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Title:

A BRIEF REVIEW OF ENVIRONMENTAL TRANSPORT OF TRITIUM
AT THE LOS ALAMOS LLRW DISPOSAL FACILITY

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Submitted to:

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Title: A Brief Review of Environmental Transport of Tritium at the Los Alamos LLRW Disposal Facility

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Abstract:

A large data base of tritium in the environment at the Los Alamos National Laboratory LLRW disposal facility, Area G, is summarized. Tritium in surface and subsurface soil moisture indicates three disposal areas significantly elevated above background with the highest results around the currently used tritium disposal shafts. Airborne concentrations are well below appropriate concentration guidelines but vary seasonally by over an order of magnitude. Surface efflux measurements show seasonal and diurnal time dependent variations with a positive correlation with ambient temperature. The integral of the surface efflux is estimated as a release on the order of ten Curies/year. A subsurface tritium plume observed in the early seventies has since been greatly reduced indicating an effective half-life in the environment which is significantly shorter than the radiologic half-life. Subsurface migration from the mesa top disposal facility has resulted in tritium in the canyon water immediately adjacent to the disposal site which is marginally elevated above background. The hydrogeologic diffusion near the tritium shafts is compared to model results to predict the vertical extent of the tritium migration. Field work to define this limit and additional quantification of tritium sources and migration is continuing.

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Introduction

Los Alamos National Laboratory (LANL) is located in a semi-arid high altitude region of over one hundred square miles on the Parajito Plateau. This plateau sits on the eastern edge of an inactive volcanic cauldron and slopes downward going west to east from about 7400' to below 6000'. The local topography forms a complex network of finger-like canyons and mesa tops with a typical mesa height of hundreds of feet from an adjacent canyon floor. The active Low Level Radioactive Waste (LLRW) disposal facility, Area G at TA54, is located on a narrow (<2000' N to S) mesa top at an average elevation of 6800', with about a 100' drop to the adjacent canyons, Canada del Buey to the north and Pajarito Canyon to the south. The unsaturated zone is estimated to be 300m deep at the disposal site, minimizing the risk of transport to saturated ground waters.

Operations at the Area G facility includes disposal of solid radioactive waste since 1957. Waste inventory data is limited prior to 1971. Inventory data is relatively complete since then, and very detailed after 1990. Tritium contaminated waste has been disposed in specifically designated shafts since about 1970, but was not segregated prior to that time. Tritium disposal inventory exceeds 10^5 Ci/yr in several years with a maximum annual disposal of 0.22×10^6 Ci in 1986.

The Waste Management (WM) Program at LANL is dedicated to a complete characterization of the disposal site and its influence on the environment. This characterization includes an enhanced annual environmental surveillance program at Area G, several on-going special studies to monitor radioactivity and geologic transport parameters, studies specifically to support the Area G Radiologic Performance Assessment and studies done in collaboration with the Environmental Restoration (ER) Program. The WM program operates active disposal pits and shafts while the ER program has responsibility for the legacy disposal pits and shafts, those no longer in active use for disposal.

Tritium movement in the environment is particularly important because it moves as tritiated water. Tritium is a dominant waste component migrating through the environment in the vapor form and therefore it acts as a tracer for understanding water vapor movement in the atmosphere and especially in the subsurface unsaturated zone.

Review of Data

Throughout the years a number of special studies as well as routine environmental surveillance measurements (see for example, [LANL,92]) have recorded tritium concentration in the environment of the Area G waste management facility as summarized in the data sources listed in Table I.

An early study of tritium migration in the soil [Purtymun,74] was motivated when soil from new shaft drillings was found to be highly contaminated. The adjacent older shafts were identified as the source and the underground plume was mapped. The plume showed a broad, somewhat isotropic spread with some preference for transport along the horizontal contact lines between geologic units. The maximum horizontal extent of the plume was about 100 ft as measured at a time estimated to be about 3 years after the disposal of the tritium source.

