mi) north of LAMPF], Bayo sewage treatment plant [2.3 km (1.4 mi) northeast of LAMPF in Bayo Canyon], locations north-northwest and east-northeast of LAMPF across Los Alamos Canyon, 0.8 km (0.5 mi) east of LAMPF, 0.5 km (0.3 mi) northwest of LAMPF in Los Alamos Canyon, several sites 1.2-2.6 km (0.7-1.6 mi) south to southwest of LAMPF on mesas, and a site west-southwest of LAMPF in Mortandad Canyon. Most of the siting took advantage of the high frequency of south to southwesterly and north to northeasterly winds caused by Rio Grande Valley channeling.

Results to date confirm that the highest external radiation levels are transported toward the northeast and north-northeast. However, the highest short-term (an hour or so) levels of over 100 mR/h were found east of LAMPF, with over-the-mesa transport. Higher short-term levels were also found north of LAMPF. External radiation dropped off by about 50% with increases in downwind distance of 0.8-2.0 km (0.5-1.2 mi). Above-background external radiation was detected at all canyon sites, especially in Los Alamos Canyon, at 0.5 km (0.3 mi) downwind. Radiation levels occasionally exceeded 50-60 mR/h at this site. Much of these levels may be a result of shine of the LAMPF plume traveling overhead. Predicted external radiation levels using onsite meteorological data and release data agree well with measured concentrations at all sites.

#### **BIOTRAN DEVELOPMENT AND APPLICATIONS (Anthony Gallegos, Malcolm Ennis, and Lars Sohlt)**

The computer code BIOTRAN, a model of environmental transport of transuranic radionuclides, was applied to an active waste disposal area at Los Alamos as part of the code's validation and verification program. A user's manual was also prepared as part of the code's documentation.

#### **RANGER VAN (Wayne Scoggins and Donald VanEtten)**

A prototype of a real-time radiation-mapping system, Ranger, was developed for DOE's Accident Response Group to respond to an accident involving the release of plutonium. Ranger has demonstrated that it can provide an efficient method of monitoring large areas for radioactive contamination. As a result of experience gained with the prototype, the external computer and software were upgraded to obtain a fully operational system.

The new system uses the prototype's commercially available line-of-sight microwave system for determining position and the same radiation detection instruments. The data obtained from the radiation detection instruments are linked back to the external computer along with the relative position of the measurement through the ranging system. The data are both displayed on a gridded map as colored circles and permanently stored in real-time. The different colors represent different contamination levels. Contours can be drawn using the permanently stored data.

#### **SURVEY OF CLEANUP COSTS OF LAND CONTAMINATED WITH TRANSURANICS (Malcolm Ennis and Thomas Buhl)**

The costs of cleaning up environmental transuranic contamination were evaluated. Cleanup operations at Palomares (Spain), Rocky Flats (Colorado), Thule (Greenland), and Enewetak Atoll (Marshall Islands) were included. Data available from these cleanup operations were not detailed enough to allow accurate projections of costs based on these data alone. A computer program, DECON, developed at Battelle Pacific Northwest Laboratory is useful for estimating cleanup costs. Given initial concentrations of radionuclides in soil and a dose or concentration limit to achieve, DECON will estimate the costs of meeting the chosen cleanup limit.

Two categories of procedures are available for reducing health risks from environmental contamination by transuranic radionuclides: in situ stabilization (including plowing and covering with topsoil or asphalt) and removal of soil and

vegetation. In situ stabilization is much more cost effective, but high levels of contamination require removal of contaminated material. Current analysis shows that when removal is necessary, costs will be much higher and will be dominated by the costs of transportation and low-level radioactive waste disposal.

#### **FURTHER CHARACTERIZATION OF DEPLETED URANIUM IN POTRILLO CANYON (Naomi Becker)**

Characterization studies of depleted uranium continued in the Potrillo Canyon watershed. These studies form part of larger investigations that describe the movement of depleted uranium deposited over the watershed through the Laboratory's dynamic testing program of weapons components.

One hundred fifty-one soil samples were collected from three transects in the canyon floor, perpendicular to the canyon axis. Samples were taken at various depths along the transects. This work is designed to determine the extent of depleted uranium storage, both spatially and vertically, in this portion of the watershed. These data also can be used to derive important geomorphologic and hydrologic information on the dynamics of semiarid basins and how these dynamics affect the transport of heavy-metal contaminants. Analytical results for these samples are pending.

One hundred thirty soil and sediment samples were submitted for analysis as part of an exercise to describe the grain-size distribution of depleted uranium from various locations within the watershed. Samples were collected from firing pads, stream channel and banks at several locations, and from presumed sediment accumulation areas. Each sample was sieved into seven distinct particle sizes before they were submitted for analyses. This information will be used in hydrologic modeling of contaminant transport through the basin and exiting the

watershed. Analytical results for these samples are pending.

Runoff samples of water and suspended sediment were collected from individual precipitation events during the summer. In addition, because of the lack of continuous discharge data, data on crest stage recorder levels and erosion chains were collected to provide information on maximum discharge and depth of scour after each runoff event. This effort is part of an ongoing monitoring program of depleted uranium transport from summer precipitation events. The program characterizes the concentration of uranium along different portions of the watershed and determines the amount of depleted uranium leaving the Laboratory.

#### **DEPLETED URANIUM INVESTIGATIONS (Naomi Becker)**

Characterization of depleted uranium mobility in the environment at Eglin Air Force Base, Florida, was started as part of a multiyear study. The Air Force uses depleted uranium under atmospheric conditions for its life-cycle testing and weapons development programs. Investigations during FY 1988 included the preparation of a report summarizing previous environmental monitoring for depleted uranium at three specific areas on the Eglin reservation. Also included are some recommendations to amplify and enrich the present, ongoing environmental surveillance program for depleted uranium.

Additionally, 38 soil and sediment samples were collected from transport pathway and background locations to assess the extent of depleted uranium transport. Samples were taken both at the surface and at various depths to assess surficial and vertical mobility. Results indicate that uranium exists at depths exceeding 1 ft in areas of considerable surface contamination. Transport from surface water runoff also seems to be present.

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## ***LOS ALAMOS NATIONAL ENVIRONMENTAL RESEARCH PARK***

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***Author: K. V. Bostick (LA/NERP Coordinator)***  
***Group: Environmental Science, HSE-12***  
***Funding Organization: DOE Office of Health and  
Environmental Research, Ecological Research  
Division***

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The Los Alamos National Environmental Research Park (LA/NERP) is an outdoor laboratory for conducting environmental research to further the basic understanding of ecosystem structure and function under various conditions of stress and to develop advanced concepts and supporting technology that will help reach national environmental goals. The LA/NERP encompasses approximately 112 km<sup>2</sup> of Department of Energy land at Los Alamos. The steep elevation gradient (1500 m in 25 km) and canyon/mesa terrain give the LA/NERP a wide spectrum of southwestern habitat types in a compact area.

A unique feature of the LA/NERP is that some areas within the park have been protected from activities such as agriculture, lumbering, and mining for more than 40 years. Also making the

LA/NERP a unique and valuable research resource are the presence of trace levels of both radioactive and nonradioactive materials resulting from technology development at Los Alamos, the appreciable technical resources of the Laboratory, and the physical security of outdoor study areas.

Much of the research conducted on the LA/NERP is performed by graduate and undergraduate students from colleges and universities located nationwide. LA/NERP management encourages this use as much as possible. Some of the work is conducted in cooperation with federal and state agencies such as the National Park Service and the New Mexico Department of Game and Fish. Use of the LA/NERP by researchers with independent funding is also encouraged.

In 1988 the LA/NERP, with the U.S. Forest Service and New Mexico Department of Game and Fish "Share with Wildlife" fund, cosponsored a study of Cooper's Hawks and Northern Goshawks. A report of that study follows.

## **HUNTING STRATEGIES AND HABITAT SELECTION PATTERNS OF COOPER'S HAWKS AND NORTHERN GOSHAWKS**

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**Author:** Patricia Kennedy

**Group:** Environmental Science, HSE-12

**Funding Organization:** Los Alamos National  
Environmental Research Park, U.S. Forest  
Service, New Mexico Dept. of Game and Fish  
"Share with Wildlife" fund

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Data collection for this project was completed in 1988. During the nesting season 12 hawks were fitted with radio transmitters and their hunting patterns monitored throughout the nesting season. A total of 34 hawks have been radio-tracked throughout the duration of the project.

Additional dietary information was obtained by systematic collections of prey remains and castings at nests and observations of prey deliveries made by radio-tagged birds. The dietary collections accumulated during the three field seasons (400+ prey remains, 220 castings, and 150 prey deliveries) were analyzed during the fall of 1988.

Estimating the abundance of prey (small mammals and birds) within the hunting ranges of hawks was conducted during the 1988 field season. Diurnal small mammal populations were sampled with line transects and live-trapping grids. Avian populations were sampled using point-count transects. The small mammal data were analyzed during the fall and a manuscript summarizing these data was submitted to a journal for publication.

The nest site habitat data collected during previous field seasons were summarized in a recent publication.<sup>1</sup>

### **REFERENCE**

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## ***EFFECTS OF ENVIRONMENTAL VARIABLES ON SORPTION ISOTHERMS***

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*Group: Environmental Science, HSE-12*  
*Cooperator: H. R. Fuentes, University of Texas at El Paso*  
*Funding Organization: DOE Yucca Mountain Project*

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Daniels et al.<sup>1</sup> have studied the sorption of radionuclides on tuff for the Yucca Mountain Project (previously the Nevada Nuclear Waste Storage Investigation Project). They investigated sorption as a function of mineralogy, temperature, particle size, solution concentration, water composition, time, temperature, and pH. Thomas<sup>2</sup> evaluated the effects of those variables on nonequilibrium distribution coefficients ( $R_d$ ), and Fuentes et al.<sup>3</sup> modeled these data sets with four conventional isotherms: Linear, Langmuir, Freundlich, and Modified Freundlich.<sup>3</sup> Not all the data sets could be modeled, however, because the above experiments were not designed for isotherm modeling. More recent work has been aimed at evaluating the effects of the above variables on sorption isotherms. The variables that could be evaluated include replication, particle size, hysteresis, and mineralogy.

We used routines developed by SAS<sup>4</sup> to perform F-tests in an attempt to determine whether the regression line for one data set was significantly different from that of another data set, the difference in the two data sets representing a particular variable (particle size, for example). To complete the evaluation, we needed the statistics for a third regression line, the combined data of the two sets.

The results of the F-tests indicate that replication and mineralogy do not affect the regression lines of the isotherms. However, the results for mineralogy were limited and cannot be generalized. A hysteresis effect, the difference between the regression lines for adsorption and desorption data sets, was observed, but the data for

this phenomenon were also limited and cannot be generalized.

With the exception of barium sorption on tuff sample YM-22, particle size apparently did not affect the regression of the isotherms either. Figure 1 shows the regression lines for barium sorption on YM-22 tuff for two ranges of particle sizes (<75  $\mu$ m and 75-500  $\mu$ m). These two data sets exhibited much less variability than did the other data sets. The large variability of the other data sets masked any significant effect of particle size on the regression of isotherm models.

The results of this study can be useful in the design and prioritization of future isotherm experiments. We have learned that sorption can be mathematically represented as isotherms in transport codes designed to evaluate radionuclide release scenarios for the candidate high-level waste repository.

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3. H. R. Fuentes, W. L. Polzer, J. Gruber, B. Lauctes, and E. H. Essington, "Preliminary Report on Sorption Modeling," Los Alamos National Laboratory report LA-10952-MS (August 1987).
4. SAS 0-917382-66-8. "SAS User's Guide, Statistics, Version 5 Edition," SAS Institute, Inc., Cary, NC.

