

Title:

HYDROLOGIC TRANSPORT OF DEPLETED URANIUM ASSOCIATED WITH OPEN AIR DYNAMIC RANGE TESTING AT LOS ALAMOS NATIONAL LABORATORY, NEW MEXICO, AND EGLIN AIR FORCE BASE, FLORIDA

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# **Hydrologic Transport of Depleted Uranium Associated with Open Air Dynamic Range Testing at Los Alamos National Laboratory, New Mexico and Eglin Air Force Base, Florida**

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## **Abstract**

Hydrologic investigations on depleted uranium fate and transport associated with dynamic testing activities were instituted in the 1980's at Los Alamos National Laboratory and Eglin Air Force Base. At Los Alamos, extensive field watershed investigations of soil, sediment, and especially runoff water were conducted. Eglin conducted field investigations and runoff studies similar to those at Los Alamos at former and active test ranges. Laboratory experiments complemented the field investigations at both installations.

Mass balance calculations were performed to quantify the mass of expended uranium which had transported away from firing sites. At Los Alamos, it is estimated that more than 90 percent of the uranium still remains in close proximity to firing sites, which has been corroborated by independent calculations. At Eglin, we estimate that 90 to 95 percent of the uranium remains at test ranges. These data demonstrate that uranium moves slowly via surface water, in both semi-arid (Los Alamos) and humid (Eglin) environments.

## **I. INTRODUCTION**

Both Los Alamos National Laboratory and Eglin Air Force Base (AFB) have a history of open air dynamic range testing, where depleted uranium is used both in weapons components evaluation testing as well as in life cycle testing of munitions in the Air Force's current inventory. Our two organizations voluntarily undertook investigations on the environmental impact of depleted uranium usage. Here we report on our results of studying depleted uranium movement in the surface water pathway. Even though uranium is initially introduced into the environment through detonation or by forceful impact, examination of particulate fallout data and comparison to surface water transport at Los Alamos has demonstrated that the surface water pathway is the dominant mechanism for uranium redistribution. Our aim is to describe uranium transport resulting from weapons testing operations, and to quantify the extent of migration on a watershed scale. Here, we describe our sampling strategies, report on results of laboratory and field investigations, and comment on our results in terms of mass balance calculations.

## **II. LOS ALAMOS NATIONAL LABORATORY**

### **A. Facilities Description and Depleted Uranium Usage**

Los Alamos National Laboratory, in north-central New Mexico, was selected for its remote location over 50 years ago as the site for development of the first atomic bomb. Much of its 43 mi<sup>2</sup> area sits above 7000 ft on a broad plateau of volcanic tuff above the Rio Grande, the master stream of the region, Fig 1. The Pajarito plateau is dissected into long, finger-like mesas by deep canyons, creating a rugged terrain. Annual precipitation of nearly 20 in., falls as snow and as rain, with 40 percent occurring during July and August during the height of the 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.