Recent measurements of tritium surface efflux or evaporation from this area show a greatly reduced tritium level compared to levels in the 1970 study. Comparisons of the 1970 soil concentrations and the 1994 soil efflux measurements are planned to quantify this relationship. Recent measurements of tritium in the soil at a nearby TRU remediation project still show elevated tritium levels with a spatial distribution consistent with horizontal migration from these older shafts as well as vertical migration up from an old disposal pit operated in the days prior to waste segregation. Recent soil moisture tritium measurements from core cuttings in a new shaft field also suggest upward movement of the tritium migrating from a deeper disposal source.

Tritiated water vapor in air has been measured monthly at several site perimeter stations at Area G since 1984 and at one station since 1976. Results from 1984 to 1992 for two colocated samplers are shown in Fig.1. An exponential fit to the tritium data versus time shows a best fit effective decay constant of 0.24/yr, corresponding to an effective half-life of only about 3 years. Comparing this to the radiologic decay constant for tritium and noting that $1/\tau_{\text{eff}} = 1/\tau_{\text{env}} + 1/\tau_{\text{rad}}$, this data suggests that the environmental dispersion decay constant, τ_{env} , is 0.18/yr. This implies an environmental transport dilution time for the Area G sources is about $1/0.18 = 5.4$ years, or a half-life due to environmental dilution is 3.8 years.

The results depicted in Fig.1 are for samplers about 700m. in the dominant downwind direction from the currently active disposal shafts for tritium. The contribution from the disposal operations is evident and a seasonal variation is strongly evident although not consistently present in every year. The annual peak for airborne concentrations occurs in the September time frame. This appears to be due to a balance between the emission source, the soil surface efflux due to evaporation which increases with temperature, and dilution in the air which is proportional to atmospheric

turbulence. This turbulent dilution also increases with temperatures and is maximum in the summer months.

Data clearly shows an annual variation in the soil efflux due to evaporation [Abeele,87] which acts as the source term for the airborne activity. Comparisons of this efflux data with average monthly temperatures shows that a linear relationship between soil efflux or evaporation and temperature explains about 70% ($r^2 \sim 0.7$) of the annual variation of the soil efflux of tritium.

Soil moisture has been collected periodically on a site-wide grid with points every 200' and analyzed for tritium. A contour plot of the results from a 1989 survey is shown in Fig.2 and shows that there are three areas above background at Area G. The hottest area encompasses the shafts actively used for tritium disposal in the south central region of Area G. The second highest level is seen near the tritium shafts used in the early 1970s and examined in the study discussed above [Purtymun,74]. The third spot was only slightly elevated and occurs over one of the earliest disposal pits at Area G. Records were incomplete on inventory at these times, and segregation of tritium waste was not practiced. Recent measurements of tritium surface efflux from this region shows it is just barely elevated above the variation in background levels.

Each of the three hot spots identified in the 1989 soil sampling program were evaluated recently in a tritium surface efflux measurement study performed by RADIANT Corporation [Eklund,et.al.,94] for the WM Program. Our discussion focuses on results from the hot spot at TR1, the area around the active tritium disposal shafts. A plot showing the efflux results as contours and the sample locations imposed on a map of the area is shown in Fig.3.

The area which was identified as a single hot spot on the coarse sampling grid across Area G in Fig.2 is now resolved into two separate plumes each with some distinct spatial structure. Tritium falls to background levels at a distance of about 125' from the 3 disposal shafts with the greatest tritium inventory. The subsurface tritium plume is limited to the mesa top except for a small adjacent area in Parajito Canyon which is slightly elevated (2-3 times) above background as seen in the lower center area of the figure.

Some of the samples taken at different times but at repeat locations show a temperature dependence for the tritium efflux, which seems to be in agreement with the temperature dependence seen in the earlier studies done on annual variations [Abeele,87]. This analysis is continuing.

Analysis of Data

This analysis focuses on the tritium efflux data from the maximum hot spot area designated TR1 as shown in Fig.3. The results from a 3-D

computational model for tritium transport by diffusion are shown in Fig.4. Comparing Figures 3 and 4 shows reasonably good agreement between the field efflux data and the surface concentrations predicted by the simple model. The computational volume on the left in the figure shows the full range of results (log scale) along the computational surface. Higher contour levels are shown in the figures to the right and indicate the positions of the highest tritium inventory in two of the shafts and its subsequent dilution from three shafts throughout the volume.

