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CONTAMINANT SIGNATURE AT LOS ALAMOS FIRING SITES

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FACILITIES DESCRIPTION AND HISTORICAL DEPLETED URANIUM USAGE

Los Alamos National Laboratory was selected for its remote location over 50 years ago as the site for development of the first atomic bomb. The Laboratory's 43 mi² area sits above 7000 ft on a broad plateau of volcanic tuff above the Rio Grande, the master stream of the region. Known as the Pajarito plateau, it is dissected into long, finger-like mesas by deep canyons, creating a rugged terrain. Annual precipitation of nearly 20 inches, falls as snow and as rain, with 40 percent occurring during the height of the summer monsoon season (Bowen, 1990). None of the canyons across the Laboratory contain perennial flow. Water in the stream channels is from snowmelt or runoff from rainfall events.

A large portion of the Laboratory is devoted to open air dynamic firing sites and their adjacent buffer zones. Components of weapons are tested at Los Alamos, whereas completed weapons were tested at the U.S. Department of Energy's Nevada Test Site near Las Vegas (The last weapon test at the Nevada Test Site occurred in September 1992). At Los Alamos, (mostly) depleted uranium has been substituted for enriched uranium since weapons testing began during the early 1940's. It is estimated that as much as 100 metric tons have been expended since operations began (Becker, 1991).

During a dynamic weapons test, a weapons component is either explosively detonated or impacted against a target in the open air environment. This results in both the production of a wide size range of depleted uranium particles as well as particle scattering over a considerable distance away from the firing pad. The explosive detonation process which creates aerial distribution over a watershed distinguishes this contaminant transport problem from others where the source term is spatially discrete (e.g., transport away from a waste pile or landfill).

DESCRIPTION OF FIELD AND LABORATORY ACTIVITIES

Investigations began in 1983 with collection of onsite soils, sediments, and rock samples to establish background uranium concentrations. Because the Laboratory is situated on volcanic Bandelier tuff which naturally contains uranium, it was decided to request isotopic uranium analyses on all soil and sediment samples. The isotopic analysis is a technique which can uniquely

distinguish uranium associated with dynamic weapons testing from the indigenous uranium present in the Bandelier tuff.

Although there are numerous watersheds at the Laboratory which contain firing sites where dynamic tests are conducted, investigations were confined to one watershed named Potrillo Canyon. Potrillo Canyon was selected because of its small size (3.1 mi²), it is completely contained within the Laboratory boundaries, it is limited to public access, and contains 5 firing sites, four of which remain active today. A conservative estimate of the total uranium source term in Potrillo Canyon is about 35,000 kg (Becker, 1991).

Field investigations (Becker, 1991) also began in 1983 with the installation of a runoff monitoring program, which was rugged and could collect flow data from spring, summer, and autumn rainfall/runoff events without power or an operator. Investigations were expanded to collect samples of fallout particles to assess uranium contribution in the air pathway; watershed-wide sampling of surface soils to quantify the spatial distribution of uranium; sampling of suspect geomorphologic deposits such as alluvial fans and point bars expected to concentrate uranium; depth sampling in three cross-canyon transects and in a 475-ft long trench; samples of snowmelt; and continuous monitoring of rainfall and crest stage measurements for flow.

Supplemental to the field investigations were laboratory studies (Becker, 1991). Leaching experiments were performed to assess uranium partitioning between particulate and dissolved phases. Deionized water was adjusted to a pH range of 4.65 to 4.75 to simulate the pH of natural rainwater measured in the Los Alamos area. Soils expected to contain depleted uranium were collected in the channel downstream from firing sites, continuously agitated, and periodically sampled to examine dissolution kinetics.

Depleted uranium-contaminated sediments were separated into individual grain sizes ranging from pebbles to the silts and clay fraction to measure how depleted uranium distributes as a function of particle size. This information was expected to be later related to the dynamics of uranium transport.