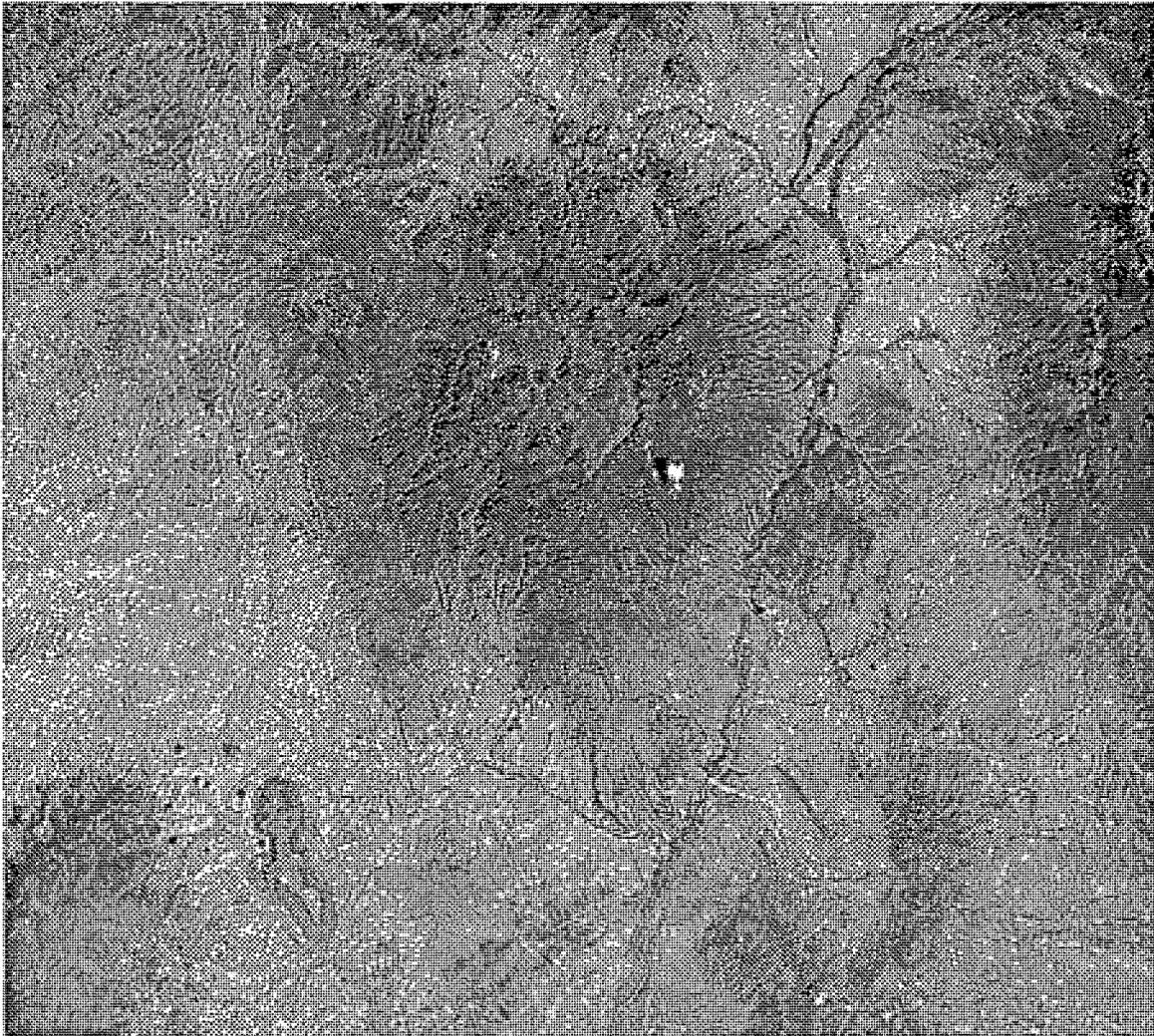


Fig. 1. 1975 LANDSAT scene of Los Alamos and surrounding area. Los Alamos sits on the eastern flank of the Valles Caldera (circular feature in center). Note location of the Rio Grande River (running from upper right to bottom middle) and Santa Fe, NM at the southwestern base of the Sangre de Cristo mountains (lower right side of image).

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 are tested at the U.S. Department of Energy's Nevada Test Site near Las Vegas. 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 explosively detonated, or is 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 large distance away from the firing pad. The explosive detonation process of 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).

## **B. 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 which uniquely distinguishes uranium associated with dynamic weapons testing from 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 called Potrillo Canyon. Potrillo Canyon was selected because of its small size (3.1 mi<sup>2</sup>), 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.

Field investigations 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 precipitation events without power or an operator. Investigations were expanded to collect samples of fallout particulates 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. 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 below firing sites were continuously agitated and periodically sampled to examine dissolution kinetics.

Depleted uranium-contaminated sediments were separated into individual grain sizes ranging from pebbles down 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.

### C. Results of Depleted Uranium Sampling in Soil, Sediment, Air and Water

In all, more than 750 contaminant measurements of fallout from air, 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  $\mu\text{g/g}$ , in soil from 2-5  $\mu\text{g/g}$ , 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  $\mu\text{g/g}$ , 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 mobilization of uranium away from firing sites. Uranium concentrations in runoff in the dissolved and suspended sediment phases were found to decline with downstream direction in the watershed, with the largest concentrations below two firing sites near the top of the watershed, implying both dilution and contaminant deposition with increasing hydrologic distance from firing sites. Leaching studies of uranium attached to channel sediments showed that uranium readily leached 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).

#### D. Mass Balance Calculations

Calculations were made to determine the amount of uranium currently coexisting on or attached to fluvial (stream) sediments in the watershed. Using average measured concentrations of uranium in fluvial sediments and subtracting off background concentrations of uranium, estimates were made of the uranium inventory in the channel, on banks, in point bars and alluvial fans, and in an area known as a discharge sink where sediment is preferentially accumulating in the watershed. For each of these five regions, the soil masses were multiplied by uranium soil concentrations above background to obtain uranium volumes. In this manner, estimates of uranium associated with fluvial sediments accounted for about 5 percent of the estimated total uranium expenditure of 35,000 kg.

