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# DISTRIBUTION OF RADIOACTIVE ELEMENTS IN THE DEBRIS OF THE SALMON NUCLEAR TEST

D. G. KARRAKER
W. C. PERKINS

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*Savannah River Laboratory*

*Aiken, South Carolina*

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# DISTRIBUTION OF RADIOACTIVE ELEMENTS IN THE DEBRIS OF THE SALMON NUCLEAR TEST

By

D. G. Karraker & W. C. Perkins

Approved by

L. H. Meyer, Research Manager
Separations Chemistry Division

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## ABSTRACT

Analysis of the debris of Salmon, an underground nuclear test detonated near Hattiesburg, Mississippi, showed that most of the isotopes formed during the explosion collected in a thin layer at the bottom of the original cavity. An explanation is offered for the vertical distribution of radioactive species in the debris.

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# DISTRIBUTION OF RADIOACTIVE ELEMENTS IN THE DEBRIS OF THE SALMON NUCLEAR TEST

## INTRODUCTION

As a part of the Project Vela Uniform, Salmon, a 5-kiloton nuclear explosion was detonated in the Tatum Salt Dome, near Hattiesburg, Mississippi on October 22, 1964. The primary purpose of Salmon was to test the capability of detecting and locating underground nuclear explosions. However, Salmon also offered an opportunity to investigate the physical and chemical effects resulting from the explosion; one of the more interesting involves the distribution of radioactive species in the debris.

The salt in the neighborhood of the detonation was principally NaCl with anhydrous $CaSO_4$ varying from 0.5 to 25%;(1) the water content of the salt was <30 ppm. Besides the materials used in the components of the nuclear device, several hundred pounds of tantalum and about 1 ton of steel were included in the shot cavity. A known amount of $^{244}Cm$ tracer was included in the device to allow an estimate of the fraction of the actinides present in post-shot debris samples.

The device was detonated 2716 feet below ground level. Post-shot drilling and pressure tests established that a spherical cavity of about 19,000 cubic meters was created. The radius of the cavity was about 56 feet, and the cavity was filled to about 1/5 its vertical height with material melted by the explosion. All material originally present in the neighborhood of the device except volatile fission products was found in this pool of salt. This report describes the results of measurement of the radioactive species present in samples taken from the floor of the cavity at different depths. These data provide a basis for speculation on the chemical behavior of each element in the debris during and following the detonation.

## SUMMARY

Chemical and radiochemical analyses of samples from a drill coring through the debris of the Salmon shot showed that about 90% of the radioactive species were in a 3-inch-thick layer at the bottom of the debris. This layer contained 22 wt % CaO, 9 wt % $Al_2O_3 \cdot SiO_2$ and 1.4 wt % Fe. The presence of these compounds indicates that this layer was heated to a high temperature by the nuclear detonation. The remainder of the debris contained essentially no CaO, and 6 to 27 wt % $CaSO_4$; apparently the temperature

of the remainder of the debris did not exceed 1200-1500°K, the decomposition temperature of $CaSO_4$. An explanation is offered for the vertical distribution of $CaSO_4$ and radioactive species in the debris.

## EXPERIMENTAL

### SAMPLING

Approximately eight months after the device was detonated, the debris was analyzed at the Savannah River Laboratory (SRL) for $\alpha$- and $\gamma$-emitting isotopes and major elemental constituents. Ten samples were taken at intervals along a vertical drill coring from the debris on the floor of the cavity. A preliminary survey of the coring showed that the gamma radiation increased irregularly starting at the top of the recrystallized melt (2748 feet below ground), reached a sharp maximum at -2771.5 feet, and decreased very rapidly at greater depths. The radiation profile and sample locations are shown in Figure 1. The debris was white salt, containing large fused crystals and occasional yellow or black deposits. The debris was sampled in a manner that avoided the rust and oil left on the exterior of the coring by the drill.

### GAMMA SPECTROSCOPY

Gamma-emitting isotopes were tentatively identified by the energy of their radiation, and these assignments were confirmed by their chemical behavior. Weighed fragments of about equal size* were analyzed by a solid state, lithium-drifted germanium detector with a 10-mm$^2$ surface area. This detector and associated components produced a 1.33-Mev $^{60}Co$ peak with a 15-kev half-width. The detector output was fed to a 4096-channel analyzer for energy resolution. The major gamma peaks observed and their assignments are shown in Table 1. Selected samples were leached with water to remove NaCl, and then with 12M HCl to remove the acid-soluble constituents. The relative weights of each fraction are shown in Table 2. After the soluble fractions were evaporated, each fraction was counted. The water-soluble and HCl-soluble fractions were then redissolved in 12M HCl and passed through a column of "Dowex 2"** anion exchange resin. The column was eluted with increasingly dilute solutions of HCl, according to the method of Hicks, et al.(2) The behavior of the isotopes during this treatment was as predicted(2,3) and was used as the basis for the gamma

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* A much smaller fragment of the sample from a depth of 2771.5 feet was analyzed because of its higher radiation level.