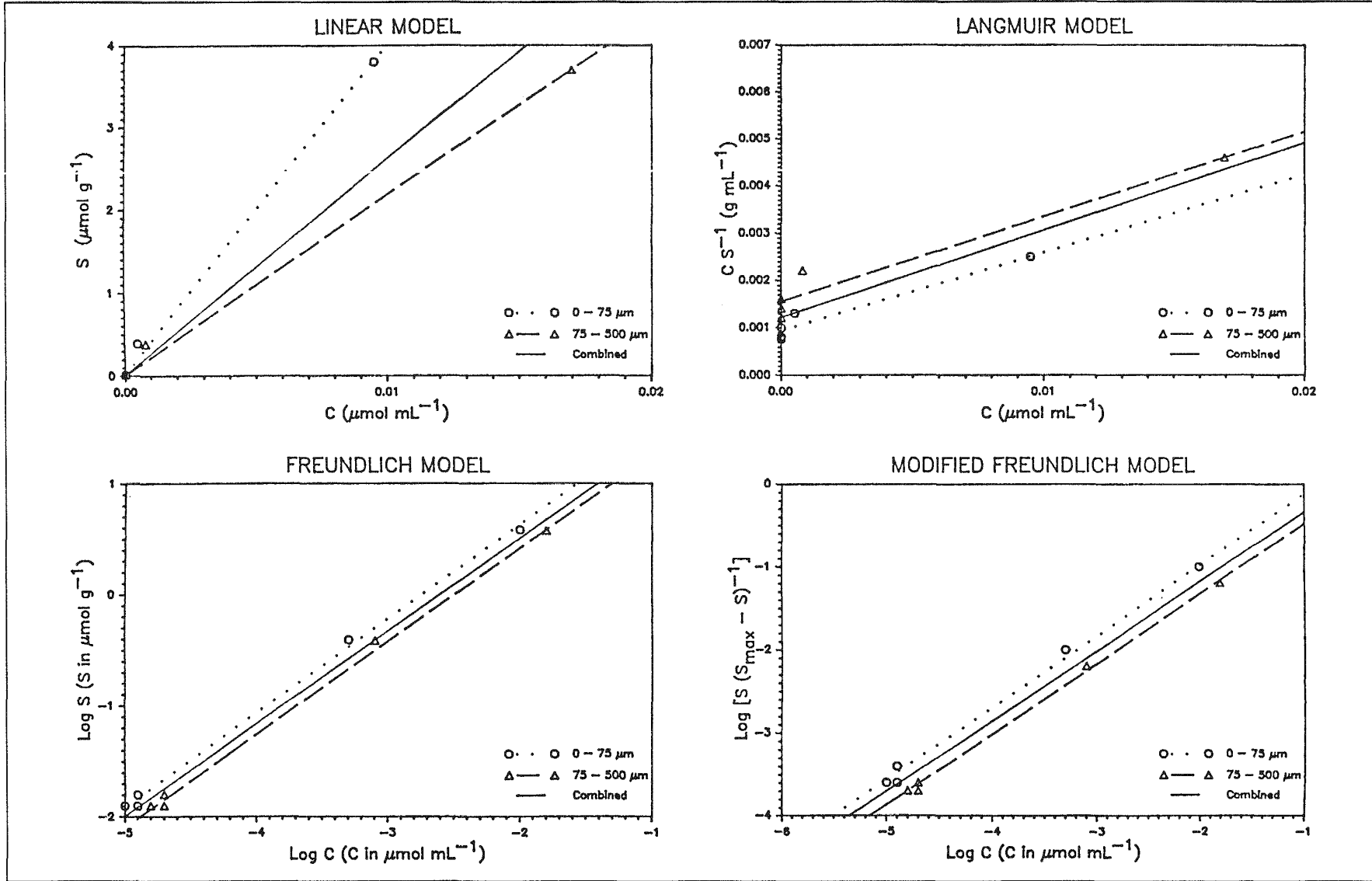


Figure 1. These isotherm plots show the effect of particle size on the adsorption of barium on YM-22 tuff for a contact time of 21 days in an air atmosphere at ambient temperature. A cation exchange capacity of  $44.0 \mu\text{eq g}^{-1}$  for the  $<75\text{-m}$  tuff and  $58.0 \mu\text{eq g}^{-1}$  for the  $75\text{- to }500\text{-m}$  tuff were used for  $S_{\text{max}}$  in the Modified Freundlich expression.

## CONTAMINANT TRANSPORT IN SOUTHWESTERN ECOSYSTEMS

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*Funding Organization: DOE Office of Health and  
Environmental Research, Ecological Research  
Division*

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Geographic information systems (GIS) are becoming increasingly important in analysis of environmental problems because they can easily manage and manipulate spatially distributed data. This capability is particularly important in confronting scale-related problems associated with hydrologic analyses of arid/semiarid watersheds. Automated data manipulation allows us to examine how various levels of detail affect the prediction of hydrologic response. In this research, conducted in cooperation with Pennsylvania State University (PSU), we used a GIS to investigate how the level of detail for watershed representation affected the predicted hydrologic response.

Our study area was Watershed 15 in the Walnut Gulch Experimental Watershed near Tombstone, Arizona, operated by the U.S. Department of Agriculture, Agricultural Research Service (USDA-ARS), Aridland Watershed Management Unit. Employees of USDA-ARS provided us with soil information and rainfall and runoff records covering 13 years.

Researchers from PSU used a U.S. Geological Survey digital elevation model (DEM) of the site as the basis for their GIS. They superimposed soil data onto the GIS by referencing to known locations on the watershed. At this point, soils and topographic information could be provided to a hydrologic model.

We selected the SPUR model<sup>1</sup> as the hydrologic model for this study. SPUR offered several advantages: (1) it was designed to operate with daily rainfall data, (2) it included a channel loss algorithm that is important in arid/semiarid environments, (3) it has a dynamic vegetation component, and (4) it is spatially distributed. This last point is important because a distributed model is needed if we are to take advantage of the GIS's rapid manipulation of spatial data.

SPUR divides a watershed into three areas: (1) upland or contributing areas, (2) channels, and (3) ponds. Associated with each channel are a maximum of three contributing areas where runoff is generated. Determination of the model parameters that govern runoff and plant growth requires knowledge of the soil properties. These properties may differ from one contributing area to another or vary within a single contributing area. The GIS was used to determine the average value of a given property on a given contributing area.

In this study we used the number of channels to represent Watershed 15 at three different levels. The simplest level had a single one-channel with three contributing areas. The next levels had 6 channels with 15 contributing areas and 1 pond. The most detailed level had 28 channels, 69 contributing areas, and 3 ponds. Data in the GIS were used to estimate parameters for each of these cases. Missing information included data on the vegetation. This deficiency forced us to calibrate this portion of the model to produce a "best" estimate of the vegetation dynamics of the area.

The simulations used the 13 years of available rainfall data to drive the model; predicted runoff and peakflow values were compared with observed data. Figure 1 is an example using runoff from the watershed with 28 channels. Considerable underprediction is obvious; that is, fewer events were predicted than observed. The level of watershed detail did not affect the predictive capability of the model, as other plots (not shown) for six channels and for one channel were similar. These results indicate that obtaining

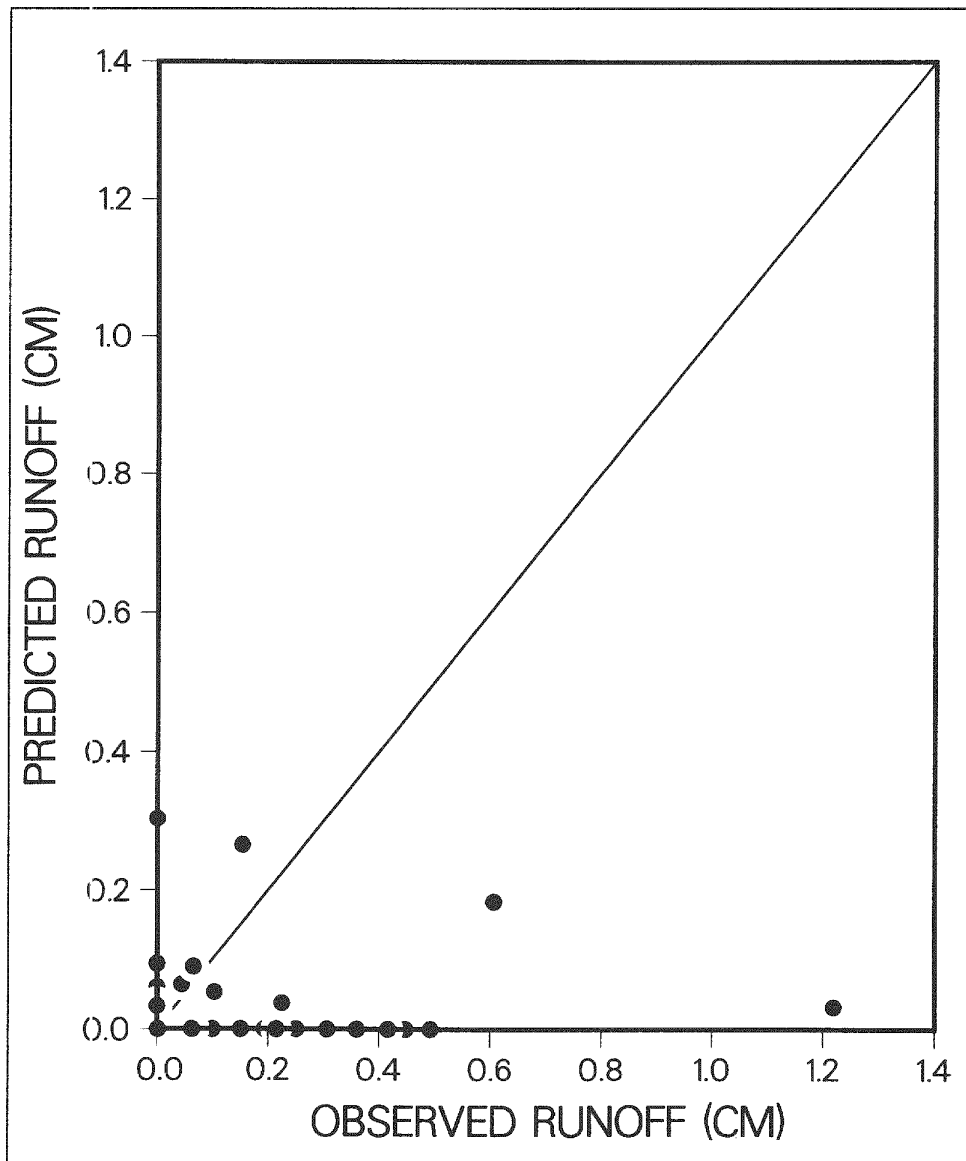


Figure 1. Plot of observed versus predicted runoff volume from Watershed 15 located at Walnut Gulch Experimental Watershed for the case where the watershed is represented by 28 channels, 69 contributing areas, and 3 ponds.

the correct parameter values is more important than accurately representing the watershed.

Another type of comparison can be seen in Fig. 2. Here the flood frequency for the 13 years covered by the data is compared with the observed data for each of the levels of watershed representation. The flood frequency data are obtained by taking the maximum peakflow discharge occurring in each year, ordering them by increasing magnitude, then plotting the ordered data on a probability scale.

This approach compares the statistical distributions of the observed and predicted values. From Fig. 2, the predicted values are still lower than the observed values, but the simplest representation is the closest to the observed values. This result was unexpected because in principle a more detailed spatial representation should lead to more accurate predictions. Again, the result stresses the need for accurate parameters regardless of the level of detail used in representing an area.

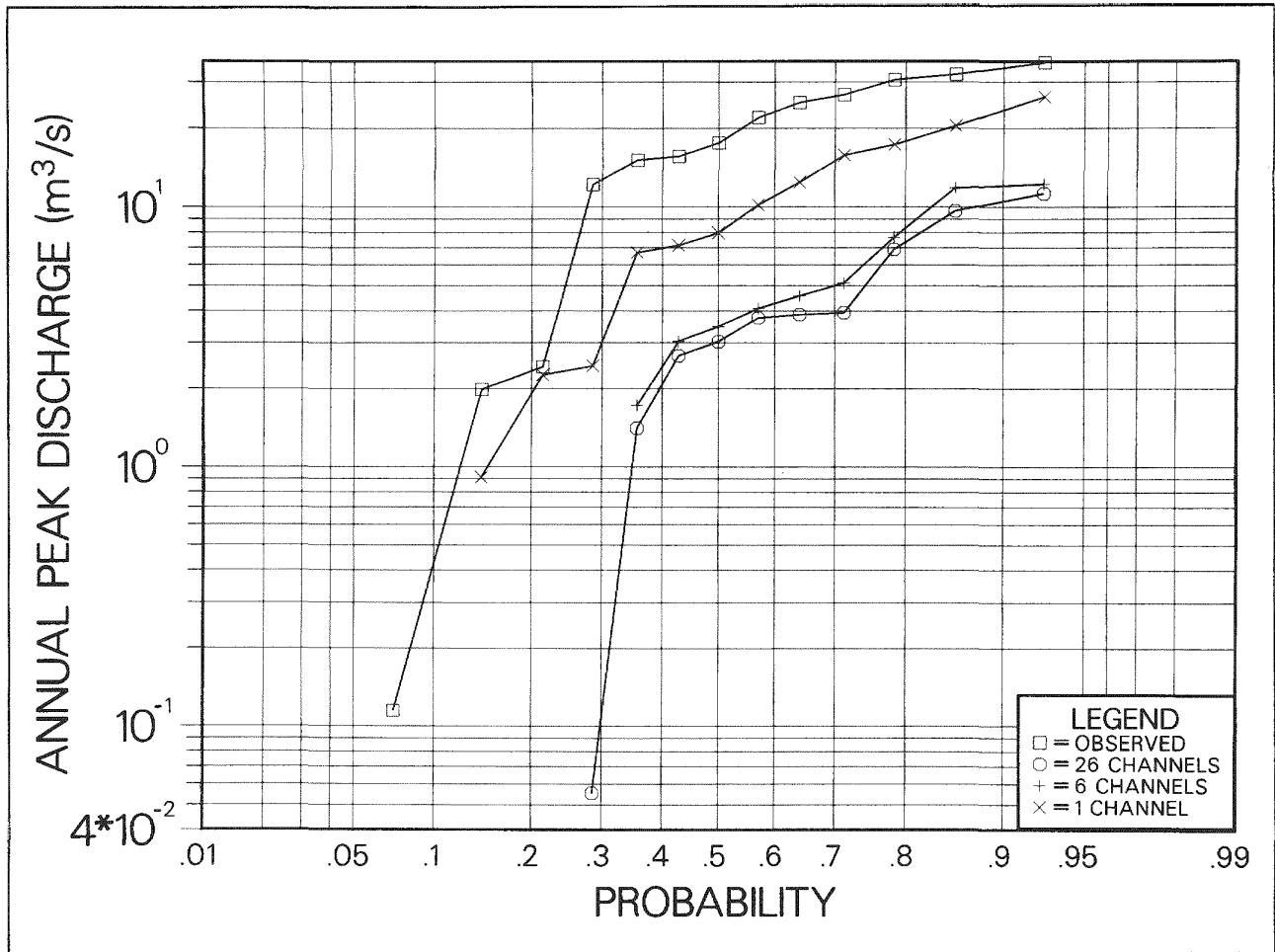


Figure 2. Flood frequency plot for Watershed 15 comparing the observed data with the various levels of representation of the watershed by the SPUR model.