From this calculation it may be concluded that most of the uranium mass 1) is not tied up in the fluvial sediments, 2) has already left the watershed, or 3) remains on or near firing sites. Flow and uranium losses can occur by vertical flow (infiltration) in the discharge sink or through horizontal flow out of the watershed. Infiltration and surface water losses are considered separately.

Examining the volume of uranium which enters the discharge sink, which is the main terminus for all flow and contaminants generated at the firing sites, there are dissolved and suspended sediment-uranium components. First consider the dissolved uranium component. Assuming an annual total inflow of 5200 m<sup>3</sup> (measured during 1990) and an average dissolved uranium concentration of 1.9 ppb (measured between 1984 and 1990), then 9.5 g of uranium annually are carried in the dissolved phase. Over 50 years of operation this would amount to an influx of about 0.5 kg of dissolved uranium transported into the discharge sink, or less than 1 percent of the estimated 35,000 kg source term.

Considering the suspended sediment component, the average annual suspended sediment load was calculated by assuming the suspended load to be 5 percent of the average discharge based upon visual observations of the volume of suspended sediment which was collected in cumulative samplers emplaced throughout the watershed. Using a range of 35,000 to 1,400,000 kg/km<sup>2</sup>-yr (Leopold and others, 1966) and multiplying by an average suspended sediment

uranium concentration of 8 ppm by weight (measured), the average annual uranium influx into the discharge sink range from 1 to 36.5 kg/yr. The combined dissolved and suspended sediment influx to the discharge sink over the 50 years constituted between 0.1 and 5 percent of the 35,000 kg uranium source term.

If large volumes of depleted uranium had exited the watershed through surface water transport at the outlet, a depleted uranium signature observable through inspection of the ratio of uranium-235 to uranium-238 is expected to have remained in the sediments in the lower half of the watershed. Because little depleted uranium signature was observed in sediments in the channel, banks, and floodplain downstream of the discharge sink, and it was inferred through chemical and historic aerial photographic data that there has been little transport across the discharge sink during the last 26 years, it was assumed that most of the uranium must remain in the watershed.

Another calculation was made to determine what the concentrations of uranium in runoff water should be if all the uranium expended were uniformly dissolved in precipitation on an annual basis. Considering 0.5 m of precipitation annually and that 80 percent of the precipitation is lost to evaporation, transpiration and infiltration, then

$$\begin{aligned} \text{Dissolved Concentration} &= 35,000 \text{ kg} / (0.2)(0.5 \text{ m})(8 \text{ km}^2)(50 \text{ yr}) \\ &\approx 1 \text{ ppm.} \end{aligned}$$

A dissolved concentration of one ppm is an underestimate because not all precipitation contacts the uranium; expected concentrations would be even higher. The dissolved concentration of 1 ppm exceeds observed dissolved uranium concentrations in runoff water by 2 to 3 orders of magnitude. Clearly, large dissolved uranium concentrations in surface water are not observed and dissolved transport in surface water is not a main uranium transport mechanism.

The hypothesis that most of the uranium mass left the watershed either by movement into the discharge sink (dissolved phase) or by flowing through the watershed outlet is rejected. Calculations show that the fluvial sediment contains about 5 percent of the expended mass. Therefore, the only plausible location for the remaining uranium is at or near the firing sites.

Results from an aerial radiological flyover in 1982 (Fritzsche, 1986) estimated that between 4 and 23 Curies of Protactinium-234m (Pa-234m) remained near three firing sites in the watershed, the variability dependent on the estimated vertical distribution. It is reasonable to assume equilibrium between Pa-234m and uranium-238 (U-238) because the half-life decay from uranium-238 to protactinium is short, of the order of about a half year, whereas the half life of U-238 is long, on the order of  $4.5 \times 10^9$  years. If equilibrium is assumed, an estimated 4-23 Curies of uranium remains at the three firing sites. Multiplying to convert to kilograms, the amount of

uranium remaining at the firing sites is calculated to range between 12,000 to 69,000 kg, bracketing the original estimate of 35,000 kg uranium expended in Potrillo Canyon.