The computational model solves the 3-D diffusion equation

$$\frac{\partial C}{\partial t} = D \left(\frac{\partial^2 C}{\partial x^2} + \frac{\partial^2 C}{\partial y^2} + \frac{\partial^2 C}{\partial z^2} \right) + \frac{C_w(x)}{\tau} \quad (1)$$

with the empirical coefficients assumed as a constant diffusion coefficient, D , and the shaft-specific release time constant, τ , and inventory concentration, C_w , which is nonzero only at the shaft locations. The source concentration of tritium is determined from the annual inventory data base. From this with the operating times of the disposal shafts the approximate depth for each years tritium disposal is known and the 3-D source location is input to the model. The simulation is run explicitly from the time of the oldest tritium disposal to the current date with the results as seen in Fig.4 considering the inventory of the three shafts with the greatest inventory. The values for shaft release time constants and the diffusion coefficient (500ft²/year) were chosen to best match the field results and in the following sections the significance of the diffusion value is discussed. The value for τ at the newer steel lined shaft #154 was taken to be 5 times larger than the value for τ at the older unlined shafts, #150 and #151.

An analytic solution is examined for comparison of the diffusion coefficient chosen in the numerical modeling effort. The 1-D planar solution is used as a rough approximation to obtain a characteristic scale length for diffusion in the field. The solution to the problem for transient diffusion of C with a specified flux of $q = -d\nabla C$ as a boundary condition [Incropera and Dewitt,81] is

$$C(x,t) = C_o + \frac{2q}{\pi^{1/2}d} (Dt)^{1/2} \exp\left(\frac{-x^2}{4Dt}\right) - \frac{qx}{d} \operatorname{erfc}\left(\frac{x}{2\sqrt{Dt}}\right) \quad (2)$$

where the flux q is proportional to the source term, C_w/τ . The coefficients, d and D are equivalent for the case of mass diffusion as governed by Eqn (2).

The analytic solution shows that the field magnitude depends on the flux boundary condition (the tritium release rate in this case) but the field distribution depends only upon the diffusion coefficient. A surface plot of the analytic solution shown as $C(x,D)$ for a fixed time ($\Delta t = 10$ years, an average time of tritium disposal in these shafts) shows that the solution has a fall-off at a characteristic distance from the source which depends upon D . Working backward from the fall-off scale length observed in the field data of about 125', the analytic solution shows that this occurs for dimensionless scale length, $L_s \sim 70$ where $L_s = \sqrt{Dt}$. For the average disposal duration of 10 years, this implies a diffusion coefficient of $D \sim 500 \text{ ft}^2/\text{yr}$. This confirms the value for the empirically chosen diffusion coefficient and supports the hypothesis that a simple diffusion law is governing the tritium transport.

The magnitude of the empirically determined diffusion coefficient can be compared to values expected on a physical basis to help our understanding of the important physical transport processes. The diffusion coefficient is considered as a sum of contributions from classical diffusion modified for a porous media D_{pm} , and from diffusion driven by oscillatory pressure sources, D_{osc} as

$$D = D_{pm} + D_{osc}. \quad (3)$$

An expression for a porous media diffusion coefficient [Koorevaar,83] is

$$D_{pm} \sim \frac{1}{2} \epsilon D_{bc} \quad (4)$$

where D_{bc} is the binary collision theory diffusion coefficient, ϵ is the porosity, and the 1/2 is a rough correction for tortuosity. The binary diffusion coefficient for water vapor in air at standard pressure is $D = 2.6e-5 \text{ m}^2/\text{s}$ [Incropera, DeWitt,81]. We neglect the small pressure correction for the altitude at Los Alamos.