RESULTS OF DEPLETED URANIUM SAMPLING IN SOIL, SEDIMENT, AIR, AND WATER

In all, more than 750 contaminant measurements of atmospheric fallout, soil, sediment, and water and suspended sediment in spring/summer/autumn runoff were collected between 1983 and 1990 and analyzed for total uranium to evaluate the magnitude of transport of uranium away from firing sites by airborne and surface water runoff mechanisms. Results for the maximum, minimum, and mean values are presented in Table 1. Background concentrations of uranium in fallout range from 1-6 µg/g, in soil from 2-5 µg/g, and in water about 1 ppb (Becker 1991). The greatest concentrations of uranium were found in transported suspended sediment carried in runoff waters where average concentrations were 51.1 µg/g, followed by sediment present in

stream banks where average concentrations were 42.2 $\mu\text{g/g}$, Table 1. Average concentrations of 17.5 $\mu\text{g/g}$ were observed in geomorphologic deposits such as alluvial fans and point bars. Average uranium concentrations dissolved in runoff water of 11.9 ppb were also found to be elevated above background concentrations. Uranium present in fallout and in surface soils were found to be at or slightly above background concentrations in most samples, which indicated that airborne transport and wind redistribution is not significant in mobilizing uranium away from firing sites. Uranium concentration in runoff in the dissolved and suspended sediment phases were found to decline with downstream direction in the watershed and increasing distance from firing sites, with the largest concentrations below two firing sites near the top of the watershed. This implied both dilution and contaminant deposition with increasing hydrologic distance from firing sites. Leaching studies of uranium attached to channel sediments showed that uranium readily leaches into the dissolved phase, often in a matter of a few hours. Equilibrium between the dissolved and sediment phases was determined to range between 24 and 48 hours. Grainsize analyses indicate that, in general, uranium concentrations increase with decreasing particle sizes and that uranium has a particular affinity for the silt and clay-sized particles.

Table 1
Uranium in Air, Water, Sediment, and Soil
Units are $\mu\text{g/g}$ (except where noted)

	Min	Max	Mean	Standard Deviation
Air (fallout)	0.8	7.5	3.5	2.1
Soil (top 5 cm)	1.2	66.	4.8	8.3
Runoff				
-dissolved (ppb)	BDL*	654	11.9+	53.4+
-suspended sediment	0.5	404.9	51.1	157.1
Sediment				
-Channel Deposits	1.0	158.1	8.6	23.0
-Bank Deposits	1.5	373.0	42.2	100.3
Alluvial Fans and Point Bars	1.6	154.5	17.5	39.8

* Below Detection Limits

+ Derived using Maximum Likelihood Estimators (Gilliom and Helsel, 1986).

USING CONTAMINANT DATA TO DEFINE A FIRING SITE

There are many ways to define the boundaries of firing sites, dependent on the justification for doing so. Examples of firing site boundaries may be based on hazard circle radii for flying debris, safety control boundaries such as safety gates, radiological control areas, the actual pad itself, etc. We sought to develop a different methodology, based on the contaminant data collected.

In many instances, samples of soil, sediment, and water collected between 1983 and 1990 were analyzed for multiple analytes, translating into over 4100 measurements. Samples were organized into a database which included information such as their geolocation, both by coordinate and geologic/geomorphologic strata, media, date collected, analytic result and uncertainty, if there were duplicates available, and so on (Becker, David, and Hoopes, 1995). It was then relatively facile to examine some statistical features of this large data set.

Uranium, uranium isotopic ratio, barium, beryllium, mercury, lead, and copper were selected for examination on how these elements distribute in the surface or near surface soils in the vicinity of firing sites. Selection was based on substantial usage of these materials through time. Element concentration as a function of distance from firing sites are shown in Figures 1 through 7.

A number of comments accompany these figures. First, for each element of interest, adequate data were available to establish background values, including some natural spatial variability. Second, the uranium and uranium isotopic ratio plots represents at least 150 values, collected over an 8-year period of time. Other analyte plots contain lesser amounts, but still remain a statistically significant data set. Third, these data were collected on a watershed scale, and therefore represent information from 5 separate firing sites. Fourth, data are representative of concentrations on mesa tops and in stream channels, which constitute two quite different hydrologic and transport environments. Finally, where there was considerable range in concentrations, the data were transformed by logarithm before plotting.