** "Dowex" is a trademark of Dow Chemical Company.

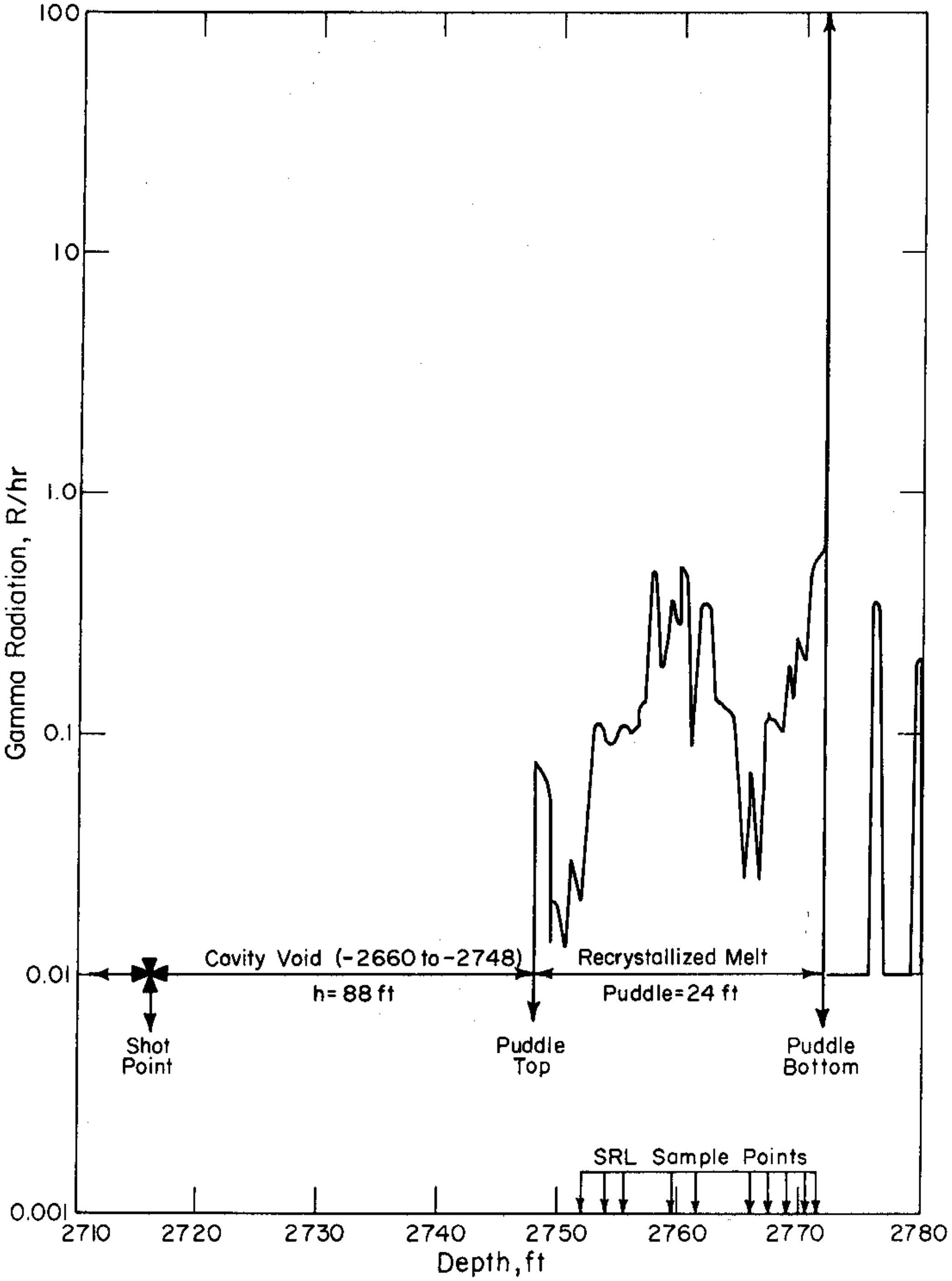

FIG. 1 RADIATION PROFILE AND SAMPLE POINTS FROM CORING OF SALMON CAVITY (t=150 days) Redrawn from Reference 1.