Consider one final calculation. If all the 35,000 kg of uranium were situated at the three firing sites, then what magnitude of soil concentration would be expected? Assuming a contaminated region of 26,000 m<sup>3</sup>, which assumes uniform uranium concentrations down to 0.6 m, then

$$\begin{aligned}\text{Uranium Soil Concentration} &= 35,000 \text{ kg} / (26,000 \text{ m}^3)(19 \text{ g/cm}^3) \\ &\approx 72 \text{ ppm},\end{aligned}$$

where 19 g/cm<sup>3</sup> is the approximate specific weight of uranium. Unpublished surface soil studies reported concentrations of uranium ranging from 400 to 3400 ppm by weight at one of the firing sites, and unpublished surface and depth data at another firing site ranged from 560 to 4580 ppm by weight. Concentrations in the vertical dimension ranged from 2 to 75 ppm by weight to 3.7 m depth with the largest concentrations in the uppermost 0.6 m. Therefore, an average soil concentration of 72 ppm is consistent with measured concentrations at firing sites. The original estimated source term of 35,000 kg may even be slightly low.

### **III. EGLIN AIR FORCE BASE**

#### **A. Facilities Description and Depleted Uranium Usage**

The Eglin AFB military reservation is located in the Florida panhandle and occupies approximately 725 square miles within portions of three counties, Santa Rosa, Okaloosa and Walton, Fig 2. Eglin also controls 44,000 mi<sup>2</sup> in the adjacent Gulf of Mexico. Eglin is comprised of two physiographic settings, a coastal lowland of swamps, flatwoods, dunes, beach ridges and bluffs, and a western highlands of sandhills 50 to 200 feet in elevation cut by deep, narrow stream valleys. All of Eglin's depleted uranium test ranges are located in the western highlands. A close relationship between the numerous streams and shallow groundwater permits rapid infiltration of rainfall to recharge the groundwater followed by a slow release back into the surface water. This results in a fairly constant streamflow year-round. The humid, semitropical climate at Eglin AFB provides an average annual rainfall of almost 65 inches and a mean annual temperature of slightly more than 66 degrees F°. Heavy rainfall can be expected between the months of June and November, the hurricane season. (Becker and others, 1990 and 1994).

Research on using depleted uranium (DU) as a conventional Air Force munitions component began in the late 1960s. This early work sought to exploit the high density of DU in

armor-piercing applications. Records indicate that testing with DU penetrators was conducted at Test Area (TA) C-74L, the Gunnery Ballistics Facility, from 1973 to 1978. In late 1978, activities at TA C-74L were shifted to testing high explosive incendiaries. Subsequently DU testing was moved to Test Area C-64, the High Explosive Test Area, where it continues. Testing in the early years was associated with research and development of DU-containing munitions whereas current efforts are directed toward life cycle testing of ammunition in the active inventory. Life cycle testing is a periodic withdrawal and testing of ammunition to evaluate whether acceptable performance is being maintained throughout the entire storage life of the ammunition. Depleted uranium-contaminated sand has been periodically collected from these test ranges, particularly from the sand-filled target butts, mixed with concrete and water for stabilization, stored in metal drums at the test sites, and eventually removed for disposal at a low-level radioactive waste site at Barnwell, South Carolina.

## **B. Description of Field and Laboratory Activities**

Environmental monitoring of depleted uranium released into the environment as a result of range testing began at Eglin AFB with sampling of area streams in 1974, soils beginning in 1976, vegetation in 1986, and groundwater in 1987. In 1990, a review of the Eglin AFB DU environmental monitoring program for the period 1974-1988 was published (Becker and others, 1990). In 1988, a supplemental hydrologic monitoring system for surface runoff water and sediments was implemented to further evaluate the fate and environmental transport of depleted uranium. Cumulative samplers based upon the design in use at Los Alamos were installed in drainage pathways from each of the test ranges to assess depleted uranium transport. In addition, soil samples were collected at various depths in the soil profile, especially within surface drainage pathways; samples of surface water and sediments from local creeks, and groundwater from local wells were collected and analyzed for total and isotopic uranium.

Laboratory investigations consisted of performing grain-size analyses and solubility/leaching studies on DU-contaminated soils collected from the test ranges. Grain-size distributions were performed to determine the total uranium concentration as a function of particle size. Leaching/solubility experiments were performed to investigate the potential rates of uranium leaching from contaminated soils and sediments via rainwater. Deionized water was