The most striking observation is the uniformity with which the element concentration falls to background within a roughly 3000 ft radius of the firing site, and that this occurs for all the elements examined. This observation is unexpected due to the different materials and testing histories at each firing site, the explosive nature of the contaminant distribution, and fundamental differences in the amounts of each material used. From a contaminant transport viewpoint, this is a very interesting observation suggests several conclusions. First, it implies that even though the weapon component is forcefully detonated into the open atmosphere, for the most part, the main mass of contaminant is confined to a 3000 ft, and for some elements, 1000-ft radius of the firing site pad. This confirms the fallout data interpretation that the airborne transport pathway is not predominant in the distribution/redistribution of the dynamic weapons testing. Second, in the semi-arid climate at Los Alamos, the hydrologic processes in the last 50 years have not moved the main center of contaminant mass beyond a 3000-ft radius of the original site of detonation, even though there is considerable relief (4 of the 5 firing sites are located in relatively close proximity to the canyon edge). Third, although each of these analytes are fairly unique with respect to their specific gravities, partition coefficients, and sorptive capacities, their net redistribution appears relatively constant. These conclusions have positive ramifications for waste management and future remediation activities.

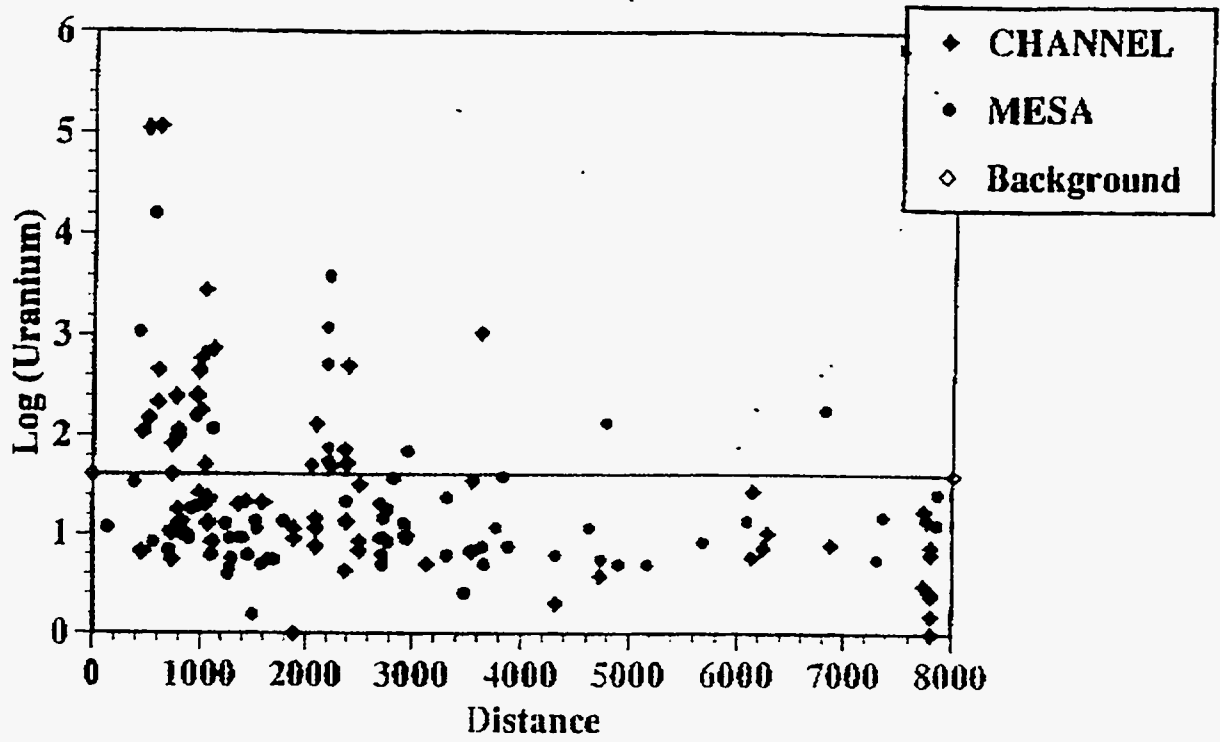


Figure 1. Uranium Concentration versus Distance in feet from Firing Sites.