The study of contaminant transport generally requires more spatial detail because contaminants are rarely spread uniformly over a watershed. Therefore, the utility of a GIS in providing representation of watersheds as was done in this study is important. Future efforts need to strive more toward identifying parameters than attempting to spatially represent the watershed.

#### REFERENCE

1. J. R. Wight and J. W. Skiles, eds. SPUR: Simulation of Production and Utilization of Rangelands. Documentation and user guide. USDA, Agricultural Research Service, ARS 63, 372 pp. (1987).

## WASTE-SITE CLOSURE DEMONSTRATION -- A FIELD-SCALE STUDY

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*Funding Organization: DOE Defense TRU Waste  
Management Program*

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The long-term integrity of a waste disposal site depends strongly on our ability to manage the water balance of the trench caps. Precipitation is the driving force for the surface hydrology of the site, but the fate of water falling on the site will be determined by both the design and the long-term behavior of the cap. If vegetation or mulches afford insufficient protection of the soil surface, the impact of raindrops can erode the trench cap and expose the waste material or provide channels for increased infiltration of water deep into the cap profile. After the precipitation event, some of the water evaporates from the soil and vegetation surfaces. Infiltrated water can be stored in the soil profile, perhaps to be transpired later by the vegetative cover (and thus "mined" from the soil and removed from storage). Alternatively, the water can be transported laterally out of the trench cap profile by barriers and drains or it can seep down to contact the waste material.

In semiarid climates, most of the precipitation is removed from the system by the combined forces of evaporation from the surface and transpiration by the plant cover (evapotranspiration, or ET). Erosion in such climates is particularly severe in part because of localized storms of short duration and high intensity typical of these areas. A relatively small change in ET can radically affect deep seepage or percolation of the water below the root zone in the trench cap.

In managing the components of the water balance of a site, we must understand the effects of a mulch that, while preventing excessive

erosion, may increase infiltration of water under some conditions. Increased water infiltration will likely result in increased plant growth (particularly in dry climates), increasing the ET from the site. Whether the higher ET can offset the increased infiltration of water should be determined along with studies on the most appropriate soil profiles and vegetative cover for minimizing soil water content and surface erosive potential at critical times of the yearly hydrologic cycle.

In FY 1987 we initiated a demonstration project to study the interactive effects on the water balance of surface mulches, vegetative cover, and soil profile design. The initial phase of the demonstration consists of a series of 12 plots installed on an inactive mixed-waste site, Area B. We are collecting data on water balance and trench cap performance for each plot under natural precipitation. After analyzing the data, we will design and apply an optimum cover to an entire site at Los Alamos with follow-up monitoring to assess cover performance.

At Area B, runoff from plots with four different surface treatments (shrubs and grass covers, both with and without a gravel mulch treatment) has shown that in both 1987 and 1988 the presence of a gravel mulch significantly decreased the runoff and erosion resulting from the intense summer storms (Table I). Decreased runoff implies increased infiltration of water and thus a subsequent increase in water available for plant growth and ET. To test this hypothesis we calculated the water balance to estimate the combined evapotranspiration and deep seepage (ET + L) components. These mechanisms removed a significant 4.6 cm more water per year from the mulched plots than from the unmulched plots. Inspection of soil moisture data at depths to 100 cm suggests that little seepage occurred and that this increase is attributable primarily to ET. Overall sediment concentrations in the runoff were much higher in 1987 than in 1988, probably resulting from progressive stabilization of the surface.

Table I. Runoff and Sediment Transport on Area B Plots.						
Treatment	Runoff Fraction <sup>a</sup>		Sediment Concentration <sup>b</sup> (g/l)		Total Sediment Load <sup>c</sup> (Mg/ha)	
	1987	1988	1987	1988	1987	1988
Shrub + Mulch	0.02	0.11	5.06	0.49	0.09	0.29
Shrub	0.20	0.22	6.99	1.34	1.44	3.25
Grass + Mulch	0.03	0.10	4.33	0.48	0.11	0.82
Grass	0.11	0.27	6.47	1.07	0.68	3.93

<sup>a</sup>Mean runoff as a fraction of precipitation over 21 events in 1987 and 32 events in 1988.  
<sup>b</sup>Mean sediment concentration for 1987 and 1988 precipitation events.  
<sup>c</sup>Total sediment load from 32.54 m<sup>2</sup> plots calculated on per hectare basis.

The effect of vegetation type on runoff, erosion, and ET + L is more variable. In 1987, a significantly higher fraction of runoff was observed from unmulched plots with a shrub cover than from the unmulched grass plots. This effect was not seen in 1988, partly because of the extremely high precipitation that year. However, over both years the shrub vegetation had a significantly higher ET + L (3.8 cm annually) than did the grass vegetation. These small but statistically significant differences could result in appreciable differences in the amount of water infiltrating into a trench cap

and eventually penetrating below the root zone toward the waste.

The demonstration study at Area B is providing us with a valuable data base on the effects of natural precipitation on trench cap performance. After another year, we will analyze the data for the interactive effects of soil profile, soil surface, and vegetative and location factors on performance. We will then compare our results with those from erosion studies using a rainfall simulator for controlled precipitation inputs.

## REACTIVE TRACER TESTS IN THE C-WELLS

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*Funding Organization: DOE Nevada Operations  
Office Yucca Mountain Project Office (YMP)*

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The Yucca Mountain Project Office (YMP) has assigned to Los Alamos the primary responsibility for geochemical characterization of the candidate high-level waste repository site at Yucca Mountain, Nevada. This characterization includes mineral composition of geologic material, solubility and adsorption of important radionuclides, and diffusion and transport characteristics of the nuclides. Because many of the experiments will be conducted in the laboratory, some concern has been expressed about extrapolating laboratory results to the site.

The study is divided into laboratory and field phases. The laboratory experiments were initiated at Los Alamos in 1987 and the field experiments are planned for the C-well complex at Yucca Mountain.

This study is an initial attempt to determine the validity of laboratory-derived parameters when applied to a field-scale experiment. A single process, adsorption, will be examined. In our attempt to understand the adsorption phenomenon and to extrapolate the results to the field, we decided to take a mechanistic approach. Adsorption is defined in many ways in many different disciplines. As a result, initial efforts have concentrated on defining the adsorption mechanisms and on identifying laboratory experiments to determine the adsorption of a particular solute by a prevailing mechanism. These efforts involved literature reviews and interactions

with experts in soil science, aquatic chemistry, and chemical engineering.

Two adsorption mechanisms are potentially important at Yucca Mountain: (1) physical and (2) chemical. In the laboratory, we plan to identify at least one tracer for each of the two mechanisms, one that is adsorbed predominantly by that mechanism. Also, the laboratory experiments will provide the parameters for the appropriate sorption expression to be used with transport models to describe the field experiment.

Lithium is a potential tracer that is adsorbed predominantly by the physical mechanism. The Prow Pass Member of the Crater Flat Tuff is one of the zones in which the proposed C-well field experiments may be conducted. Using tuff samples from this member, we conducted laboratory experiments to characterize lithium and confirm the prevailing adsorption mechanism.

We performed both batch equilibrium and kinetics experiments at temperatures of 25°C to 45°C and initial lithium concentrations of 1 to 2000 g/mL. The kinetics experiments revealed that lithium equilibrated rapidly (within a couple of hours), but a 24-h period was used for all equilibrium experiments.

Figure 1 shows the measured concentration in solution (C) versus the adsorbed concentration (S) for equilibrium experiments conducted at 38°C. These data are used to determine parameters for the four adsorption expressions selected for use in this study---Linear, Langmuir, Freundlich, and Modified Freundlich. Inconsistencies in our measurements of the lithium concentration led us to use an alternative method for calculating the adsorbed lithium. Figure 2 compares data obtained by this method with the original measured data. The lines represent the fit of the data with the modified Freundlich isotherm. The closeness of the two lines suggest that the parameter values are similar. However, errors associated with the values obtained by the alternative technique are much smaller than those associated with the observed data.

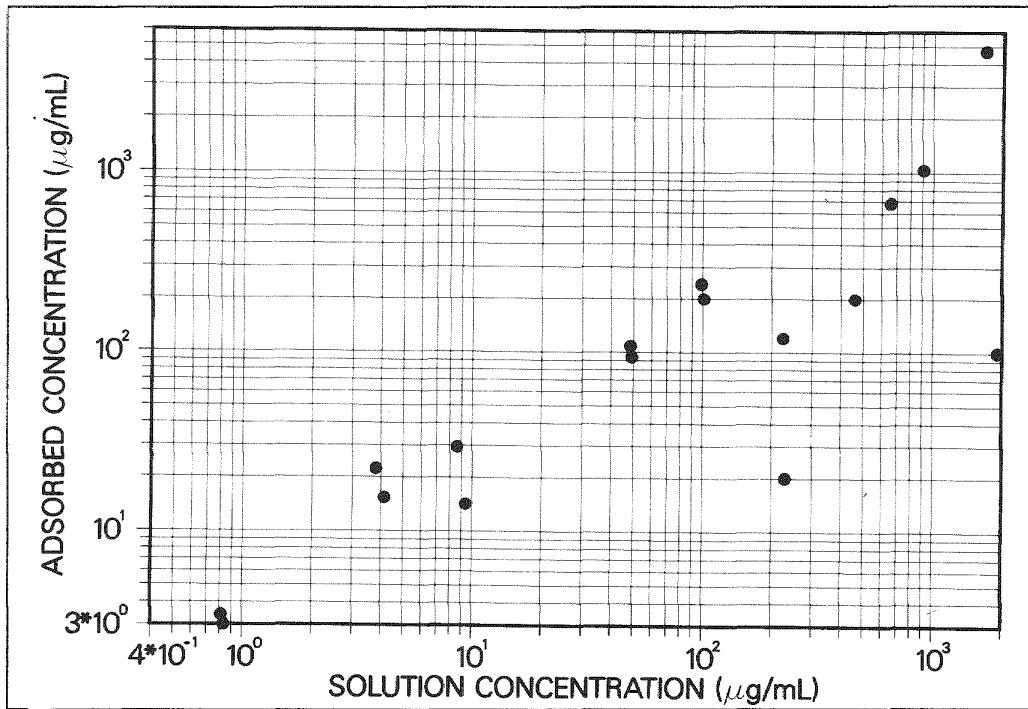


Figure 1. Observed and predicted adsorption of lithium on a Prow Pass suspension in J-13 well water at 38°C for an initial concentration range of 1 to 2000  $\mu\text{g mL}^{-1}$  and a solid:liquid ratio of 1:20. Adsorbed lithium is determined in two ways: (1) based on measured lithium (Li) in solution and (2) based on a combination of measured lithium and other cations in solution (best estimate). Predicted lithium is based on the Modified Freundlich isotherm.

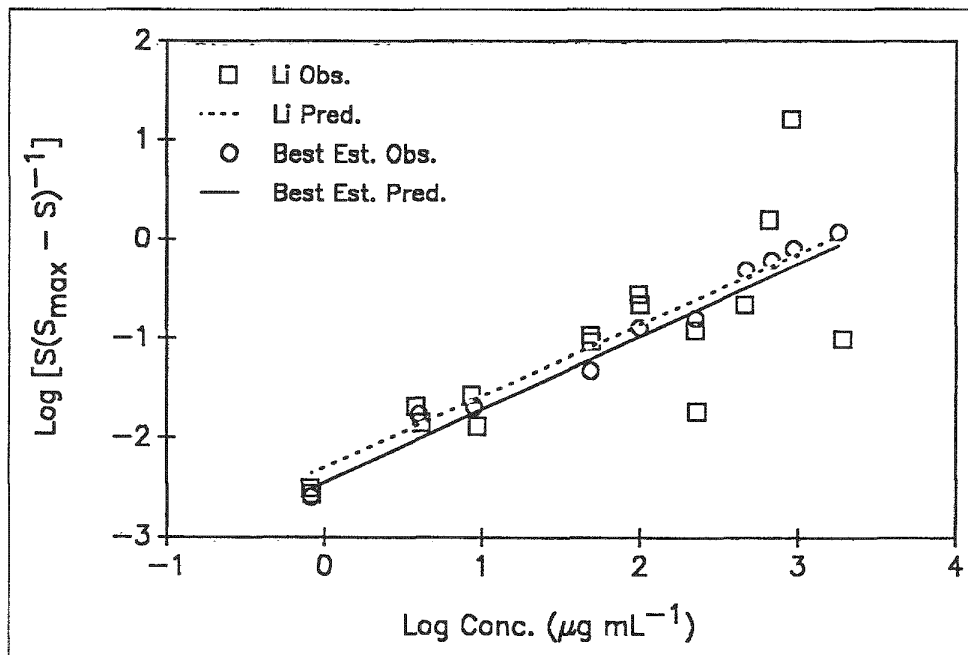


Figure 2. Observed and predicted adsorption of lithium on a Prow Pass suspension in J-13 well water at 38°C for an initial concentration range of 1 to 2000  $\mu\text{g Li mL}^{-1}$  and a solid to liquid ratio of 1:20. Adsorbed lithium is determined two ways: (1) based on measured lithium in solution (Li) and (2) based on a combination of measured lithium and other cations in solution (best estimate). Predicted lithium is based on the Modified Freundlich isotherm.