TABLE 1

Gamma Spectrum and Peak Assignments of Salmon Samples

| Energy, Mev | Assignment | Basis for Assignment[(a)] |
|---|---|---|
| 0.135 | $^{144}Ce$ | A, C, E |
| 0.42 | $^{127}Te$ | A, C, F(eluted by 1M HCl) |
| 0.42, 0.46, 0.60 | $^{125}Sb$ | A, C, F(not eluted by HCl) |
| 0.51 | $^{106}Ru$-$^{106}Rh$ | D, F(not eluted by HCl) |
| 0.66 | $^{137}Cs$-$^{137}Ba$ | B, E |
| 0.69 | $^{127m}Te$ | A, C, F(eluted by 1M HCl) |
| 0.73 | $^{95}Zr$ | A, D, F(eluted by 6-9M HCl) |
| 0.76 | $^{95}Zr$-$^{95}Nb$ | A, D, F(eluted by 1-6M HCl) |
| 0.84 | $^{54}Mn$ | B, E, G |
| 1.12 | $^{182}Ta$ | A, D, F(not eluted by HCl) |
| 1.17 | $^{60}Co$ | A, B, F(eluted by 3-6M HCl) |
| 1.19 | $^{91}Y$ | C, E |
| 1.22 | $^{182}Ta$ | A, D, F(not eluted by HCl) |
| 1.27 | $^{22}Na$ | B, E |
| 1.29 | $^{59}Fe$ | C, F(eluted by 3M HCl) |
| 1.33 | $^{60}Co$ | A, B, F(eluted by 3M HCl) |
| 2.18 | $^{144}Pr$ | A |

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(a) A. Other peaks found from decay scheme, parent, or daughter.
B. $H_2O$ soluble.
C. HCl soluble.
D. Partly HCl soluble.
E. Not absorbed by anion resin from HCl.
F. Chloride complex absorbed on anion resin.
G. Scavenged by $MnO_2$ precipitate.

TABLE 2

Solution Properties of Salmon Samples

| Sample Depth, ft | Weight % Soluble in $H_2O$ | Weight % Soluble in 12M HCl | Weight % Insoluble in HCl |
|---|---|---|---|
| 2752 | 96 | 2.0 | 2.3 |
| 2754 | 87 | 2.1 | 9.8 |
| 2755.5 | 76 | 2.1 | 24 |
| 2759.5 | 74 | 8.6 | 16 |
| 2761.5 | 70 | 5.6 | 23 |
| 2766 | 92 | 2.0 | 5.9 |
| 2767.5 | 91 | 2.5 | 6.4 |
| 2769 | 89 | 1.5 | 8.3 |
| 2770.5 | 90 | 5.9 | 0.6 |
| 2771.5 | 56 | 33 | 5.7 |

peak assignments (Table 1). To confirm the identification of $^{54}$Mn, the 12M HCl fraction from the sample at -2766 feet was redissolved; and the 0.84-Mev peak was removed from the solution by precipitation of MnO$_2$.

Quantitative data for each isotope were obtained from the area under its peak in the $\gamma$ spectrum of the solid weighed fragments from each sample. For those isotopes whose spectra had multiple peaks, the most highly resolved peak was used. The energies of the peaks and the corrections for each are shown in Table 3. Background corrections were made graphically to exclude the contributions of Compton peaks. Normally, interferences were not severe because of the high resolution of the detector. In a few cases, because one of the interfering isotopes emitted an additional gamma ray, the intensity ratio of the two peaks was used to calculate the contribution of the masked isotope to the gamma peak at the energy studied. In this manner, the activity of $^{91}$Y was corrected for $^{60}$Co and $^{182}$Ta. The peaks for $^{22}$Na and $^{59}$Fe were resolved after chemical separation of the $^{22}$Na into the water-soluble fraction.

TABLE 3

Selected Peaks and Correction Factors for Interpreting $\gamma$ Spectra

| Isotope | Energy of Peak Measured, Mev | Corrections | | $\gamma$ Abundance,$^{(4,5)}$ % |
|---|---|---|---|---|
| | | Decay | Efficiency (Multiplied by $10^5$) | |
| $^{106}$Ru-$^{106}$Rh | 0.52 | 1.66 | 21 | 21 |
| $^{125}$Sb | 0.60 | 1.23 | 17 | 24 |
| $^{137}$Cs-$^{137}$Ba | 0.66 | 1.02 | 15 | 82 |
| $^{95}$Zr | 0.76 | 21 | 12 | 49 |
| $^{54}$Mn | 0.84 | 2.09 | 10 | 100 |
| $^{91}$Y | 1.19 | 31 | 6.0 | 0.2 |
| $^{182}$Ta | 1.22 | 5.64 | 5.8 | 28 |
| $^{59}$Fe | 1.27 | 83 | 5.3 | 43 |
| $^{22}$Na | 1.27 | 1.24 | 5.3 | 100 |
| $^{60}$Co | 1.33 | 1.11 | 5.0 | 100 |
| $^{144}$Pr | 2.18 | 2.01 | 2.4 | 0.8 |
| $^{242}$Cm$^{(a)}$ | 6.1 | 4.18 | - | - |

---

(a) Alpha peak values.

The efficiency of the detector was determined at each energy with standard counting sources. When a standard was not available for an isotope, the calibration was made with another standard that emitted a gamma ray of approximately the same energy, and a decay scheme correction was made.(4,5) Finally, each activity was corrected for the time elapsed between detonation of the device and sample analysis. The corrected data are given in Figures 2, 3, and 4 and in Table 4.