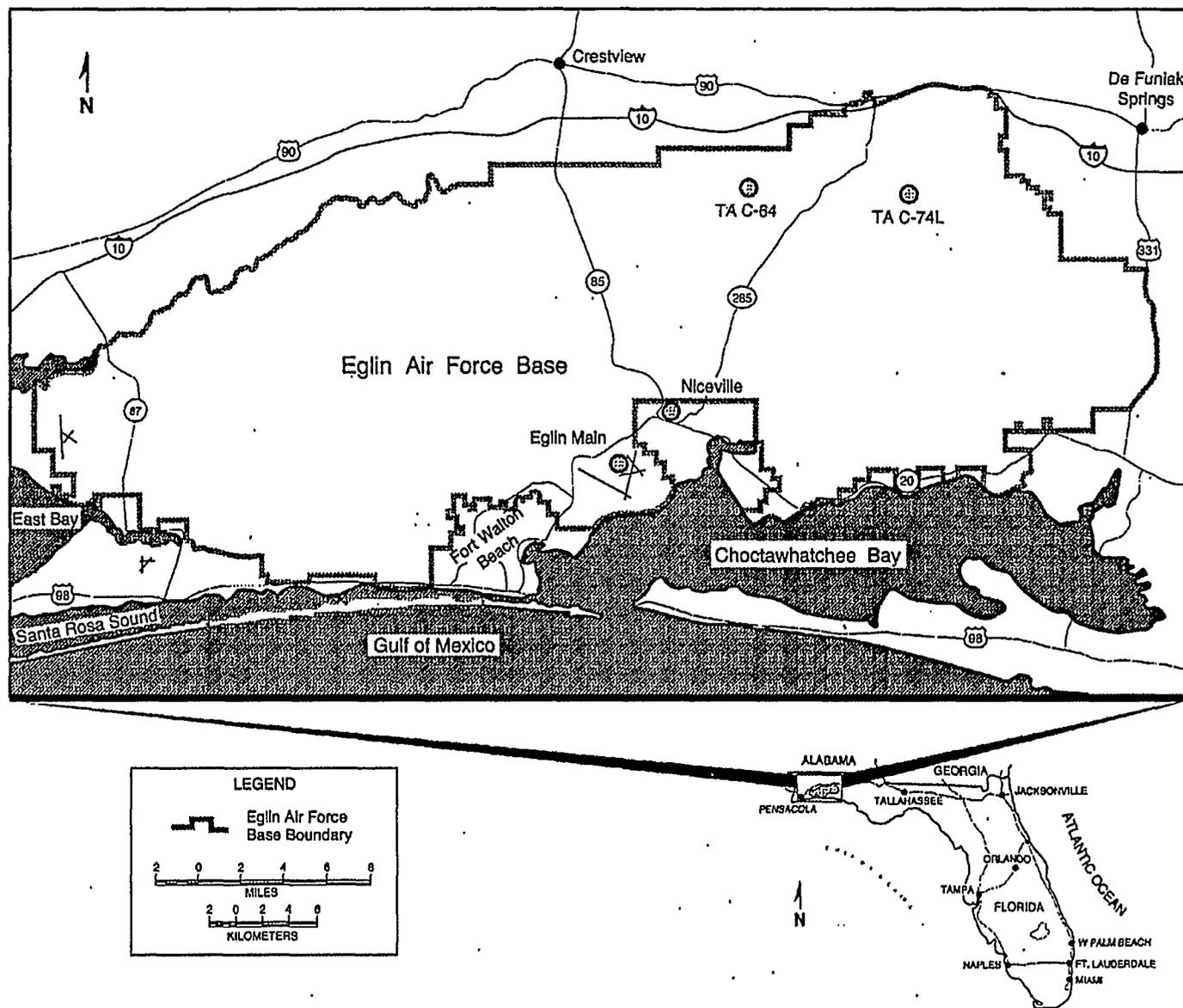


Fig. 3. Location map of Eglin Air Force Base.

adjusted to a pH of 4.65 to 4.75 to simulate the pH of natural rainwater measured in Florida (Brezonik et al, 1980). Ten grams of DU contaminated soil were mixed with 2 liters of the pH-adjusted deionized water and subjected to continuous mixing. The mixing was discontinued periodically for the brief intervals required to allow removal of 55 ml of the agitated mixture. The 55-ml samples were immediately filtered using a 0.45-micron size filter to separate sediment from solution. The filtered solution was then analyzed for total uranium content.

### C. Discussion of Results

Uranium concentrations in the natural background waters, soils, and sediments at Eglin AFB are, in general, very low as compared to many parts of the United States, with the uranium in both groundwater and surface water measuring in the tenths of parts per billion (ppb). Background concentrations in both soils and sediments at Eglin AFB generally ranged from 0.2 to 0.5  $\mu\text{g/g}$ , although in some locations on the reservation natural background uranium content was measured to be as high as 1.4  $\mu\text{g/g}$ . In areas where there is a predominance of silts and clays, such as in claypits, the natural background uranium concentration may be as high as 12  $\mu\text{g/g}$ . A background location rainwater runoff collector installed in the vicinity of an abandoned clay pit yielded background uranium concentrations in suspended sediment of up to and exceeding 10  $\mu\text{g/g}$ , suggesting that uranium has an affinity for the smaller sized clay particles.