Potentiometric titration was another technique used to characterize lithium adsorption behavior in these experiments. This technique measures residual proton concentration resulting from addition of a known amount of potential-determining ion ( $H^+$  or  $OH^-$ ). These titrations are conducted initially with no tracer (such as lithium) added and then with tracer added. If the zero point of charge (ZPC) shifts after the addition of the

solute, the adsorption mechanism may be chemical. If the ZPC does not shift, then the mechanism may be physical. Figures 3 and 4 show the results of these experiments. At  $38^\circ C$ , the ZPC ( $S_o k^{-1} = 0$ ) did not shift after  $900 \text{ g mL}^{-1}$  of lithium were added. This is another piece of evidence that confirms lithium adsorption is governed predominantly by the physical mechanism.

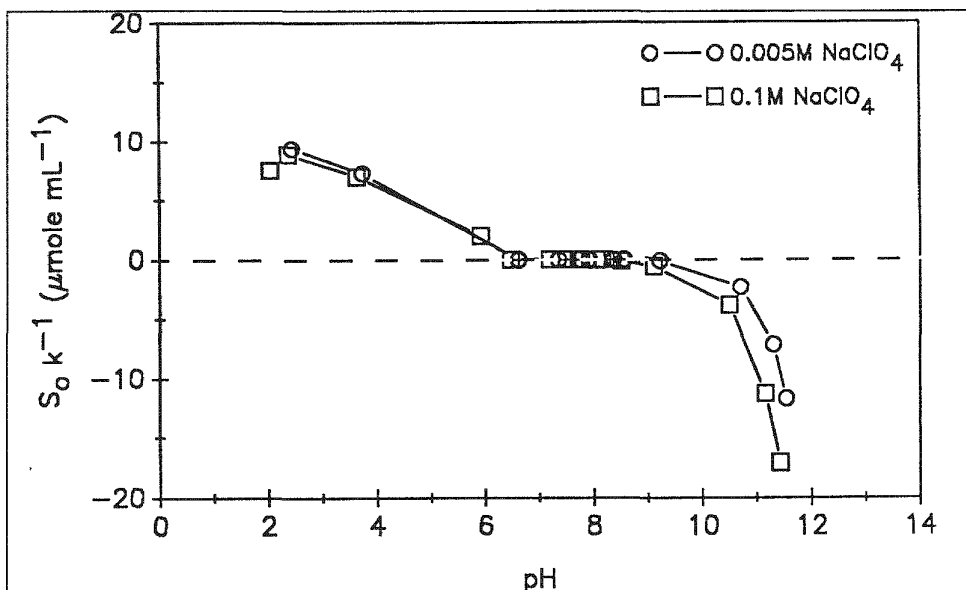


Figure 3. Potentiometric titration of Prow Pass in suspension with J-13 well water and  $\text{NaClO}_4$  electrolyte at  $38^\circ$  without the addition of lithium.

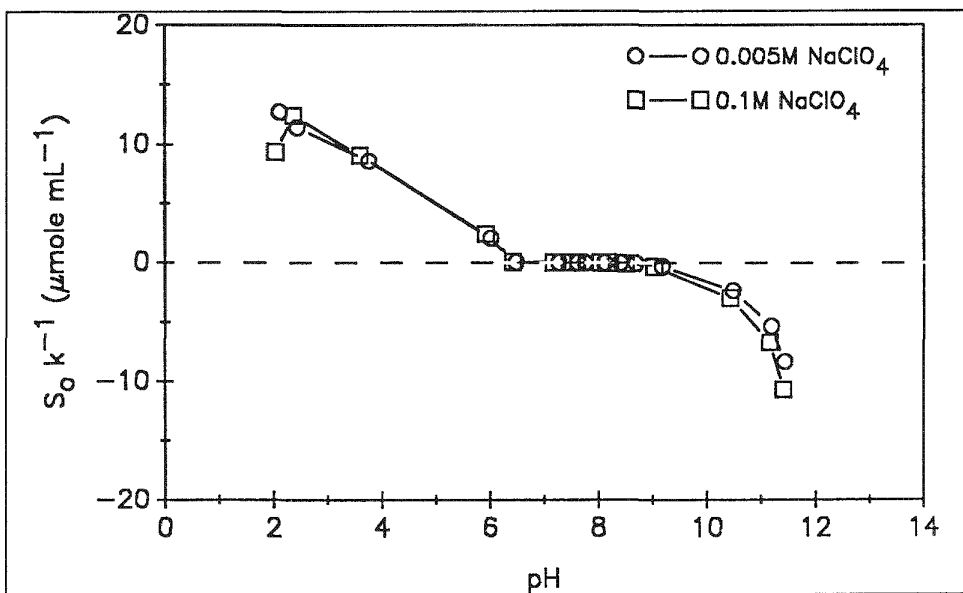


Figure 4. Potentiometric titration of Prow Pass in suspension with J-13 well water and  $\text{NaClO}_4$  electrolyte at  $38^\circ$  with the addition of  $900 \mu\text{g Li mL}^{-1}$ .

## ***SURFACTANT FLOODING TECHNOLOGY FOR IN SITU CLEANUP OF CONTAMINATED SOILS AND AQUIFERS - A FEASIBILITY STUDY***

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*Author: C. Porzucek*

*Group: Environmental Science, HSE-12*

*Funding Organization: ISR*

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In November 1987 Los Alamos began a feasibility study to investigate the use of surfactant flooding to clean up contaminated aquifers and soils. Such technology is applicable to, for example, fuel spills. Some spills occur in areas that are impossible to excavate and decontaminate. For these areas some form of in situ cleanup is necessary.

A nonaqueous-phase liquid (NAPL) can be trapped in soils by capillary forces and sorptive forces. Work on surfactant flooding in enhanced oil recovery has shown that surfactants can reduce the capillary forces by several orders of magnitude, making it easier for a flooding solution to displace the NAPL phase. NAPL can also be trapped in soils by sorption onto mineral surfaces or by association with organic matter.

I estimate that approximately 10% of a TCE phase will sorb onto the surfaces of an aquifer containing 3.0% organic carbon. The effect of a surfactant on sorbed contaminants is unknown. Current research efforts with Howard University (through the HBCU program) will give more insight into this effect. If a surfactant has no effect on sorbed contaminant, then surfactant flooding will never be able to remove all the contaminant from the soils. For the hypothetical soil just mentioned, 10% of the TCE will be left behind even if the surfactant flood is 100% successful at displacing TCE held by capillary forces.

Surfactants can also displace NAPL by increasing the solubility of the aqueous phase. In one study by General Electric,<sup>1</sup> a particular surfactant increased the aqueous solubility of the PCB Arochlor 1260 by a factor of  $10^4$  to  $10^5$ .

Unfortunately, field observations show that the increased solubility phenomenon is limited by mass transfer (aqueous NAPL concentrations are less than expected in the presence of surfactant at equilibrium conditions).

Hunt et al.<sup>2</sup> have presented a model for the dissolution time of a NAPL in ground water. The effect of surfactant was incorporated into this model to study its effect on the dissolution times. Calculations were performed for different-size droplets of TCE under different ground water flow velocities. Figure 1 shows results for a 10-cm droplet. In this figure, the effect of surfactant is shown by an increased aqueous TCE solubility. The curve representing a ground water velocity of 0.1 cm/day shows that, in the absence of surfactant, a TCE drop may require up to 30 years to dissolve. However, in the presence of surfactant the dissolution time can be less than a year. Other results are found in the FY88 ISR report.<sup>3</sup>

Remedial action consisting of in situ surfactant flooding is risky from several standpoints. The phase behavior (single phase, multiple phases, gels, precipitates, and so forth) of surfactant systems is sensitive to temperature, pH, and NAPL concentration. In an in situ situation, it may not be possible to control all variables so as to achieve desirable phase behavior. Unknown heterogeneities in the soils can also make the flood risky. In addition, the process can be used only on soils having desirable physical properties. Disposal of effluent waste from the flood is also a problem. From past field work on the process, no successful method has been developed to separate the NAPL from the aqueous phase. Finally, the surfactant is an expensive chemical. Therefore, the cost of the process must also be weighed against other available remedial actions.

One area in which surfactant-enhanced soil washing could be more attractive is in vadose zone cleanup. Previous work has shown that the method does drain the capillary zone of NAPL.<sup>4,5</sup>

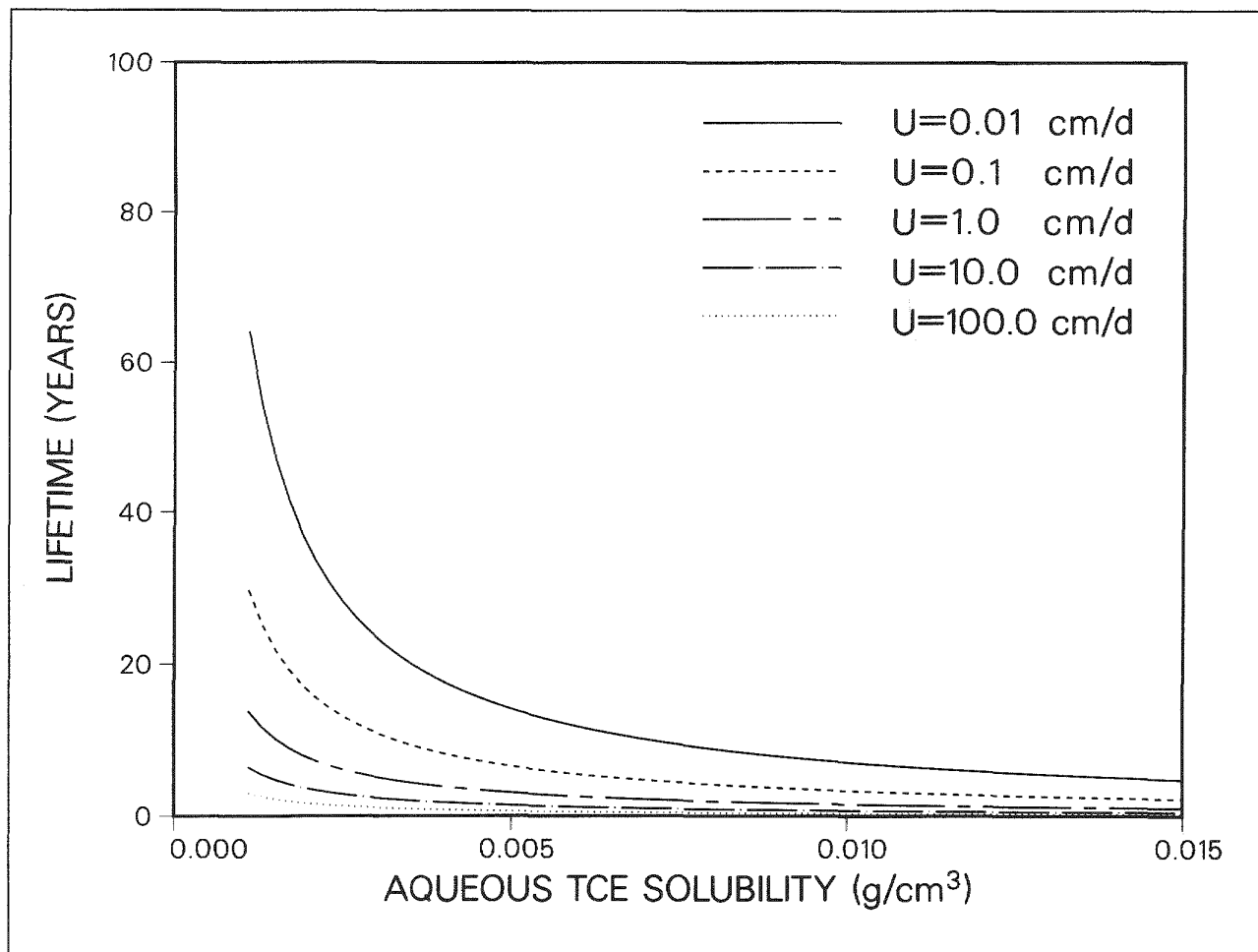


Figure 1. Droplet lifetimes for a 10-cm TCE droplet in an aquifer for different aqueous-phase TCE solubilities and different ground water flow velocities.

Finally, surfactant-enhanced soil washing does not decontaminate soils to within the EPA standards. Reported recoveries of residual oil from two oilfield surfactant flooding projects are 25% and 25.4%, respectively.<sup>6,7</sup> In the latter case, the recovery was only 38% to 43% of the value predicted from laboratory core flood experiments. Therefore, it seems logical that a surfactant flooding remedial action will have a similar recovery. Surfactant-enhanced soil washing must be combined with some other cleanup method if the soil is to be decontaminated to within the EPA standards. Bioremediation seems a logical remedial action to follow surfactant-enhanced soil washing.