TABLE 4

Gamma Activity Concentrated in Bottom Samples,[a] d/m/g

| Sample Depth, ft → | 2752-2769 | 2770.5 | 2771.5 |
|---|---|---|---|
| $^{91}Y$ | $<5 \times 10^{8}$ | $<10^{8}$ | $2.7 \times 10^{11}$ |
| $^{59}Fe$ | $<10^{6}$ | $1.1 \times 10^{6}$ | $9.1 \times 10^{8}$ |
| $^{144}Pr$ | $<6 \times 10^{5}$ | $4.2 \times 10^{5}$ | $1.65 \times 10^{9}$ |
| $^{22}Na$ | $<5 \times 10^{3}$ | $<7 \times 10^{3}$ | $2.1 \times 10^{5}$ |

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(a) Corrected to 10/22/64.

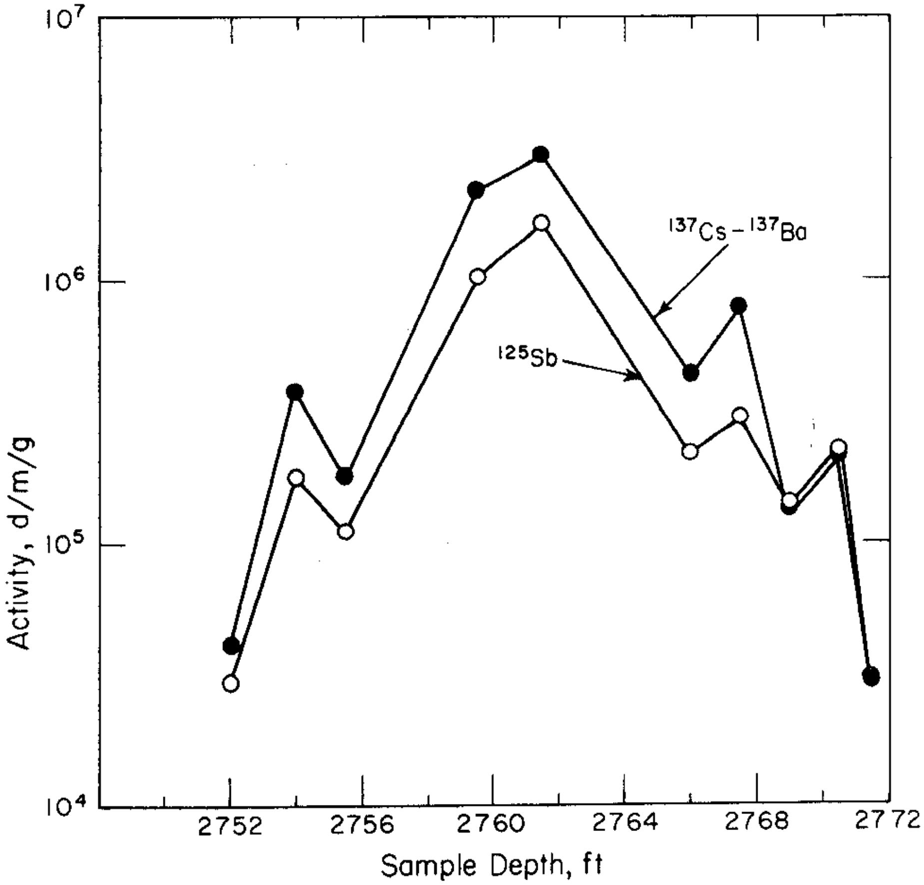

FIG. 2 $^{137}Cs$ AND $^{125}Sb$ DISTRIBUTION IN SALMON DEBRIS

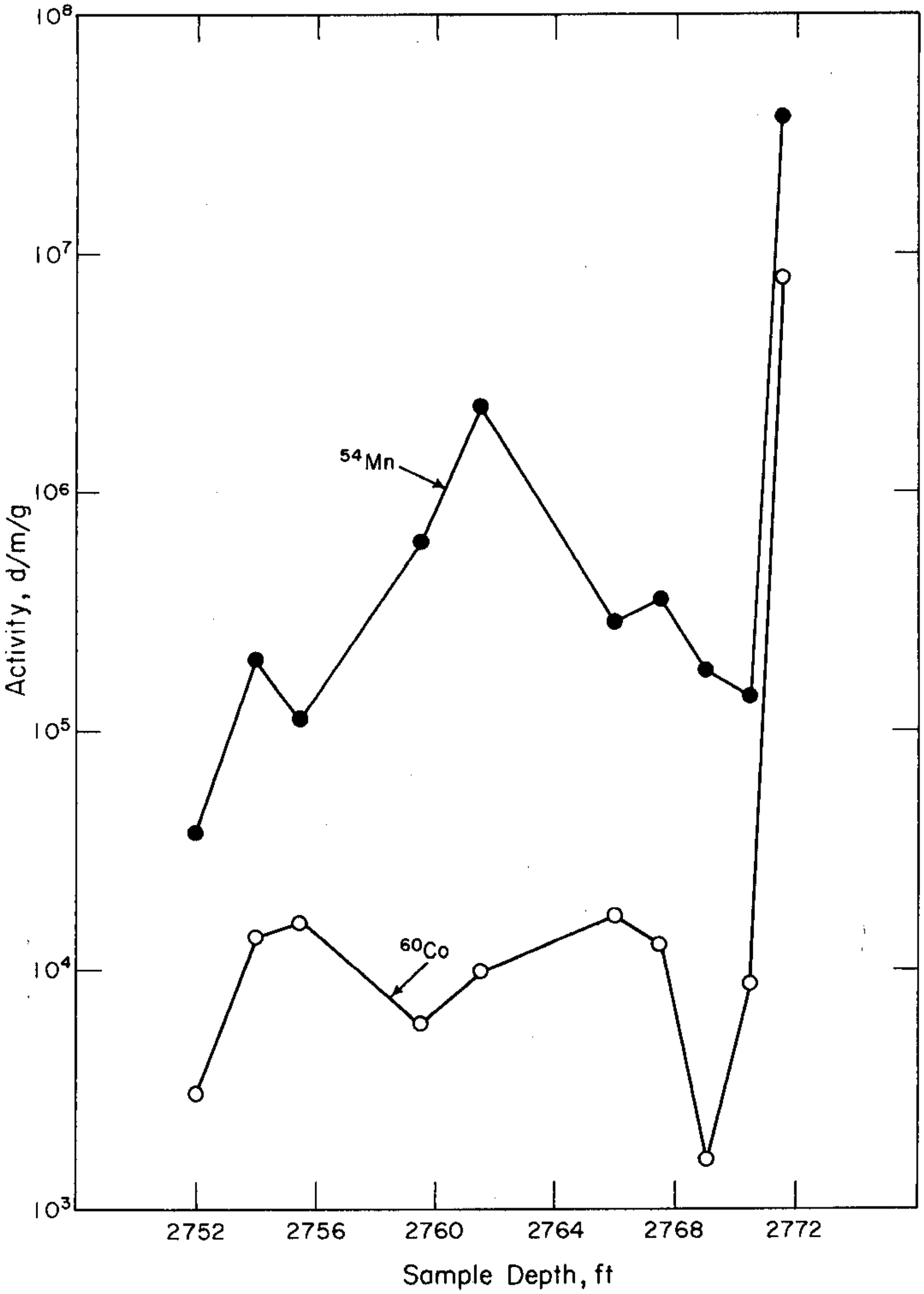

FIG. 3 $^{54}$Mn AND $^{60}$Co DISTRIBUTION IN SALMON DEBRIS

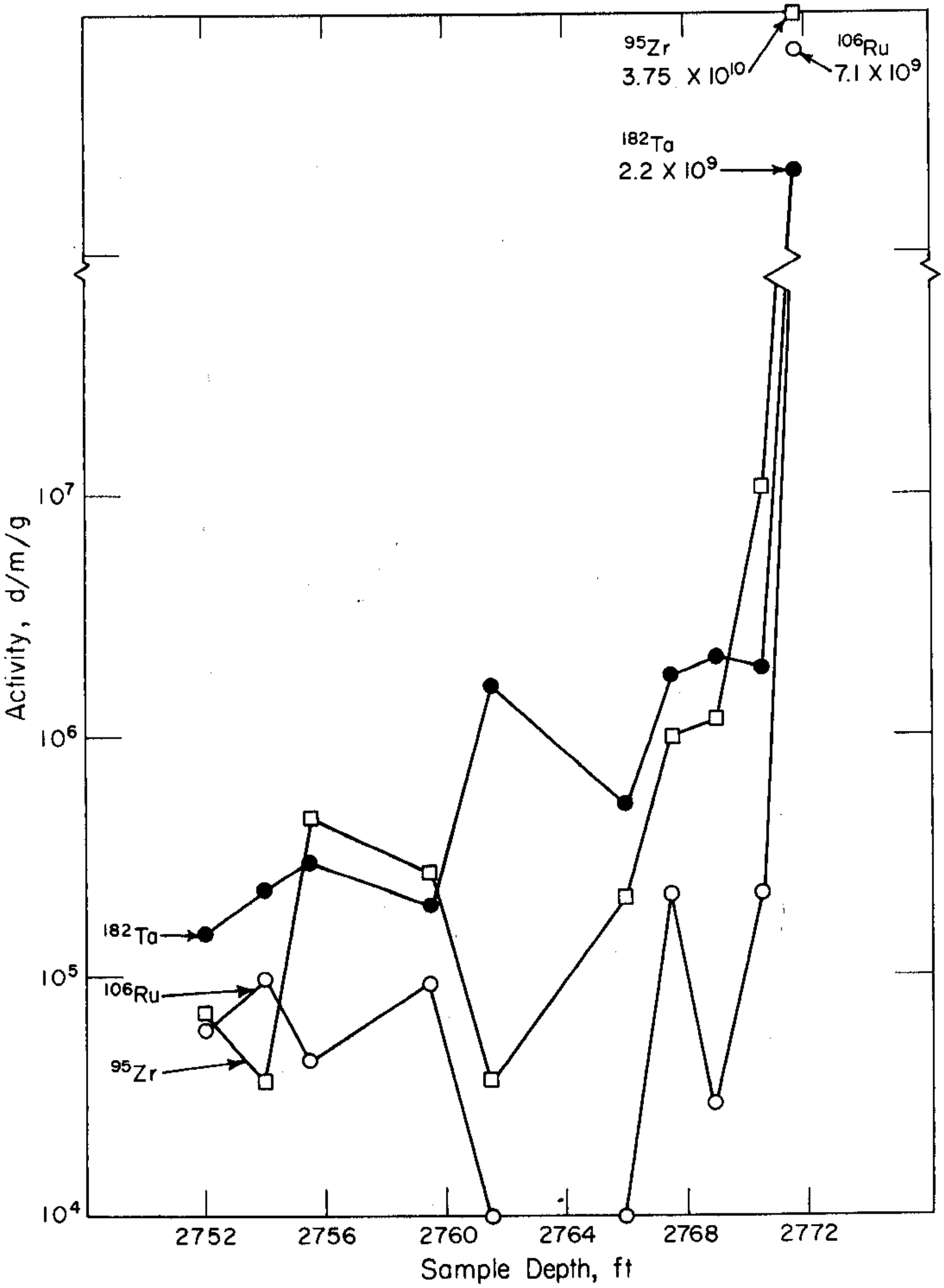

FIG. 4 $^{95}$Zr, $^{106}$Ru, AND $^{182}$Ta DISTRIBUTION IN SALMON DEBRIS