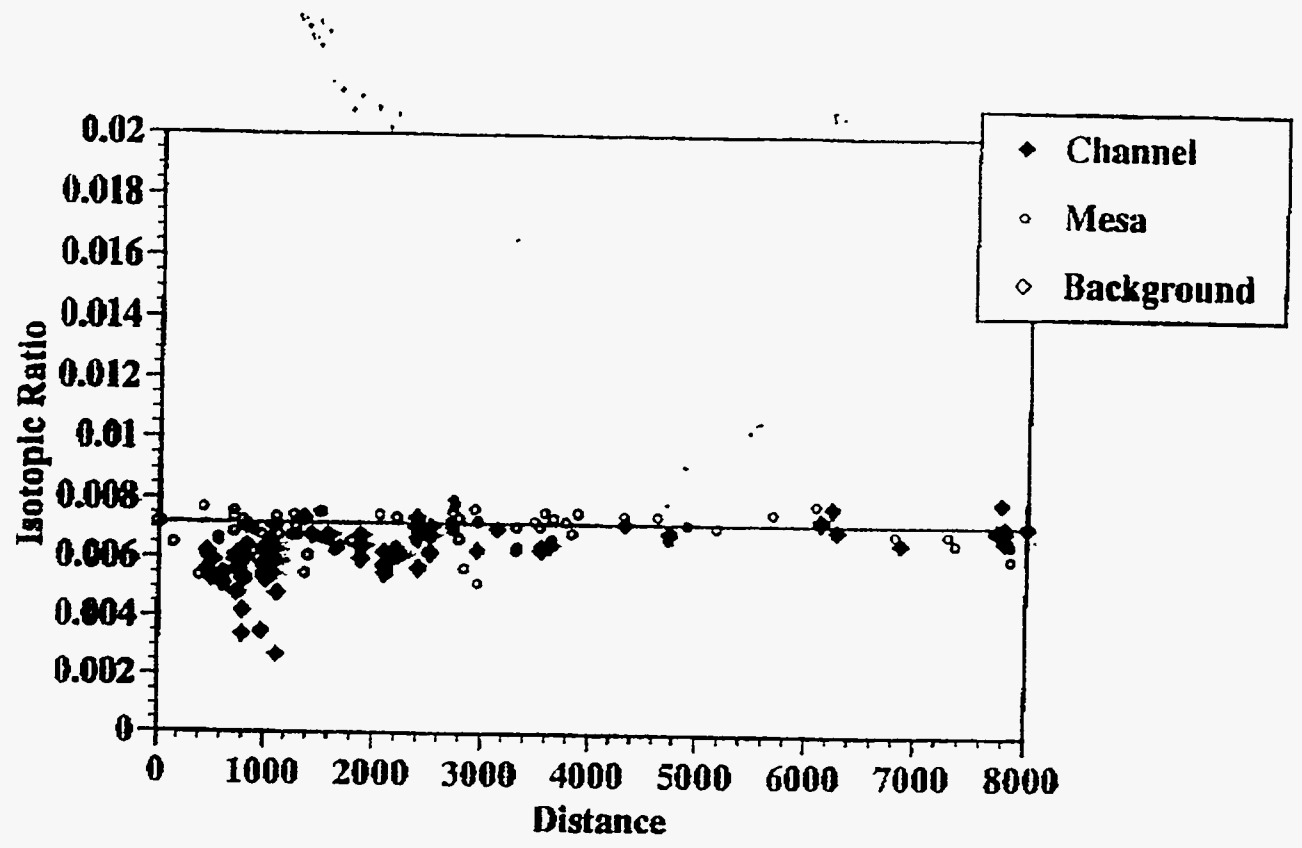


Figure 2. Uranium Isotopic Ratio versus Distance in feet from Firing Sites.