With $\epsilon \sim 0.5$, this gives a porous media diffusion coefficient of about $5.e-6 \text{ m}^2/\text{s}$, or $1500 \text{ ft}^2/\text{yr}$. This is a factor of three too large compared to the diffusion coefficient derived from the comparison of the empirical data with the numerical computation or with the analytic solution. One interpretation of this discrepancy, if these analyses are correct, is that the expression in Eqn (4) to correct the binary diffusion coefficient to that in a porous media is not correct and a greater reduction due to the porosity needs to be considered.

An expression for the pressure oscillation-driven diffusion coefficient can be derived by considering the pressure gradients driven by pressure fluctuations at the mesa surface and attenuated through the porous mesa volume. These fluctuating pressure gradients drive fluctuating velocities, v , over a finite period, T , which leads to a characteristic diffusion step size, $L_d = vT$. In this case a diffusion coefficient associated with a single dominant mode (velocity component and period) is considered and generally written as

$$D = v L_d = v^2 T. \quad (5)$$

The largest amplitude pressure oscillations are associated with diurnal fluctuations so the period of concern is 24hrs, and the maximum velocity component is that associated with this diurnal pressure variation. The velocity driven by pressure gradients in a porous media is given by

$$v = - \frac{k}{\mu} \nabla p. \quad (6)$$

As will be seen, this is a negligibly small number for convective flow in the Area G subsurface and suggests that convective flow can be ignored in vapor phase contaminant transport under Area G. The oscillating component can however contribute to a diffusion effect which may be important as in the following.

The pressure gradient is approximated as $\nabla p \sim \Delta p_s / \delta$, the observed pressure fluctuation amplitude at the surface, Δp_s , divided by the pressure attenuation distance in the porous media δ . This distance is derived from the solution of the governing pressure equation and is well known [Neeper,91], given approximately by

$$\delta = \sqrt{\frac{T k}{\pi \mu}} P_o. \quad (7)$$

Combining these expressions gives the diffusion coefficient due to the pressure oscillation,

$$D = v^2 T = \left(- \frac{k}{\mu} \nabla p \right)^2 T = \left(- \frac{k}{\mu} \frac{\Delta p_s}{\delta} \right)^2 T, \quad (8)$$

and this results in

$$D_{osc} = \pi \frac{k}{\mu} \frac{\Delta p_s^2}{P_o}. \quad (9)$$

Note that the period, T , cancels and so becomes irrelevant in the final expression. The expression is linear with the local permeability so that an order of magnitude change in k , which is possible between different geohydrologic units in this area, leads to a proportionately large change in the vapor diffusion coefficient.

Based on field observations a reasonable maximum for Δp_s is $\Delta p_s \sim 10^3 \text{Pa}$ for $T = 24 \text{hr}$. Using site specific values, $k \sim 10^{-12} \text{m}^2$, $\mu \sim 2 \times 10^{-5} \text{Ns/m}^2$, and $P_o \sim 800 \text{mbar} = 8 \times 10^4 \text{Pa}$, we find that the velocity driver

by this dominant diurnal mode from Eqn(6) is about 20 cm/day confirming our notion that convection is a negligible contribution to transport, while the diffusion result for D_{osc} from Eqn(9) is

$$D_{osc} \sim 2 \times 10^{-6} \text{ m}^2/\text{s} = .560 \text{ ft}^2/\text{yr}. \quad (10)$$

This value is significant and in remarkably good agreement with the diffusion coefficient numerically determined to best fit the field data and derived in the analytic solution. This suggests that the pressure driven oscillation diffusion is a viable candidate for the actual physical mechanism driving tritium transport in the mesa at Area G.

The classical diffusion coefficient with porosity correction in Eqn(4) was found to be larger than the value consistent with the field data, therefore this is also a viable candidate to explain the observed tritium transport. However, the discrepancy in that case suggests that the porosity correction factor may be a greater reduction than that predicted by Eqn(4) and should be reevaluated.

Conclusions

Several conclusions can be made regarding airborne transport and geologic transport for site characterization during the operational phase and for the site Performance Assessment during the post-operational phase.

Implications of this study for airborne transport concern annual site boundary airborne concentrations of tritium in relation to limits in DOE orders, integral release rates of tritium as related to the RAEM (radiologic air emissions monitoring) program, and the tritium release rate per source which impacts the WAC (waste acceptance criteria).