## ALPHA PULSE HEIGHT ANALYSIS

Samples used for gamma analyses were also analyzed for actinide elements by alpha pulse height analysis. The samples were dissolved in a large excess of hot 1M HCl, and the actinide elements were electrodeposited from the NaCl-HCl solution by the method of Donnan and Dukes.(6) The alpha energies were resolved by an Ortec Surface Barrier (silicon solid state) Detector with a 300 $mm^2$ surface area and a resolution of about 50 kev. The output from the detector was resolved by a 400-channel analyzer. The gross alpha count was measured with a standard windowless, gas-flow counter with an efficiency of 50%. The assignments of peaks are based on comparisons with the alpha peaks from standard actinide sources. The results are shown in Table 5 and Figure 5.

As shown in Table 2, none of the samples dissolved completely in HCl solution. Analysis of the residues from the 1M HCl treatment showed that, with the exception of three samples, the amount of alpha-emitting isotopes in the residue was negligible compared to the amount in HCL solution. No Cm was found in the residues. Multiple analyses were made on these three samples, and the results were reproducible, within the limits of analytical accuracy.

TABLE 5

Alpha Activity of Salmon Samples(a)

| Sample Depth, ft | Gross α, d/m/g |
|---|---|
| 2752 | 330 |
| 2754 | 1310 |
| 2755.5 | 2840 |
| 2759.5 | 1200 |
| 2761.5 | 6900 |
| 2766 | 1410 |
| 2767.5 | 2100 |
| 2769 | 2570 |
| 2770.5 | 8230 |
| 2771.5 | $1.66 \times 10^7$ |

(a) Corrected for decay of $^{242}$Cm after detonation.