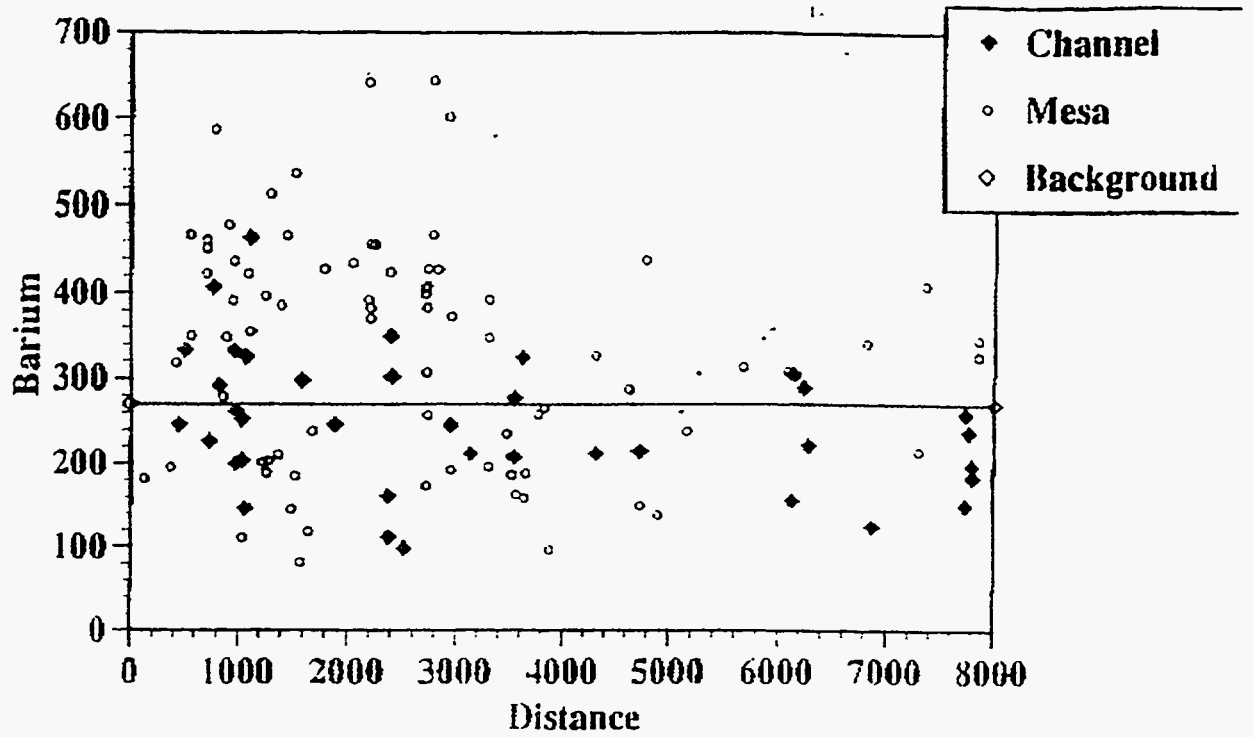


Figure 3. Barium Concentration in ppm versus Distance in feet from Firing Sites.