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## **QUALITY ASSURANCE FOR ENVIRONMENTAL RADIONUCLIDE ASSESSMENT PROGRAMS AT THE NEVADA TEST SITE**

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*Funding Organization: DOE Nevada Operations Office, Environmental Protection Division, Environmental Science and Technology Branch*

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This past year the Environmental Science Group participated in two programs sponsored by DOE/NV dealing with evaluating radionuclide inventory, distribution, and environmental compliance and monitoring at the Nevada Test Site (NTS).

The first is the Radionuclide Inventory and Distribution Program (RIDP), which concluded in assessing the radionuclide distribution and inventory of Areas 5, 11, 12, 15, 17, 18, 19, 25, 26, and 30. LANL not only provided consultation in soil sampling, preparation, analysis, and interpretation but also designed and conducted the quality assurance program required for DOE-sponsored programs.<sup>1</sup> Results of the study and the quality assurance task are documented in the report by McArthur and Mead.<sup>2</sup>

The second is the Basic Environmental Compliance and Monitoring Program (BECAMP), conducted by participating experts in environment, modeling, particle resuspension, soil, mammology, desert vegetation, collection, and analysis. LANL provides consultation in soils oriented studies, for example, sampling, analysis, and data interpretation, and designs and conducts the required quality assurance program.<sup>1</sup> During this past year the major task was to prepare a quality assurance plan.<sup>3</sup> Participants were instructed in required aspects of quality assurance so that data collected or derived in support of environmental studies and radionuclide monitoring would be acceptable and defensible.

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3. E. H. Essington, "Quality Assurance Plan for the Basic Environmental Compliance and Monitoring Program (BECAMP)," Los Alamos National Laboratory report, Draft (1988).

## **DEVELOPMENT OF A SOLAR-BLIND RAMAN LIDAR FOR EVAPOTRANSPIRATION MEASUREMENT**

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*Funding Organization: DOE, Office of Health and Environmental Research, Ecological Research Division*

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At the Los Alamos National Laboratory we are developing a remote optical sensing tool to monitor the rate of emission of water from plants and soil. The mobile uv solar-blind LIDAR (light detection and ranging) system will be capable of remote, noninvasive measurement of water vapor concentration in three dimensions. Rapid scanning of the selected volume of air will permit future software development to correlate the observed patterns from successive time scans and then calculate both the horizontal and vertical wind vectors. The patterns are created from rising eddies of heated moist air. The LIDAR offers the possibility of developing an instrument that can bridge the gap in the measurement scales between localized point measurements and broad-scale estimates from satellite sensors (that cover from several kilometers to global dimensions).

The LIDAR system can be immediately applied to various areas of atmospheric research, including the study of evapotranspiration (ET), and will be more versatile than point monitors. A LIDAR-based instrument will permit (1) remote operation at places inaccessible to point monitors; (2) rapid data acquisition at rates exceeding 100 000 bits/s with measurements that are nearly instantaneous and repeatable up to many times per second; (3) noninvasive measurements that disturb neither the atmospheric nor surface environments; (4) high spatial resolution with better than 5-m resolution along the laser axis; (5) high

precision and sensitivity H<sub>2</sub>O number density; and (6) three-dimensionality allowing pattern recognition of eddies, turbulence, and diffusion.

In 1988, the system was capable of acquiring only one-dimensional arrays of water vapor concentration vs range as a function of time. The azimuth and elevation were determined solely by selecting the truck orientation. Data acquisition and transfer to permanent storage had been tested only over relatively short periods of field tests and was only capable of 1.2-Hz operation, and not at the 10-Hz laser rate we can now achieve through direct memory access.

In June 1988, we participated in the third collaborative experiment sponsored by the U.S. Water Conservation Laboratory at the University of Arizona Maricopa Agricultural Center (MAC) south of Phoenix. These experiments are designed to study surface-atmosphere interactions, with the ultimate goal of using remotely sensed data for agricultural management purposes. The opportunities to use the uv LIDAR in its limited operational mode was seen as an advantage in enabling us to test and debug the system, verify absolute calibration with other point sensors, and gain the experience of acquiring data for long periods of time. It proved extremely useful in this and demonstrated the ability to see moving atmospheric eddies of water vapor.

During the MAC experiments we obtained both vertical and horizontal scans along linear paths above various crops. An example (Fig. 1) showed a rise in water vapor from 100 to 250 m from the truck, and a gradual decrease at longer ranges. Spatial resolution of 1.5 m allowed us to detect small spatial changes in water vapor concentration. By plotting successive scans (Fig. 2) we observed temporal changes that reflect the typical midday changes in water vapor above an actively transpiring crop as well as the rapid changes in water vapor concentrations caused by horizontal eddy movements. These eddy movements are not observable using traditional devices.

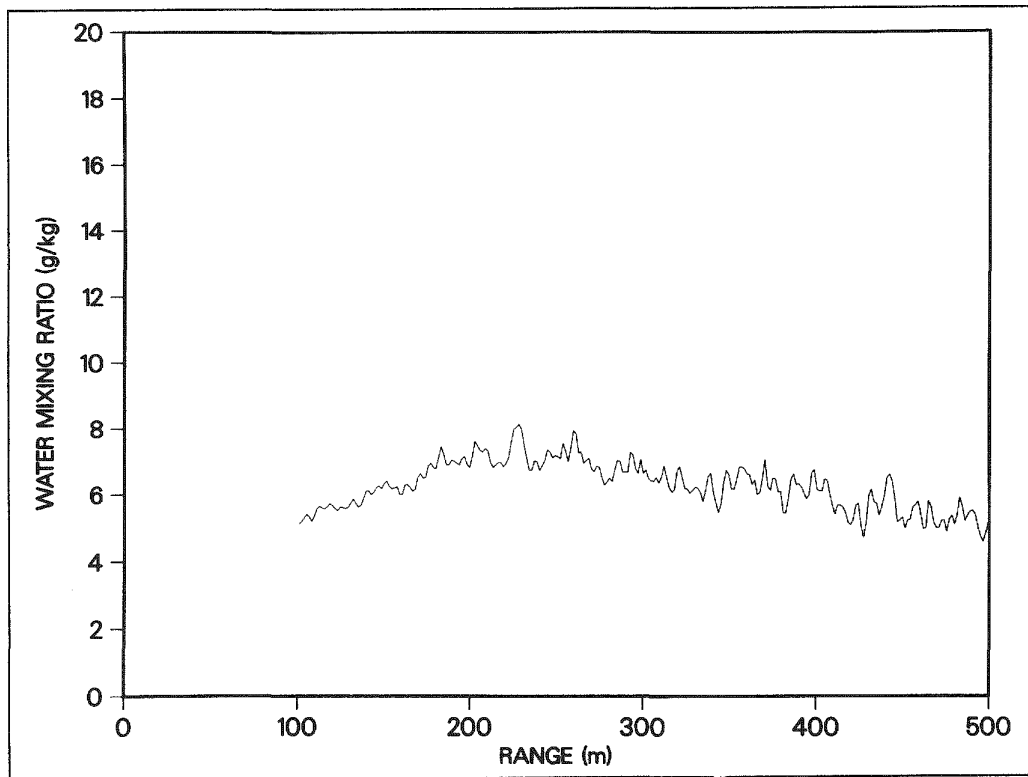


Figure 1. Example of water vapor concentration as a function of range at 1145 h, 13 June 1988.

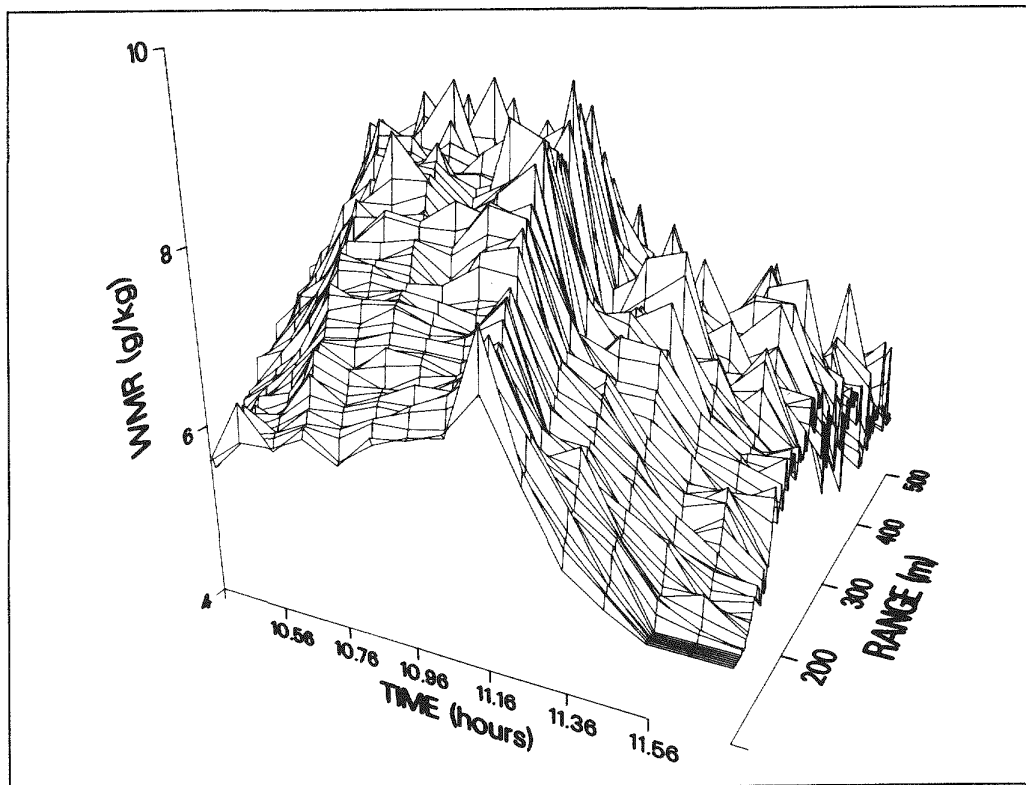


Figure 2. Water vapor concentration (WMR) as a function of range from 1000 h to 1200 h, 13 June 1988. Five point smoothing over both time and range was used to facilitate display.

A series of tests was designed to test the LIDAR's ability to detect small vertical gradients in water vapor close to ground level. Using the hydraulic jacks on the outside of the truck, the truck orientation was changed so that the LIDAR was aimed at the upper and lower arms (2.3 and 0.8 m above ground level) of a Bowen ratio micrometeorological station situated 180 m from the truck. The vertical gradients as measured by the LIDAR from 1205 to 1240 h (+0.35 to -0.55 g/kg) were comparable to those measured by the instrumentation on the meteorological tower (-0.43 to -0.53 g/kg). The higher variability in the LIDAR data is expected since each file records the average concentration over 30 to 40 s, in contrast to the 20-min averages from the tower instruments. A fast-moving downward burst of dry air apparently occurred during the tests, resulting in the temporary positive gradient measured by the

LIDAR. The long averaging time of the meteorological instruments smooths out such high frequency events.

In June 1988, we did not have an expedient display technique to process and display the copious quantities of data the LIDAR produces. However, our results suggest that the LIDAR is very capable of detecting moving parcels of air with distinctive water vapor concentrations. After azimuth and elevation scanning hardware is installed in early 1989, we will be able to accurately locate eddy cells at different times using software currently under development. From the spatial movement of these cells over time, we will be able to calculate localized moving cell velocities, including vertical vectors. This will enable calculation of water vapor flux rates.



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# DOSIMETRIC CHARACTERIZATION OF AN ALPHA-PARTICLE EXPOSURE FACILITY FOR RADIOBIOLOGICAL STUDIES

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*Funding Agency: LANL and DOE-OHER*

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## ABSTRACT

HSE-1, in collaboration with LS-4, has supported the design, construction, and dosimetric characterization of an alpha-particle exposure facility for radiobiological studies. Basic dosimetric quantities have been determined and the exposure facility is now operational. This paper describes the radiation source, exposure mechanism, dosimetry theory, and dosimetry results. Future work will include microdosimetric characterization of the source and enhanced quality assurance procedures.

## INTRODUCTION

Studies of effects from alpha-particle radiation on cultured cell populations have received increased interest in recent years. Two developments in the 1980's - risk from radon inhalation and the "reverse dose rate effect" (that is, increased radiobiological effects at lower dose rates, Roberts and Goodhead 1987) - may have implications for radiation protection guidelines. The importance of these developments and how they affect applied health physics will depend upon how well cellular damage mechanisms and biological risk from alpha-particle irradiation are understood.

Determination of dose rate effects on cell systems from neutron and charged-particle interactions will directly affect the quality factor (Q) used in the estimation of dose equivalent. Recognition that inhalation of radon progeny contributes substantially to the radiation dose of the general population results in a need to quantify risks associated with radon environments. The primary contribution to radiation induced risk from radon inhalation is thought to be exposure of the

respiratory tract to alpha particle emissions from the decay of radon progeny.

Radiobiological studies aimed at these questions are currently in progress at LS-4, and are based upon exposing cultured cell populations to an alpha particle source. These investigations require a well-characterized source of alpha particles with minimum gamma contamination. This article describes the construction and dosimetric characterization of a Pu-238 alpha irradiator in use at Los Alamos.