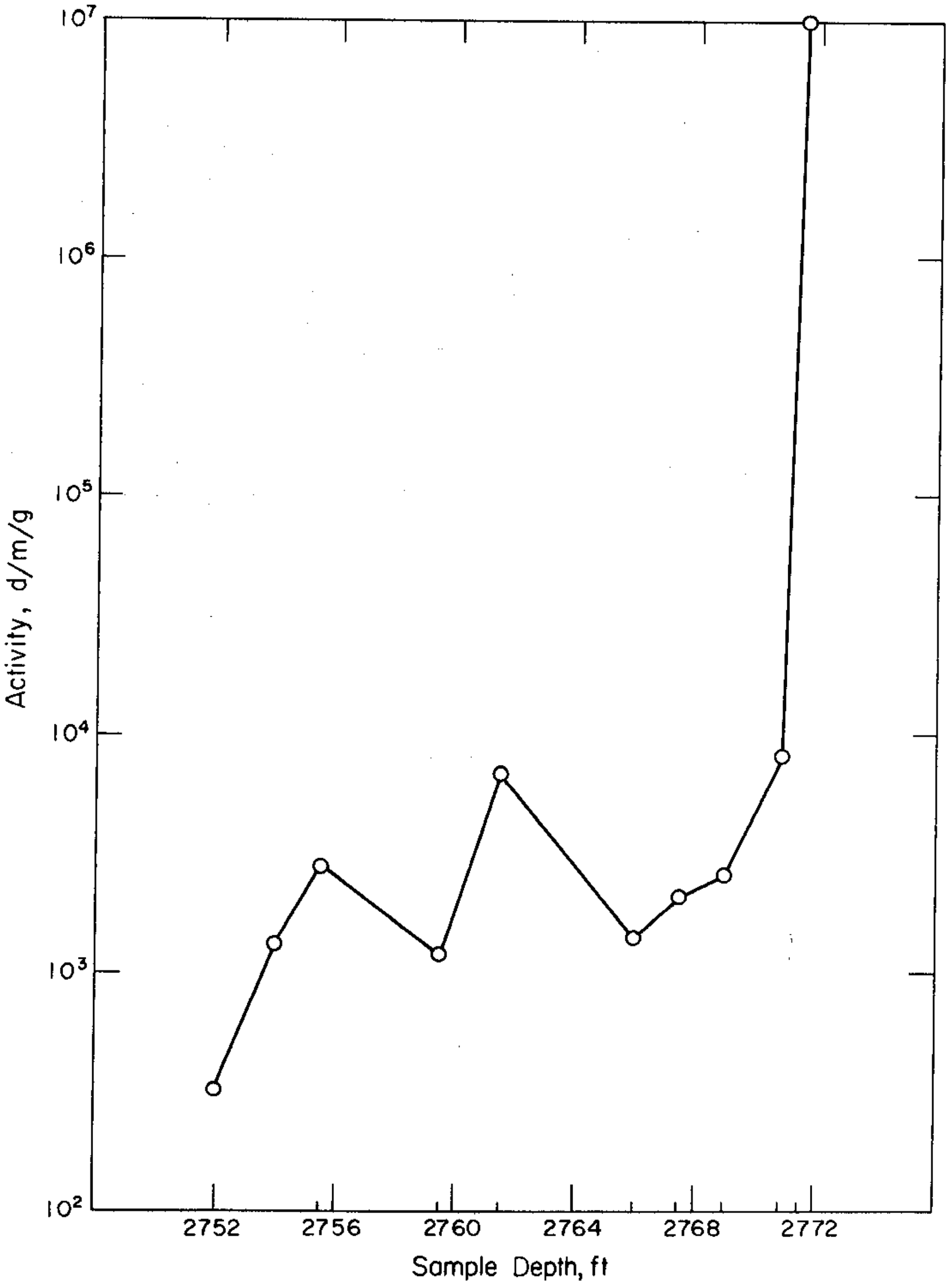

FIG. 5 GROSS ALPHA ACTIVITY DISTRIBUTION IN SALMON DEBRIS

## CHEMICAL ANALYSIS

Samples of material from each level were analyzed for major elemental constituents by conventional chemical and physical methods. Sodium, calcium, aluminum, iron, and magnesium were measured by atomic absorption spectrometry; sulfate was measured by its absorption in the infrared. Sodium chloride was calculated from the sodium content.

The water leach treatment removed NaCl from the insoluble fraction efficiently. Anhydrous $CaSO_4$ dissolved only partially during the water and HCl leaches, and was the only major constituent of the insoluble fractions from the upper eight samples. The results are summarized in Table 6.

### TABLE 6

Composition of Salmon Debris

| Sample Depth, ft | % NaCl | % $CaSO_4$ | Other |
|---|---|---|---|
| 2752 | 93 | 6.2 | - |
| 2754 | 83 | 13 | - |
| 2755.5 | 71 | 26 | - |
| 2759.5 | 74 | 20 | - |
| 2761.5 | 69 | 27 | - |
| 2766 | 89 | 10 | ~1% $Al_2O_3$ |
| 2767.5 | 89 | 10 | - |
| 2769 | 89 | 10 | - |
| 2770.5 | 84 | 7.1 | ~1% $SiO_2 \cdot Al_2O_3$; ~1% Fe; Mg |
| 2771.5 | 60 | 2.8 | 22% CaO; ~9% $SiO_2 \cdot Al_2O_3$; 1.4% Fe; also Mg, Cr, and Pb (Ta not detected: <100 ppm) |

## DISCUSSION

The principal conclusion derived from the data is that all radioactive constituents except $^{137}Cs$ and $^{125}Sb$ are concentrated in a thin layer at -2771.5 feet. This layer, about 3" thick, is estimated to contain about 90% of all radioactivity in the shot debris. There is a second, and much shallower, maximum in the vertical distribution at -2761.5 feet for $^{137}Cs$, $^{125}Sb$, $^{54}Mn$, and possibly $^{182}Ta$; $^{106}Ru$ and $^{95}Zr$ show minima in the same region. The chemical composition of these two layers differs from that of other layers; the layer at -2771.5 feet contains 22 wt % CaO and 9 wt % $Al_2O_3 \cdot SiO_2$, neither is present in any significant concentration in any other layer. Samples from -2755.5, -2759.5, and -2761.5-feet contain over twice as much $CaSO_4$ as samples from layers immediately above and below. The data, together with the known physical and chemical properties of these compounds, allow some deductions on the physical and chemical processes that occur between detonation of the device and solidification of the debris.