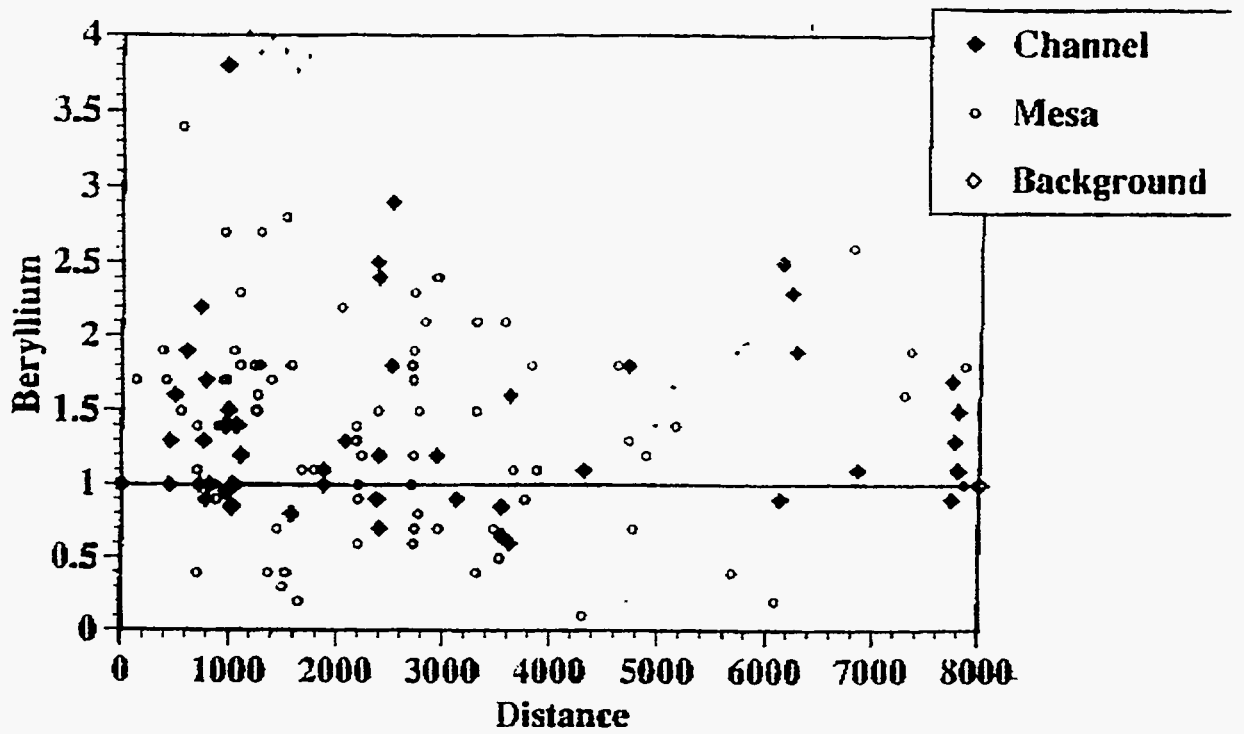


Figure 4. Beryllium Concentration in ppm versus Distance in feet from Firing Sites.