## DOSIMETRY: THEORETICAL CONSIDERATIONS

Dosimetric and microdosimetric parameters are estimated from measurements of the energy spectrum and particle fluence of the alpha particle beam. In this investigation the concept of continuous slowing down approximation (CSDA) was used. CSDA implies that alpha particles lose their energy continuously at a rate defined as stopping power (Attix 1986). Based upon CSDA, absorbed dose is obtained from the following (Carlsson 1985):

$$D = \frac{1}{\rho} \int_0^{E_{\max}} \phi(E) \cdot S(E) dE \quad , \quad (1)$$

where

$E$  = energy of the alpha particle;

$E_{\max}$  = maximum energy of measured spectra;

$D$  = absorbed dose;

$\rho$  = density of irradiated medium;

$\phi(E)$  = particle fluence at energy  $E$ ;

$S(E)$  = stopping power at energy  $E$  in irradiated medium.

The quantities  $E$  and  $\phi(E)$  should be measured for each irradiation configuration (Carlsson 1985, Eisen 1985). Values of  $S(E)$  may be obtained from published tables (Barkas & Berger 1964, Bichsel 1968, ICRP 1970, Janni 1982, Northcliff and Schilling 1970, Walsh 1970, Ziegler 1977). In the case of tables referring to protons, stopping powers are scaled by the following approximation (Bichsel 1968):

$$S_{\alpha}(E) = z_{\alpha}^2 S_p(E') \quad (2)$$

where

$S_{\alpha}(E)$  = stopping power of alpha particle;

$z_{\alpha}$  = the charge (atomic number of alpha-particle = 2);

$S_p(E')$  = stopping power of proton at equivalent energy;

$$E' = (Mc_p^2 / Mc^2) * E = (938/3727) * E = .25 * E.$$

This procedure is based on the following assumptions (Ziegler 1977):

1. The effective charge of the alpha particle is independent of the material traversed.
2. Effective charge is dependent only on particle velocity.
3. Protons and alpha particles display identical stopping mechanisms in similar materials.
4. All dielectric effects between electrons temporarily bound by the alpha particle and electrons in the medium do not contribute significantly to particle stopping.

When  $E < 1.6$  MeV an effective or average charge  $z_{\alpha}^*(E)$  should be used. As velocities decrease, an alpha particle will show increased transient binding of electrons, reducing its average charge (Bichsel 1968, Ziegler 1985). Therefore  $z_{\alpha}^*(E)$ , the expected charge of the alpha particle should be used in Eq. (2) for energies less than 1.6 MeV. Table I contains values of  $(z_{\alpha}^*(E))^2$ .

Table I. Expected Charge for Alpha-Particles (from Bichsel 1968).

E (MeV)	$(z_{\alpha}^*(E))^2$	E (MeV)	$(z_{\alpha}^*(E))^2$
0.2	1.8	0.9	3.4
0.3	2.0	1.0	3.5
0.4	2.4	1.2	3.7
0.5	2.8	1.4	3.9
0.6	3.0	1.6	4.0
0.7	3.2	1.8	4.0
0.8	3.3	2.0	4.0

Figure 1 contains a plot of the values in Table I. When using these values and any extrapolations or interpolations from them, it should be realized that these are approximations and care should be exercised in interpreting results.

Figure 2 compares mass stopping power in muscle,  $S(E)/\rho$ , as a function of particle energy from the works of Janni (1982), Walsh (1968), and Ziegler (1989). Appropriate scaling algorithms were applied to Janni's proton data. Note that the Walsh data has higher values for stopping power below 1.5 MeV. Walsh did not discuss and, presumably, did not account for the effect of transient electron binding on alpha particle stopping powers (Walsh 1968). Ziegler (1989) has provided a computer code based upon his method of stopping power calculation (Ziegler 1985). In this publication Ziegler addresses expected charge of low-energy ions, discussing how they are employed in his code. Note the data sets of Janni and Ziegler agree very well at all energies.

Based upon this result and the flexibility and convenience of the computer code, Ziegler's stopping power and ranges for tissue were used for all calculations. The range of an alpha particle in tissue is defined as the expected value of the path length it will follow until it comes to rest. Figure 3 is a plot of mass stopping power for alpha particle energies ranging from 10 keV to 4.5 MeV. Figure 4 is a plot of alpha particle range in tissue for energies ranging from 10 keV to 4.5 MeV. Data for both Figs. 3 and 4 were generated with Ziegler's code.

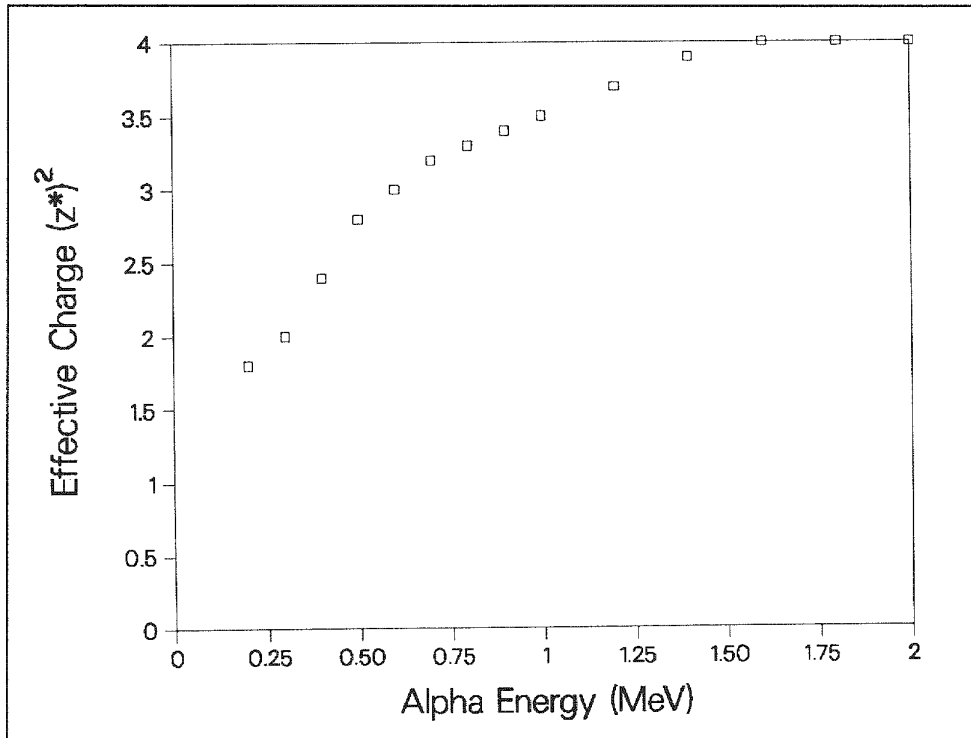


Figure 1. Expected charge as a function of alpha-particle energy (from Bichsel 1968).

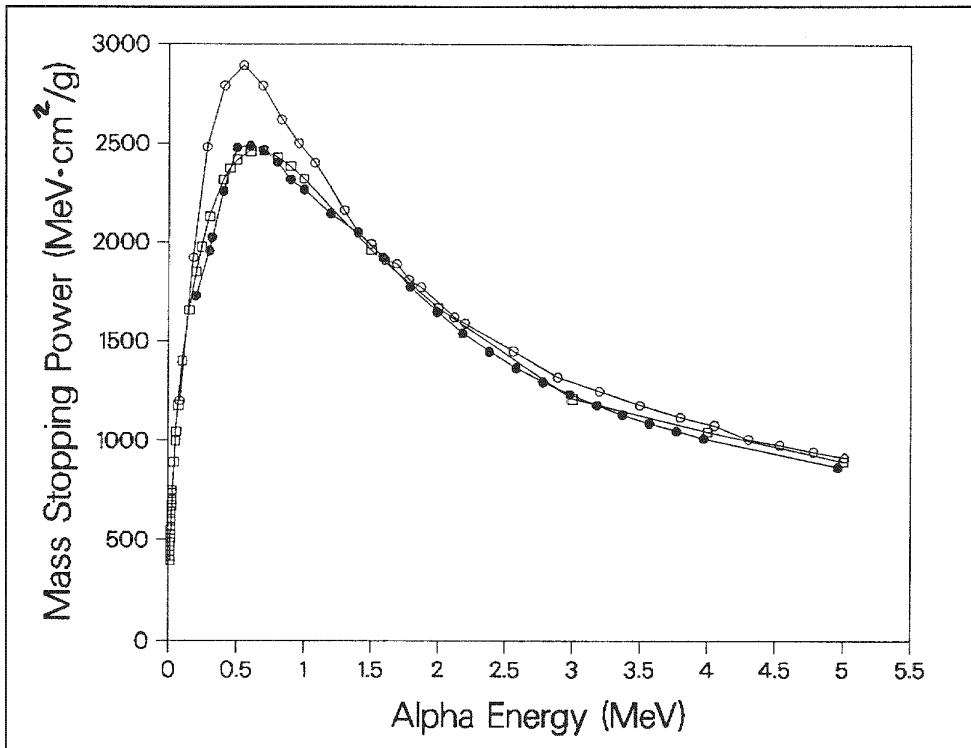


Figure 2. Comparison of mass stopping power as a function of alpha-particle energy from Walsh (1970)  $\circ$ , Janni (1982)  $\bullet$ , and Ziegler (1989)  $\square$ .

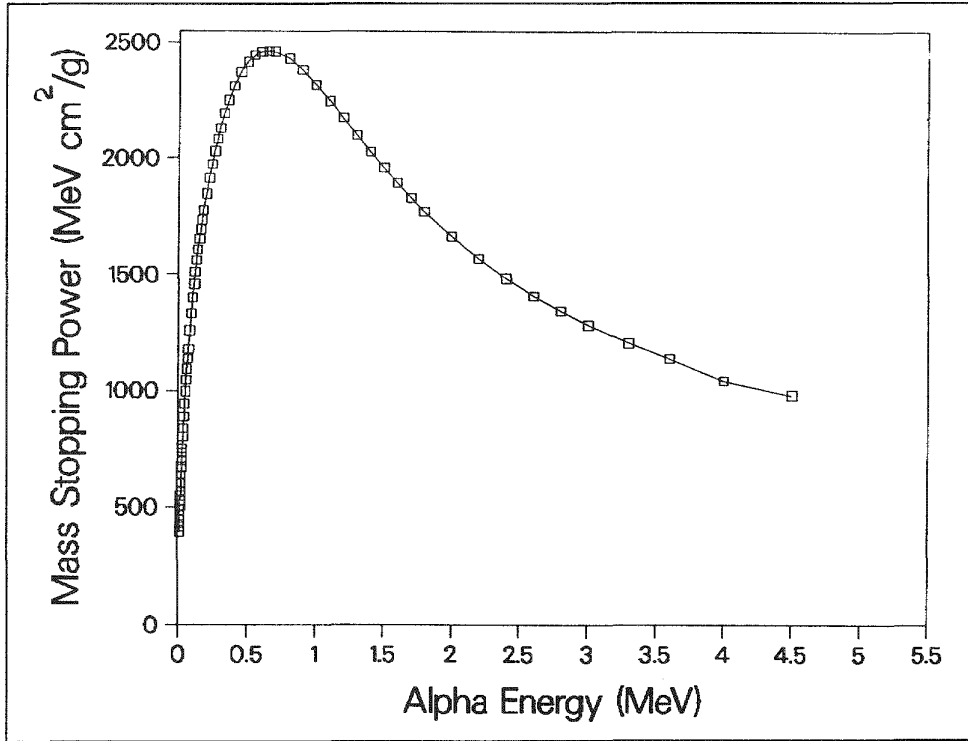


Figure 3. Mass stopping power as a function of energy, 10 keV to 4.5 MeV (from Ziegler 1989).

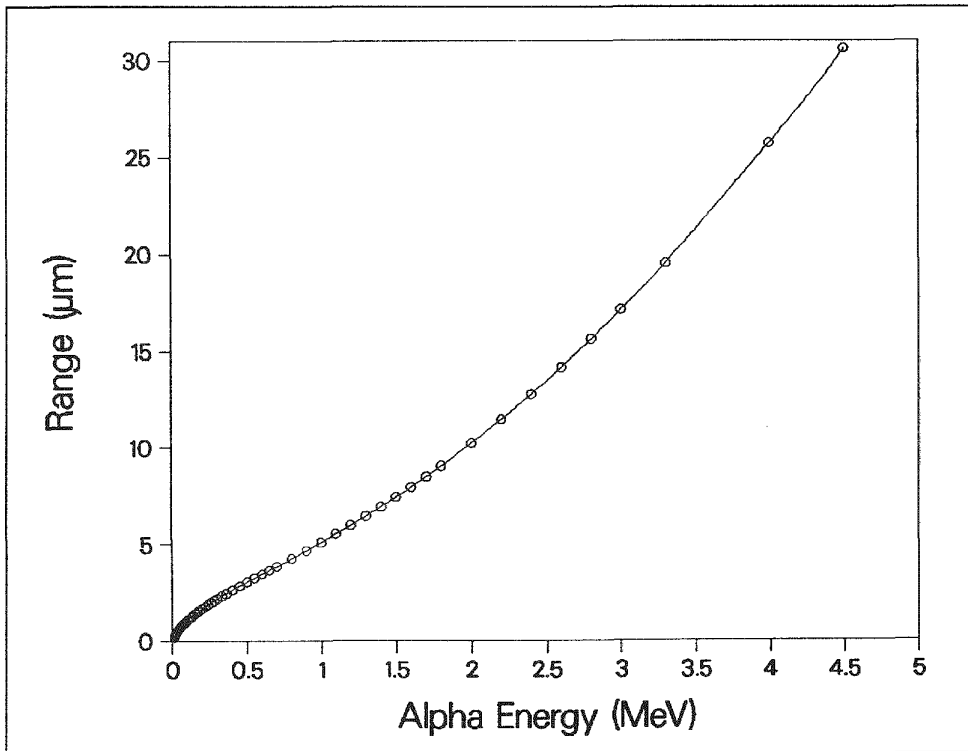


Figure 4. Mean alpha-particle range as a function of energy (from Ziegler 1989).