The sample from -2771.5 feet is a gray, extremely radioactive-mass. Large amounts of CaO, $Al_2O_3 \cdot SiO_2$, and Fe in this sample - and not in other samples - indicate that this layer, and only this layer, was heated above 1200°K, the decomposition temperature of $CaSO_4$. The estimated 50-100 tons* of debris in this layer probably contains most of the elements of low volatility from the "fireball" produced by the nuclear reaction.

This deduction is supported by crude calculations, which show that a 5-kT detonation would heat a mass of 50 tons to a temperature of $\sim 10^5$ K (10 kev) and create a pressure of $10^6$-$10^7$ atmospheres in the immediate vicinity of the explosion. If this mass of material were expanded adiabatically to the size of the cavity, it would have a pressure of about 400 atmospheres, or 6000 psi. The pressure on the expanding mass from the weight of the overburden would be about 3000 psi. Because the expansion of the gas is not adiabatic (energy is carried off by the shock wave, by compression of the surroundings to produce the cavity, and by heat loss to the surroundings), the calculations are in reasonable agreement with the data.

The radioactive species in the -2771.5-foot layer are expected to be present mainly as oxides or oxychlorides. During the cooling of the "fireball", elements compete in the formation of chemical compounds. The radioactive elements present - actinides, lanthanides, manganese, cobalt, zirconium, ruthenium - will be forced to form oxides (or oxychlorides), either because

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* Reference 1, p. 11 estimates 50 tons on the basis of 2% Fe content. SRL analyses on small samples show 1.4 wt %.

their oxides are more stable than their chlorides or because the system $MCl_n + n/2\ Na_2O$ is less stable than $MO_{n/2} + n\ NaCl$. The densities of the oxides all exceed the density of molten salt.

A convenient hypothesis(1) that explains the sharp demarcation of the radioactive species in the -2771.5-foot layer is that the denser oxides settled rapidly from the molten salt into a distinct layer. The major objection to this hypothesis derives from the vertical distribution of $CaSO_4$ (specific gravity = 2.96) (Table 6), which reaches 27% at -2761.5 feet, decreases to a constant 10% from -2766 feet to -2769 feet, and then decreases to 2.8% at -2771.5 feet. This distribution is not consistent with the theory of sedimentation, which would predict increasing $CaSO_4$ concentration with depth.

A more reasonable explanation is that the material in the -2771.5-foot layer condensed while the salt was still vaporized, collected at the bottom of the cavity, and solidified. Later, the vaporized salt condensed on the walls of the cavity, melted more salt on the walls, and flowed to the floor of the cavity, forming a pool over the solidified oxides. At this point the pool contained little $CaSO_4$, because most of the melt derived from NaCl vapor ($T > 1700^{\circ}K$). While the pool was cooling to its freezing point, more salt melted and flowed from the walls of the cavity, bringing down $CaSO_4$. The loss of heat from the pool was more rapid by conduction through solid salt than by transfer through the gases above the melt; therefore the pool of salt froze slowly from the lower periphery. As the pool was freezing, $CaSO_4$ in the melt settled through the liquid NaCl. The distribution of $CaSO_4$ in the melt suggests very rapid freezing of the layer at -2771.5 feet, comparatively rapid freezing up to -2766 feet, then a relatively slow freezing from -2761.5 feet to the surface of the pool.

This simple model is consistent with the accumulation of most radioactive species in the bottom of the melt, but requires further assumptions to explain the vertical distribution of the fission products - particularly in the region of -2760 feet. The vertical distributions of $^{125}Sb$ and $^{137}Cs$ show distinct maxima at -2761.5 feet. The distribution of these activities can be explained by assuming that $SbCl_3$ or $SbCl_5$, which are volatile at the temperature of molten salt, and $^{137}Xe$ (the precursor of $^{137}Cs$) were diffusing to the surface of the melt and escaping. As the pool froze, the volatile species were trapped. The concentrations of volatile species trapped near the surface of the melt were lower, because the upper part of the pool froze last. The accumulation of $^{54}Mn$ and $^{182}Ta$ at -2761.5 feet was probably due to sedimentation from liquid salt; freezing of the salt below this level prevented further settling.

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