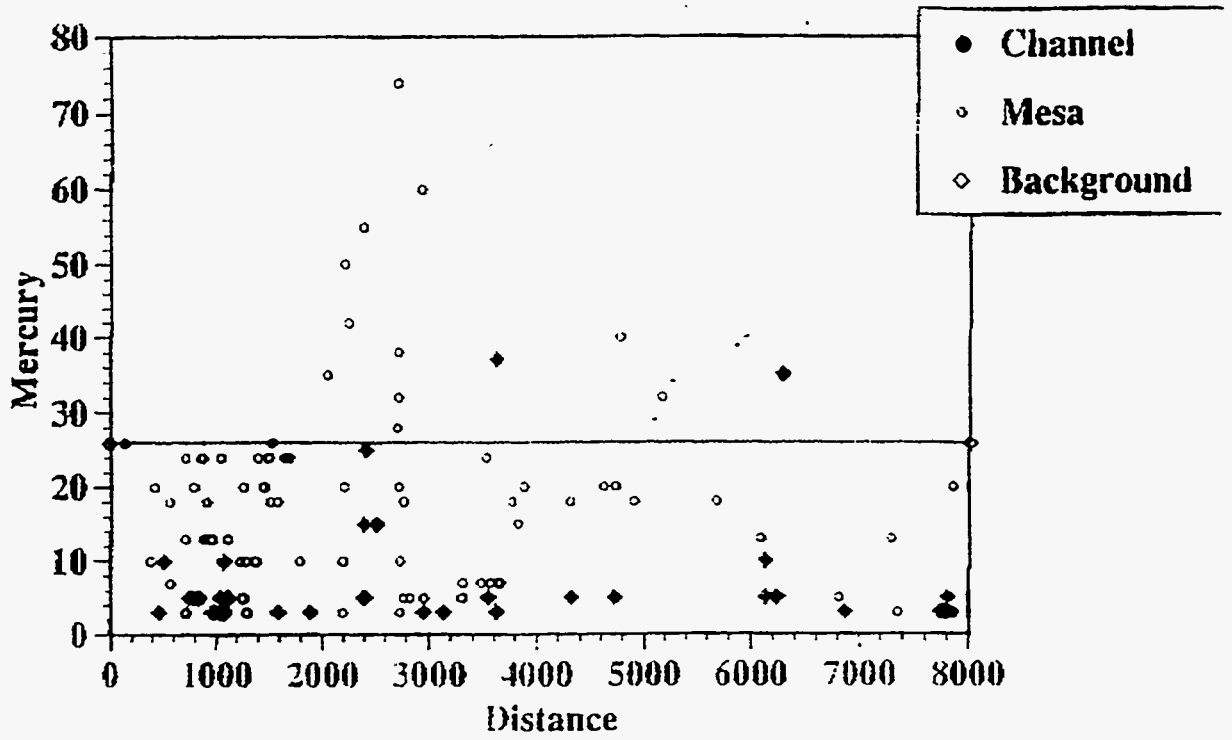


Figure 5. Mercury Concentration in ppb versus Distance in feet from Firing Sites.