Uranium's affinity for smaller sized particles was verified in the laboratory investigations on uranium concentrations as separated by grain-size within the soil samples collected. Although more than 90 percent of the soil collected at Eglin AFB sites was within the sand category (#20 to #200 sieves), the greatest concentrations of uranium were seen in the fine silt and clay particle sizes. A clear trend of increasing uranium concentration with decreasing particle size was established. In one sample from Test Range C-64, uranium concentrations varied from 25-350  $\mu\text{g/g}$  in pebbles and gravels, to 360-1090  $\mu\text{g/g}$  in coarse to medium sands, 1920-4420  $\mu\text{g/g}$  in fine to very fine sands, and 15,500  $\mu\text{g/g}$  in silts and clays, or a span of almost 3 orders of magnitude (Becker and others, 1994).

Laboratory experiments to investigate solubility and leaching potential of uranium at the pH of local rainwater showed that uranium particles present in soils and sediments in parts per million concentrations, will rapidly dissolve at concentrations in the parts per billion range at the acidic pH of the simulated rainwater. Firing site soil from Test Range C-64 with an initial uranium concentration of 451  $\mu\text{g/g}$  leached into simulated rainwater to a dissolved concentration of 3000 ppb and persisted at that concentration for the next 5 days (Becker and Vanta, 1992). Dissolution kinetics operate on the order of hours, Fig 3.