## DESCRIPTION OF THE ALPHA SOURCE

The radionuclide chosen as the alpha particle source was Pu-238. This nuclide has several physical parameters that make it a desirable source for an alpha-exposure system. These qualities include a high specific activity, a low photon yield, and low-energy photon emissions. The source is a 70-mm-diam, stainless steel disk with 3.9 mCi of Pu-238 uniformly electrodeposited on one side. A honeycomb aluminum collimator was placed on top of the active side of the disk to produce an approximate parallel alpha particle beam normal to the exit window. The source and collimator were placed in an aluminum housing with a 28-mm-diam, Mylar exit window for the particle beam.

To precisely control exposure times, a mechanical shutter was mounted directly above the exit window. The shutter was modified to support custom-fabricated tissue culture dishes with Mylar bottoms, parallel to the exit window and normal to the particle beam. Cells are grown directly on the Mylar providing a consistent irradiation geometry: each alpha particle encounters the same thickness of Mylar before interacting with a cell. Previous studies required alpha particles to penetrate varying thicknesses of residual cell culture medium before encountering a cell (Raju and Jett 1974). An electronic timer was coupled to the shutter mechanism for accurate measurement of exposure times. Table II contains specific technical information relating to the exposure system.

## DOSIMETRY PROCEDURES

Measurement of the distribution of energies and particle fluence were performed with a surface barrier detector. Because of the high particle fluence, the detector was fitted with a brass collar to support a 25- $\mu\text{m}$  thick, stainless steel disk with a laser cut, 1-mm-diam aperture that was 0.5 mm from the detector surface. Because of the physical dimensions of the detector, E and  $\Phi(E)$  were measured 3.3 mm from the position of the Mylar dish bottom during cell population exposures. The particle energy spectrum at the Mylar dish bottom position was calculated based upon Ziegler's range-energy relationships in air. A 1.5- $\mu\text{m}$  thick

Mylar disk was glued to the brass collar, between the aperture and particle source, to account for the Mylar dish bottom between the source and cell populations being exposed. Figure 5 shows the measured energy spectrum. Note the area under the spectrum represents the total particle fluence  $\Phi$  over the time interval the spectrum was measured:

$$\Phi = \int_0^{E_{\text{max}}} \phi(E) dE \quad (3)$$

Table II. Technical Data on Alpha-Particle Exposure System.	
<b>Source</b>	
Nuclide:	Pu-238
Half-life:	87.74y
Specific Activity:	17.1 Ci/g
Alpha Energies:	5.495 MeV 71.6%
	5.452 MeV 28.3%
	5.352 MeV 0.1%
Activity:	3.9 mCi, 140 MBq
Diameter:	70mm
Construction:	electro-deposition on stainless steel
Producer:	LANL
Collimator Material:	aluminum
Channel length:	7 mm
Channel diameter:	3 mm
<b>Exit Window</b>	
Material:	mylar
Density:	1.39 g/cm <sup>3</sup>
Thickness:	1.5 $\mu\text{m}$ (2.1 e-4 g/cm <sup>2</sup> )
Diameter:	27.8 mm
<b>Dish Bottom</b>	
Material:	mylar
Thickness:	1.5 $\mu\text{m}$
Distance to Source:	1.6 cm
<b>Alpha-Particle Beam</b>	
Mean Energy:	3.7 MeV (at cell-mylar interface)
FWHM:L	250 keV
Mean Energy Range:	22 $\mu\text{m}$
Mean Energy LET:	1.12 MeV*cm <sup>2</sup> /mg
Mean Fluence Rate:	1900 p/mm <sup>2</sup> *s $\pm$ 7%
Mean Dose Rate:	3.4 rad/s (at cell-mylar interface)
<b>Gamma Component</b>	
Photon Energies:	L X-ray 13.6 keV 0.7%
	L X-ray 17.1 keV 1.0%
	L X-ray 20.3 keV 0.2%

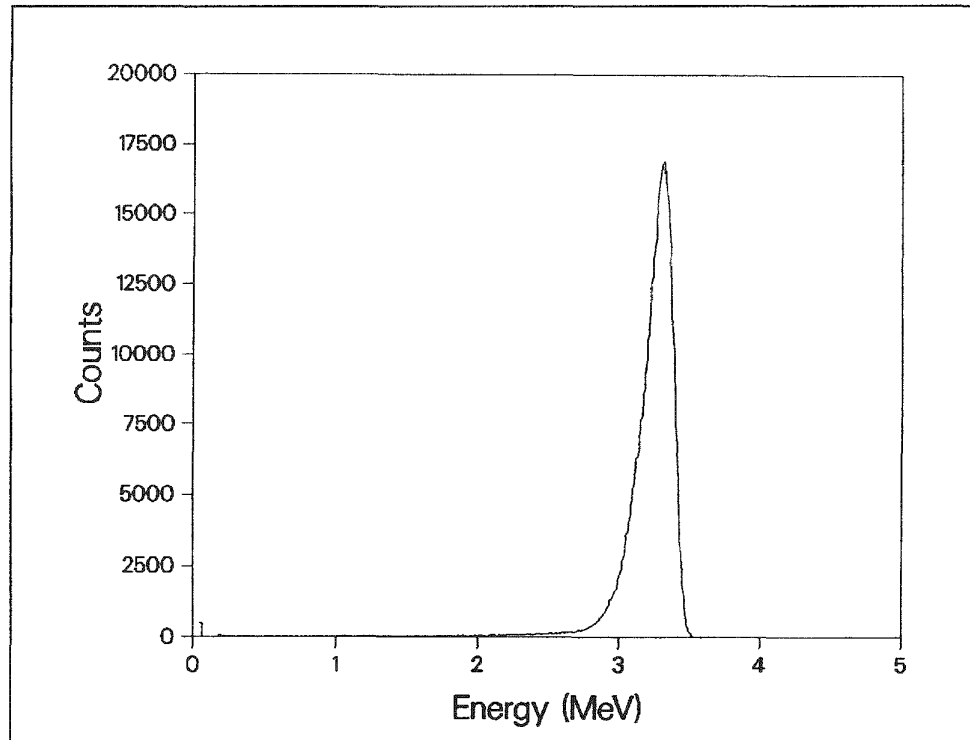


Figure 5. Measured energy spectrum of alpha-particle exposure system.

For purposes of verification,  $\Phi$  was measured utilizing CR-39 track-etch technology. This procedure eliminates edge effects from the stainless steel aperture and provides a quantitative measure of source uniformity. CR-39 foils, 28 mm in diameter, were placed in the exposure well at the same position as the Mylar dish bottoms during biological exposures. Foils were exposed for known time intervals, etched in 6.5 N, 60 C KOH, for 1.5 h. After etching, a CR-39 foil will contain pits on the surface exposed to the beam at each location where an alpha particle interacted with the foil. The number of pits per unit area is a measure of  $\Phi$ . Pits were counted using a light microscope coupled with a Biotran colony counter. The average fluence rate measured over the entire exit window area was  $1900 \text{ p/mm}^2\text{s} \pm 7\%$ . Particle fluence as a function of distance from the center of the exit window was evaluated to identify any shading resulting from shutter effects.

No trends were noted in this analysis, indicating that the particle fluence was uniform during exposure procedures. Results from the surface barrier measurements were collected on a multichannel analyzer (MCA). In this case,  $\phi(E)$  is represented by the number of net counts stored in

the channel corresponding to energy E. Based upon the discrete nature of the measured energy spectrum, absorbed dose is obtained from a modification of equation (1):

$$D = \sum_{i=1}^n \frac{1}{\rho} \phi_i S_i \quad (4)$$

where

D = absorbed dose;

$\rho$  = density of the absorbing medium;

$\phi_i$  = number of net counts in channel i;

i = channel corresponding to energy E;

$S_i$  = stopping power for alpha-particle of energy E;

n = channel corresponding to  $E_{\text{max}}$ .

The mean energy of the energy spectrum was estimated by

$$\bar{E} = \sum_{i=1}^n E_i / I, \quad (5)$$

where

$\bar{E}$  = Mean energy of the energy spectrum;

$E_i$  = energy corresponding to channel  $i$ ;

$n$  = channel corresponding to  $E$ ;

The mean energy,  $\bar{E}$ , was used in Ziegler's TRIM-89 (Ziegler 1989) code to obtain mean energy stopping power or LET and the mean energy range. The value  $\bar{E}$  was used in a Monte Carlo portion of the TRIM-89 software to estimate the linear dose-depth distribution in tissue. This distribution is shown in Fig. 6.

## DISCUSSION AND CONCLUSIONS

The measurement results and computed dosimetric quantities reported in this paper have been used to design radiobiological studies now under way in LS-4. Estimation of microdosimetric quantities, such as an energy-spectrum weighted dose-depth distribution, is planned before interpretation and publication of the results from these biological studies. Measurements with an ionization chamber will be performed to verify the dosimetric results presented here.

The choice of stopping range tables was based upon the agreement of two sets of recently published data (Janni 1982, Ziegler 1985). Investigation into differences between other stopping range tables and those used here are in progress. These results will be of particular importance for intercomparison of biological results from different laboratories and exposure systems.

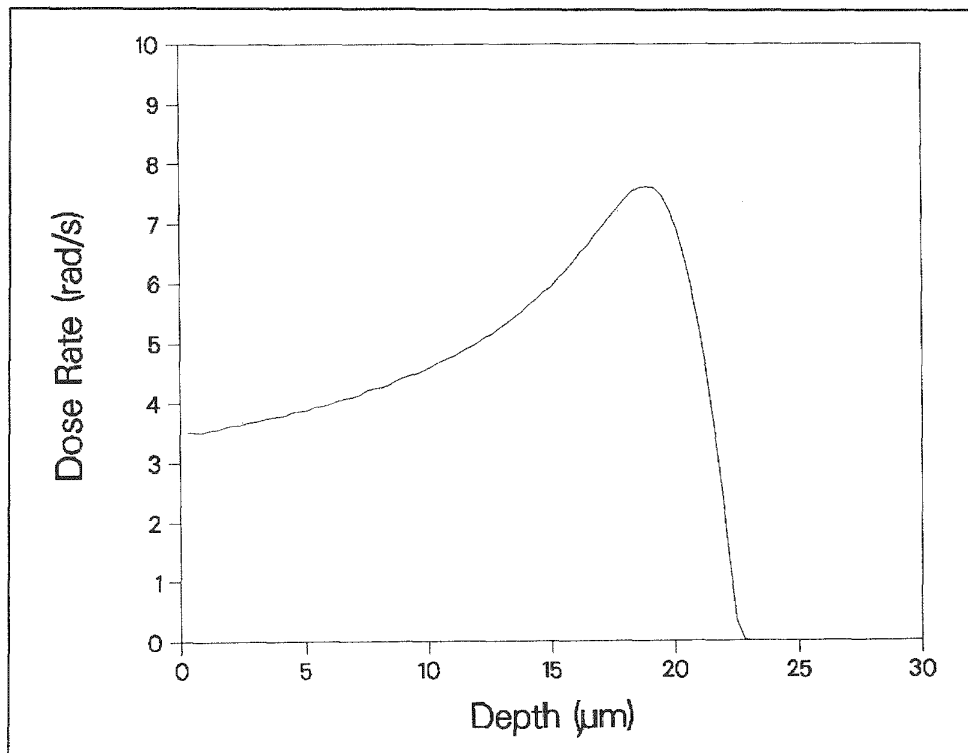


Figure 6. Estimated mean energy dose rate as a function of depth in tissue for the alpha-particle exposure system.

Work based on positioning of the Bragg peak at a known depth in a cultured cell population has been proposed. These studies will use varying thicknesses of Mylar filters to degrade the alpha particle beam energy. Dosimetry for the Mylar filters is under way.

CR-39 measurements are performed on a periodic basis as a quality control procedure for the exposure system. This protects against undetected changes in particle fluence caused by media spillage during biological exposure procedures. Uniformity and size relationships of CR-39 pits will be used in future work as a qualitative measure of the energy spectrum.

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## EVALUATION OF INTERNAL DOSIMETRY CODES

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*Funding: Los Alamos National Laboratory*

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### INTRODUCTION

Internal dosimetry codes are being reviewed, intercompared and compared to current Los Alamos methods. To date three codes have been evaluated for one radionuclide, plutonium 239, and one mode of exposure, inhalation. All three codes are based on International Commission on Radiological Protection Publication 30 (ICRP-30)<sup>1</sup> dosimetry methodology. The evaluation is being performed to (1) understand the various methods used to estimate intake of radionuclides and assign internal dose, (2) determine acceptability for use in the LANL internal dosimetry program (specifically for meeting new DOE dose assessment and reporting requirements) and (3) to compare the codes against the current methods used at Los Alamos.