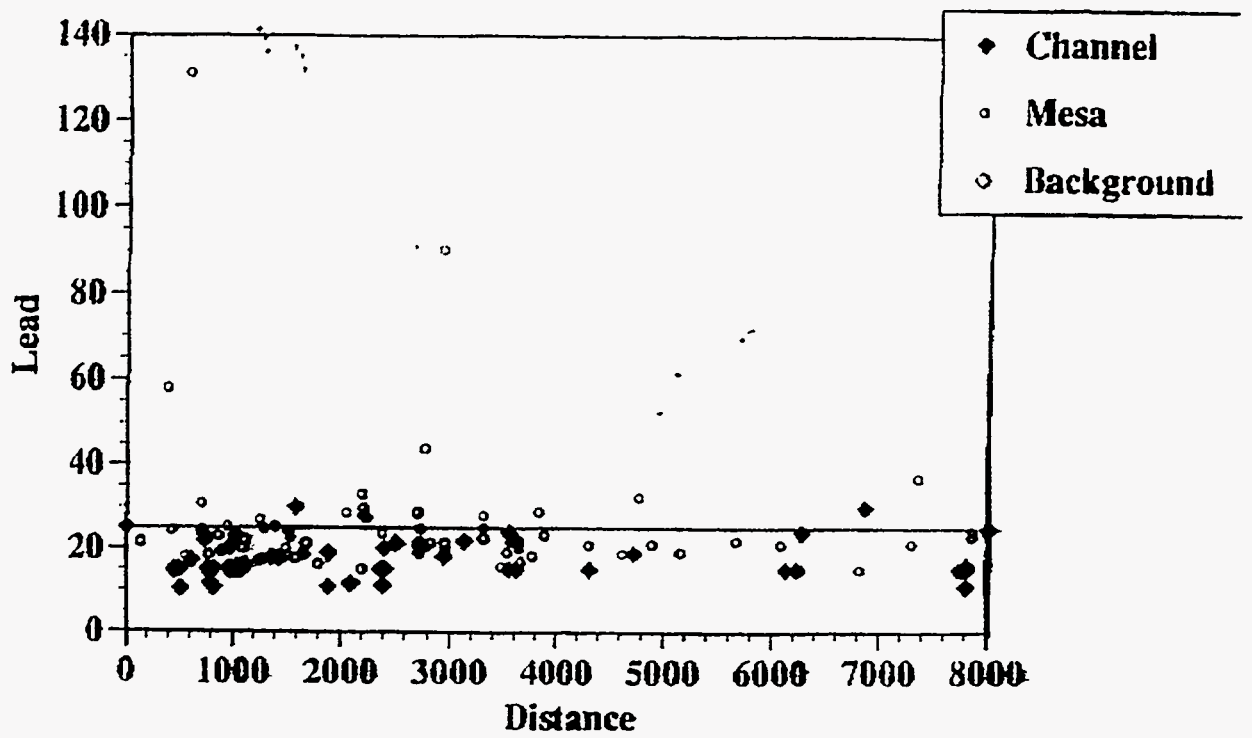


Figure 6. Lead Concentration in ppm versus Distance in feet from Firing Sites.

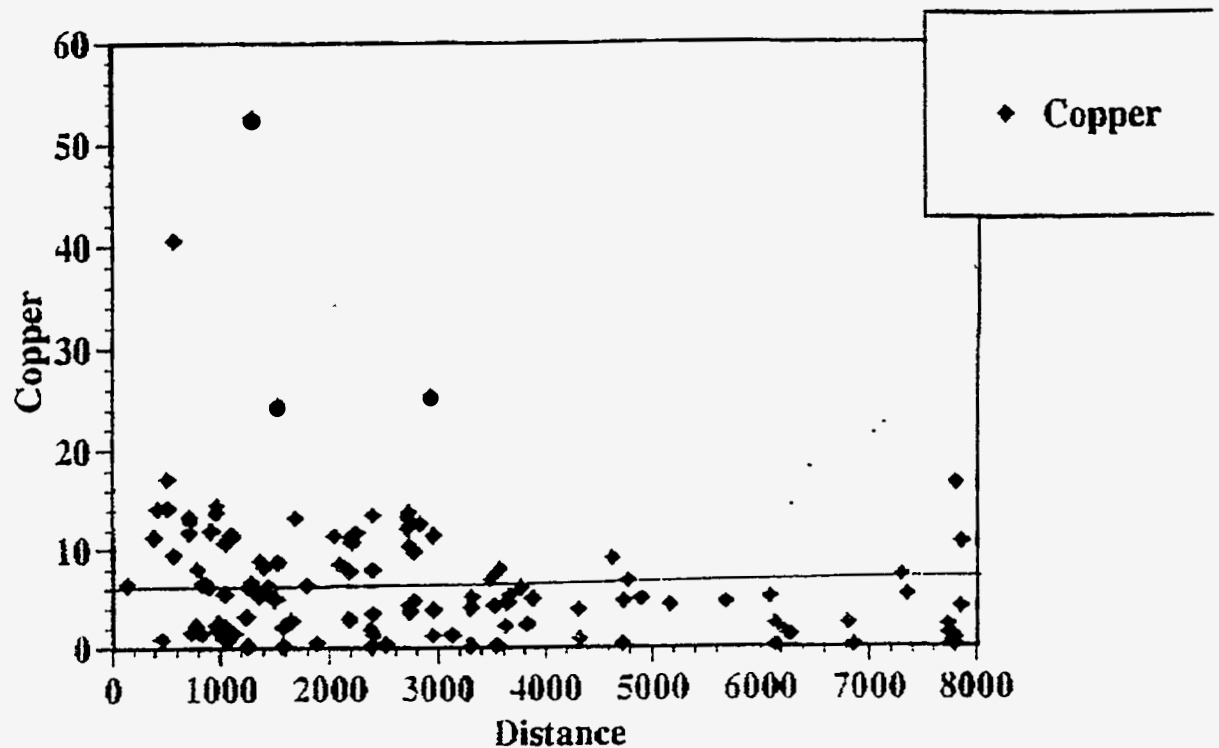


Figure 7. Copper Concentration in ppm versus Distance in feet from Firing Sites.

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