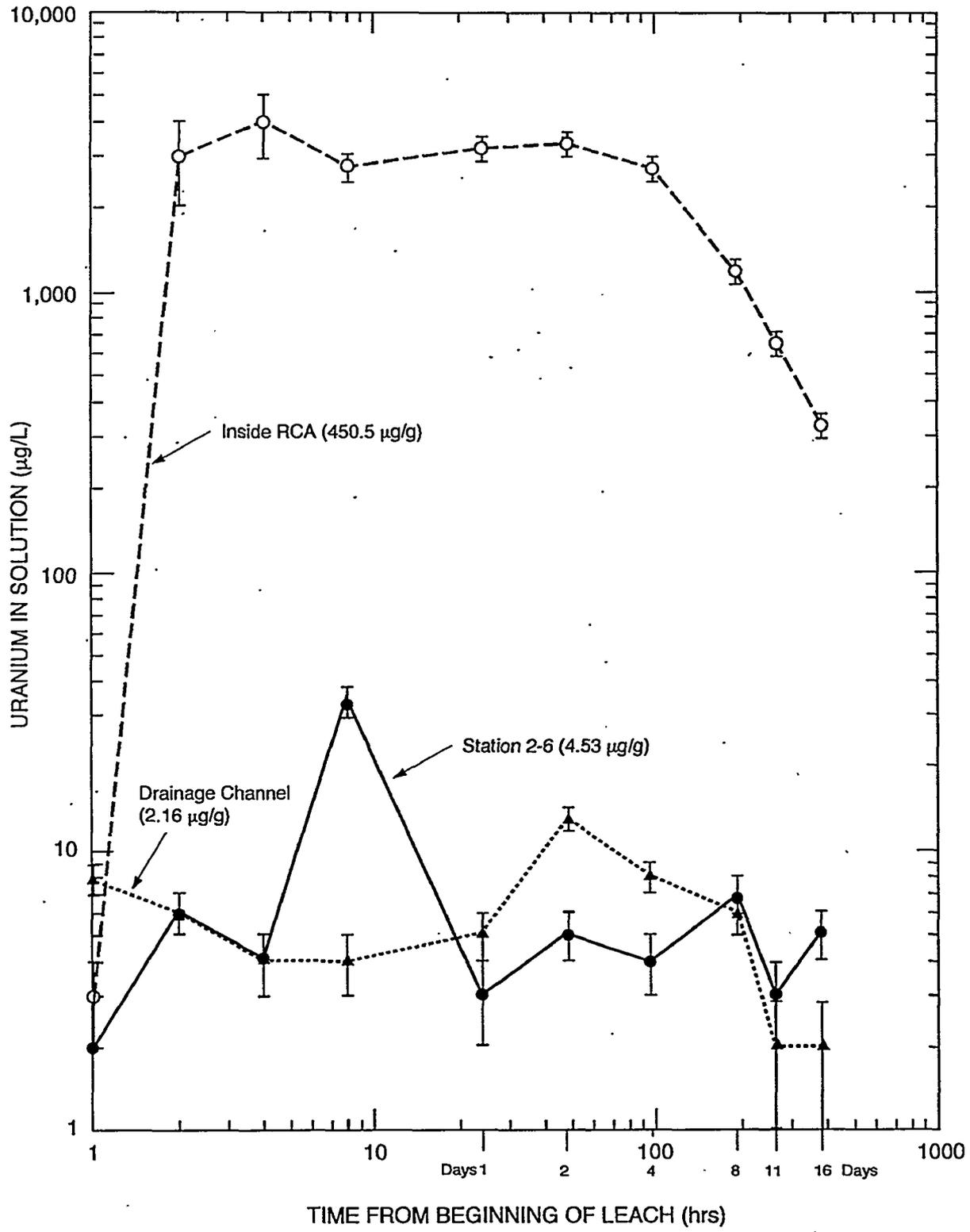


Fig. 3. Dissolution of depleted uranium from Ta C-64 radiation control area soils, Eglin AFB.

Field investigations of uranium concentrations in rainwater runoff water showed that uranium concentrations in the sediment particulate form exceeded the concentrations of dissolved uranium in the runoff water by 2 to 3 orders of magnitude. In runoff, dissolved uranium concentrations were measured ranging from below detection limits to 10 ppb at TA C-64, and from below detection limits to 15 ppb in runoff collected at TA C-74L. Uranium concentrations in the associated suspended sediment averaged 50  $\mu\text{g/g}$  (ppm) at TA C-64 and 200  $\mu\text{g/g}$  at TA C-74L (Becker and others, 1994).

Soil sampling conducted in the vicinity of the test areas showed that uranium concentrations generally declined with depth. The total depths of the holes sampled varied from 36 to 43 inches at TA C-74L and from 33.5 to 39 inches at TA C-64. In the Radiation Control Area (RCA) adjacent to the target butt at Test Range C-64, uranium concentrations declined from more than 1100  $\mu\text{g/g}$  at the surface to 0.8  $\mu\text{g/g}$  at the bottom of the hole at 39 inches. In all holes samples, the uranium concentrations at the deepest sampled point were within the range of background uranium and the isotopic ratios associated with most of these samples indicated the presence of some depleted uranium (Becker and others, 1994).

#### **D. Mass Balance Calculations**

Mass balance calculations were performed for the depleted uranium usage, disposal, and remaining inventory at TA C-64 and TA C-74L. We began with the supposition that any uranium remaining after target butt or remediation activities must remain close to or inside the RCA. This was based on analysis of sampling results for depleted uranium at various distances from the target areas, and finding soil concentrations for uranium at background levels. This supposition was also supported by the results from Los Alamos which predicted that most of the expended depleted uranium there remains close to their firing sites. Verification of this supposition was made by estimating the mass of remaining depleted uranium from best available records, computing what the concentration of uranium in soil would be if this mass remained in the RCA, and then comparing the computed concentration to actual measurements.

At TA C-64, records showed that about 56,430 kg of depleted uranium was expended, beginning in the 1970's and extending into the early 1990's. Periodic cleaning of the target butt and consequent disposal of expended depleted uranium has taken place, however there was no definitive information regarding the specific amount of uranium mass removed, just estimates based on predicted cleaning efficiencies. It is proposed that approximately 3 to 10 percent, or between 1692.9 and 5642.9 kg of depleted uranium is believed to remain at TA C-64 based on the following calculations (Becker and others, 1994). Assuming that the depleted uranium

concentration is largely confined to the Radiation Control Area at TA C-64 and that the depleted uranium concentration is fairly uniform down to a depth of 0.6 m, then the total volume of contaminated soil would be 1739.1 cubic meters. Using a specific gravity of 19 g/cm<sup>3</sup> for uranium:

At 3 percent depleted uranium remaining,

$$\text{Uranium Soil Concentration} = 1692.9 \text{ kg}/(1739.1 \text{ m}^3)(19 \text{ g/cm}^3) = 51.2 \text{ ppm.}$$

At 10 percent depleted uranium remaining,

$$\text{Uranium Soil Concentration} = 5642.9 \text{ kg}/(1739.1 \text{ m}^3)(19 \text{ g/cm}^3) = 171 \text{ ppm.}$$

These estimates were compared to the measured soil concentrations of uranium collected within the RCA in 1988. The average measured uranium concentration in the soil within the RCA during 1988 was about 160 ppm. This is within the range of calculated values and suggests that between 5 and 10 percent of depleted uranium expended at the range remains on-site exclusive of target butt contents, or visualized as a volume, between 0.15 and 0.3 cubic meter.