The current method of evaluating plutonium exposure at Los Alamos uses the computer code PUQFUA, which calculates body burden from urine analysis results using the Langham equation modified to account for long term excretion.<sup>2</sup> Doses currently must be estimated using a method developed by J.N.P. Lawrence. Past DOE reporting requirements allowed reporting of internal exposure as body burden.<sup>3</sup> The new DOE requirements state that calculated dose equivalents will be assigned to individuals with internal contamination.<sup>4</sup>

The ICRP-30 codes model the complex metabolism of the human body with groups of chains of equations. The metabolism is depicted as linear first order kinetics, or one-way transfers between compartments beginning with an intake compartment and ending with an excretion compartment. The models are used to estimate intake based on bioassay results. Dose is assigned based on the estimate of intake.

The codes reviewed to date are REMedy from SAIC Inc., INDOS from Skrable Enterprises and AERIN, which was developed at LLNL. Two additional codes, GENMOD developed at Chalk River and PUCHEL developed at Savannah River, have been obtained and previewed, but are not yet quantitatively evaluated.

### EVALUATION METHOD

A test case was used to compare dose calculations with the three codes. The test case was an acute one-microcurie inhalation of class Y, 1 micron AMAD Pu-239. Hand calculations using the methods presented in ICRP-30 were also performed for comparison purposes using the test case input. The results are presented in Table I.

The codes were then used to evaluate an exposure of a former LANL plutonium worker. The particular case was chosen because it was a significant acute inhalation exposure that provided good urine analysis results for an extended period of time. The urine analysis data were followed 3200 days after exposure for the purposes of this study.

The codes were first used to estimate intake. Intake estimates are generated by the INDOS and REMedy codes. AERIN requires a hand calculation to determine intake. The version of REMedy used, allows only one twenty-four hour urine analysis result to be entered at a time. The intake is estimated based on that result. The result used was 3200 days after exposure. INDOS prompts the user for as many results as are available. In this case all available results out to 3200 days were used. The user is then asked to select several "fitting procedures" to estimate intake. The method used for this comparison was the unweighted least squares fit. NUREG/CR-4884<sup>5</sup> was also used to estimate intake. In addition, the methods presented in NUREG/CR-4884 were used with the intake retention functions generated by the AERIN code to estimate intake. Results of estimates of intake are presented in Table II.

Table I. Comparison of Dose Equivalent Estimates for a 1 Microcurie Inhalation of Class Y, 1 Micron AMAD Pu-239.				
Compartment	Estimate of Dose Equivalent (rem)			
	REMedy	INDOS	AERIN	ICRP-30
Effective Whole Body <sup>a</sup>	3.40E2	3.40E2	3.32E2	3.3E2
Lung	1.20E3	1.05E3	1.15E3	1.2E3
Liver	--	7.04E2	7.67E2	8.1E2
Red Marrow	2.82E1	2.55E2	2.79E2	2.8E2
Bone Surface	3.52E3	3.15E3	3.45E3	3.5E3

<sup>a</sup>Effective Whole Body dose is the weighted sum of individual organ doses as defined in ICRP-30.

Table II. Estimated Intakes in pCi.			
AERIN	INDOS	REMedy	NUJREG/CR-4884
5.84E5	3.11E5	2.45E5	2.15E5

Committed dose equivalents were then calculated with the three codes, and by hand calculation using NUREG/CR-4884 and the Los Alamos method.<sup>6</sup> INDOS and REMedy calculate dose equivalents based on input of bioassay data. AERIN requires that the user provide an estimate of intake. The Los Alamos method uses the estimate of body burden calculated by the PUQFUA code from urine analysis results using the modified Langham equation. The total disintegrations (nCi-y) in the body are calculated and partitioned to the various organs to determine dose equivalent. INDOS and REMedy claim to estimate dose from intakes by the same method as NUREG/CR-4884. The committed dose equivalent is estimated by dividing the estimated intake by the stochastic ALI and multiplying by 0.05 Sv, or 5 rem. However on examination, REMedy uses the non-stochastic ALI for Pu-239. The committed dose equivalent calculated by AERIN used the estimated intake presented in Table II as input to the code. The comparison of committed dose equivalents is presented in Table III.

## DISCUSSION

The ICRP-30 codes evaluated above model the metabolic pathways of the body from intake to excretion. The models are first order linear kinetics. The models predict the amount of a radionuclide in a compartment at any time after intake. Intake of a radionuclide is estimated based on the fraction of the initial intake in the compartment of interest and the amount measured via bioassay. Dose is then calculated based on the estimate of intake.

PUQFUA calculates body burden from urine analysis results using a modified Langham equation. The total disintegrations in the body are calculated. By hand calculation the total disintegrations are partitioned to various organs to determine dose equivalent in the organs. The assumption used to calculate dose is instantaneous deposition in the organs on intake with no temporal variation other than uniform excretion from all organs. This assumption appears to better account for energy depositions in the lung. Both autopsy results and other studies have demonstrated that the ICRP-30 500 day clearance half time in the lung is too short.<sup>7</sup>

The Los Alamos dose calculation method developed by J.N.P. Lawrence (and described above) is based on observations of Los Alamos plutonium workers who have come to autopsy. The fraction of plutonium distributed to each organ in the body is based on an average of the above

Table III. Comparison of Committed Effective Dose Equivalents.				
AERIN	INDOS	REMedy	NUREG/CR-4884	LANL
194 rem	96 rem	83 rem	66 rem	110 rem

observations. The Los Alamos method makes no attempt to model the transfer of plutonium through the lung or model the chronic feed from the lung to the systemic organs. The ICRP-30 based codes attempt to estimate intake and comprehensively model lung clearance, systemic deposition and excretion; and finally estimate dose. The results in Table III are surprisingly close in spite of the differences of the methods.

The test case of Table I demonstrated that for the same intake, the doses are comparable. The differences noted in committed dose equivalent in Table III can be attributed to the estimates of intake and not dose calculation methodology in the codes. The significantly higher intake and resulting dose calculated by the AERIN code are the result of the intake retention functions generated. Intake retention functions predict the fraction of intake that will be in the urine at a specific time post intake.

No attempts were made in this evaluation to modify models in any of the codes to fit the predicted urine results to the actual results. This is the next project that needs to be performed after initial evaluations of GENMOD and PUCHEL are completed (specifically, making adjustments in the current lung model to better determine intake and calculate dose).

The evaluations of the codes to this point have only considered urine analysis results from Pu-239 exposures. Intercomparisons of intake or body burden estimates from fecal and in vivo analyses are needed as well as intercomparisons for other radionuclides and routes of intake that are important in internal dosimetry at Los Alamos.

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## PERFORMANCE EVALUATION OF CONTINUOUS AIR MONITOR (CAM) SAMPLING HEADS

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*Funding Organization: DOE*

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Continuous air monitor (CAM) samplers detect radioactive aerosol particles in nuclear facilities and provide alarm signals should the concentrations exceed threshold levels. Aerosol particles drawn into a CAM sampler are collected on a filter. Radioactivity of the particles is determined with a detector near the filter face. An important determinant of CAM performance is the ability of the inlet and body of the CAM to transport inhalable particles to the filter without substantial loss or bias with respect to particle size.

We tested three types of CAM samples in a low-velocity aerosol wind tunnel in an attempt to determine how well particles penetrate through the flow systems to the collection filter under conditions typical of normal room air exchange rates. The CAM units were prototypes provided to Los Alamos

for test and evaluation by Kurz Instruments, Eberline Instrument Corporation, and Victoreen Inc., each unit incorporating a different CAM head design. (The performance of the instruments are available from the vendors.)

The CAM samplers were operated primarily at a flow rate of 28.3 L/min and two air velocities: 0.3 and 1.0 m/s. At an air speed of 1 m/s, aerosol penetration to the filters of the Kurz unit was essentially 100% for particle sizes of 3 and 7  $\mu\text{m}$  aerodynamic equivalent diameter (AED) and was 86% for an AED of 15  $\mu\text{m}$ . For the Eberline sample, the penetration was over 80% for 3- $\mu\text{m}$  AED particles but was reduced to less than 2% for 7- $\mu\text{m}$  particles and larger. The Victoreen sampler showed penetration values of 98% for 3- $\mu\text{m}$  AED particles, 88% for 7- $\mu\text{m}$  particles, and 4% for 15- $\mu\text{m}$  particles. Thus air speed had little effect on the penetration results for the two speeds tested.

In tests to determine the uniformity of aerosol deposits on the filters of the CAM samplers, the deposits for 10- $\mu\text{m}$  AED particles were nonuniform for all three of the instruments. We expect that nonuniform sample deposits will degrade instrument performance under certain circumstances, particularly large dust loads.

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## *Organization*

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Protection of its employees, the public, and the environment is a major concern of the Los Alamos National Laboratory. The mission of the Laboratory's Health, Safety, and Environment Division is to ensure that this concern is successfully transformed into sound engineering and administrative approaches to health and safety at the Laboratory.

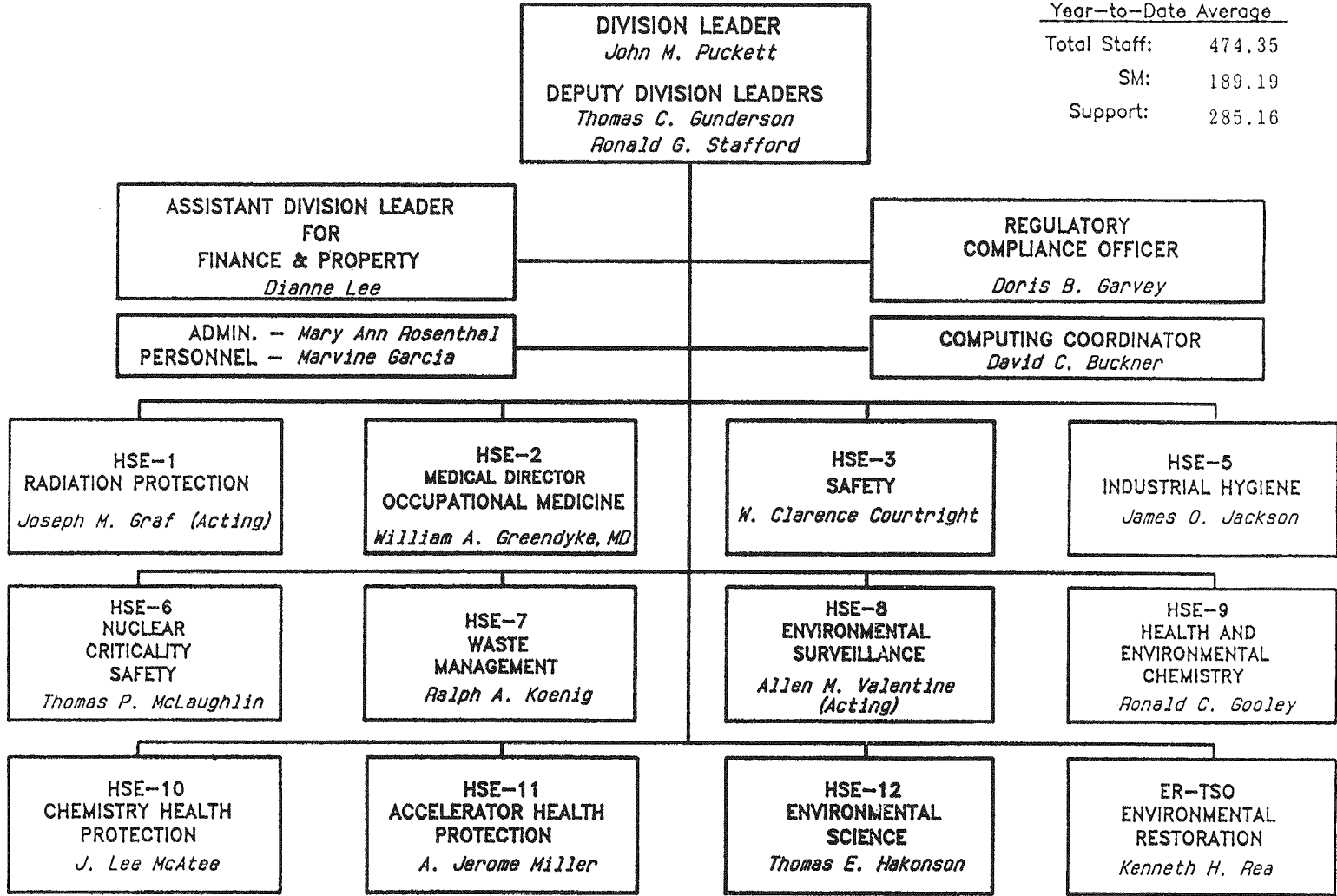
An important accessory in achieving this goal is occupational health and environment research for identifying and solving problems affecting Laboratory operations. Technical resources of the Division are also applied to nationally significant occupational health problems. These objectives are achieved through an integrated approach that combines technical support and applied research activities in health physics, industrial hygiene, occupational medicine, epidemiology, industrial safety and fire protection, nuclear criticality safety, waste management, and environmental surveillance (see accompanying chart).

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# HEALTH, SAFETY, AND ENVIRONMENT DIVISION LOS ALAMOS NATIONAL LABORATORY

Year-to-Date Average	
Total Staff:	474.35
SM:	189.19
Support:	285.16



November 1, 1988