At TA C-74L, it is estimated from records that 3201 kg of depleted uranium remains within the RCA, which encompasses an area of 13,192 square meters. Again using a depth of contamination of 0.6 m and a uranium density of 19 g/cm<sup>3</sup>, the concentration of uranium in soil was calculated to be 21.3 ppm (Becker and others, 1994). This favorably compares with the average measured uranium concentration in soil in the RCA during 1988, which was about 64 ppm.

Comparing these mass balance calculations to actual data, runoff data collected at TA C-64 and TA C-74L during 1991 and 1992 showed that depleted uranium has moved from the RCAs primarily via suspended sediment. A sampler located approximately 100 feet outside of the RCA at TA C-64 collected an average of 50 µg/g uranium in sediment in runoff water. A sampler located approximately 110 feet outside of the RCA at TA C-74L collected an average of 202 µg/g uranium in sediment in runoff water. Uranium concentrations in soils and sediments declined with increased distance from the RCAs, indicating that the major portion of the DU inventory remains within the RCAs, even though our data shows that uranium is mobile. Uranium concentrations in soils were measured at background concentrations within the boundaries of the test ranges. Expected uranium concentrations in soils derived from mass balance calculations were relatively close to measured field values and certainly within the same order of magnitude.

#### IV. CONCLUSIONS

First, an attempt was made to develop the best estimate of the total mass of expended uranium from dynamic weapons testing from records, and develop a methodology to distinguish weapons uranium from indigenous uranium present from the native rock and soil. This was accomplished primarily through analyses for isotopic uranium, because the isotopic mixture of weapons uranium presents a unique signature. Field investigations and laboratory experiments were structured to investigate multiple aspects of depleted uranium transport in the environment. Broad spectrum sampling of soils and sediments on a watershed-size scale permitted the development of a comprehensive characterization on how uranium redistributes in the environment and development of a conceptual model on how uranium transports in surface water. Measurements confirmed that uranium was moving with every runoff event in concentrations that frequently well-exceeded established background concentrations. Uranium was also observed to move vertically into the soil profile in regions close to the firing sites. At both Los Alamos and at Eglin we observed definite patterns of declining uranium concentrations in soils and sediments with increasing distance from the firing sites. Additionally, Los Alamos data confirmed the initial hypothesis that uranium movement by airborne mechanisms was small.

Laboratory experiments enhanced our field investigations. Data showed that uranium has a preference for the smallest particle sizes, silts and clays in particular. Actually, uranium distribution by particle size is bimodal, distributed by the largest mass in the large chunks and fragments created by the detonation or impaction, and by the largest concentration in (on) the silts and clays. Simulation of dissolution of uranium particles revealed that, given a sizable uranium concentration, dissolution can be great, elevating the uranium concentration of the contacting water to thousands of ppb. Dissolution is also rapid, on the order of hours, which is the timeframe of most rainfall events.

Mass balance calculations corroborated results from the laboratory and field programs. Calculations on expected uranium concentrations in soils were based on established or suspected contaminated areas, assumed uniform uranium concentrations down to 0.6 m, and compared to actual measurements on uranium in soils at these sites. Comparison between calculated and observed uranium concentrations were quite good, and within the same order of magnitude in all instances. At Los Alamos, results were corroborated by independent analyses. Interpreting these results, we show that in fact very little of the total expended mass has been transported, by either surface water or airborne mechanisms. Uranium transport, although measurable, is slow when one considers that some of the firing sites at Los Alamos have been active for over 50 years, and that at Eglin depleted uranium from munitions tests has been present for over 25 years at TA C-74L.

Perhaps the most interesting conclusion from these investigations is the similarity in the slow rates of uranium transport between the semi-arid climate at Los Alamos and the humid analog at Eglin. Eglin receives more than 3 times the average annual precipitation compared to Los Alamos, and precipitation is the predominant mechanism behind the sediment and contaminant transport process. Yet, we observe no sizable mass movement of uranium away from Eglin firing sites, as evidenced by uranium at background concentrations in sediments at distance away from their firing pads. As well, measured uranium concentrations in the sediments and waters of their onsite streams were at background values.

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