The maximum airborne concentrations of tritium at the fenceline (of order 4.e-9 Ci/m³ in 1986) corresponded to about 0.4 mrem/yr in that year, conservatively (but not realistically) assuming continuous occupancy at the fence perimeter. These concentrations are several orders of magnitude below the DOE DCG (Derived Concentration Guide) for tritium in an uncontrolled area, and the recently observed airborne concentrations have been reduced by several orders of magnitude below the maximum levels observed in 1986.

The integral release rate of tritiated water vapor from the disposal site has been calculated from the field data to be about 14 Ci/yr [Eklund,et.al.94] but will be slightly less, about 10 Ci/yr, after temperature and other environmental factor corrections are complete. This release corresponds to a conservatively calculated dose rate at an off-site location (where $\chi/Q = 1.e-6$ s/m³) of about 5.e-6 mrem/yr. Seventy percent of this release is from the top of the three disposal shafts with the largest tritium inventory, and 90% from a subsurface plume within 100-200 feet from these shafts.

The data on airborne tritium concentrations together with the data on tritium efflux could be used to define a tracer study and to determine the validity of atmospheric transport models. Such an exercise is planned for the future. The measured release rates, airborne concentrations and projected doses suggest that the current disposal practices for tritium are effective in minimizing the dose to the public and are consistent with the ALARA (As Low As Reasonably Achievable) philosophy.

Implications for geologic transport concern the diffusion of vapor at the mesa-top disposal site. The diffusion coefficient value in the numerical and analytic models which best matches the field data was found to be about $2 \times 10^{-6} \text{ m}^2/\text{s}$ or $500 \text{ ft}^2/\text{yr}$. Diffusion due to pressure oscillations or classical diffusion with porosity correction are both the right order of magnitude to account for the observed dispersion of tritium at the site.

Convective vapor phase flow is seen to be negligible, and liquid phase transport is expected to be too slow at the observed moisture content to play a role in the observed tritium movement. This implies a somewhat symmetric plume is expected and the vertical extent of the tritium plume should be less than 200'. Field measurements to confirm this have been made and analyses are underway.

If we take the effective environmental half-life ($T_{\text{Env}} \sim 4 \text{ yrs}$) observed for the airborne tritium and assume it applies to the subsurface tritium, then the tritium is dispersed sufficiently rapidly into the environment so that the concentration of tritium eventually reaching aquifer (deep ground) waters or alluvial (surface) waters will be negligible. A more detailed calculation of the long-term transport to the ground waters is warranted in support of the Area G Performance Assessment.

Acknowledgements

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Table I. Data base for tritium migration at Area G.

Routine Environmental Surveillance

- Tritium in atmospheric moisture
 - 6 locs around Area G (~84-'94)
 - 5 locs added near TRU remediation (1993+)
 - 4-6 locs being added (1994+)
- Tritium in moisture: soil / sediment / surface waters
 - sediment at 9 locs since '82
 - H3 contours in near surface soil (1993+data)
 - H3 in water samples. 5-10 locs / yr since '82
- Tritium in honey/bees - 1 loc at Area G (~1984+)
- Enhanced monitoring of vegetation, mammals, etc.. (1994+).

Environmental Surveillance Special Studies

- Tritium in soil/sediment moisture
 - H3 contours in surface soil (3depths),
LAUR-9656, 1983 (1980data)
 - H3 contours in surface soil, unpub. (1989data)
- Tritium and moisture in horizontal boreholes under old pits
S. McLinn unpublished study - 1992
 - Tritium in vegetation - ~16 samples, 1980, '85, '86.
 - Abeele'87 (1980-83data) - H3 in air, soil efflux, in soil

Core Drilling Samples

- Purtymun'74 study (1970data)
 - H3 in soil moisture around shaft 13 ($Q(H_3) = ?$)
- shaft field, shafts 235 - 253
 - H3 in soil moisture in shaft core samples
- TRU remediation
 - pre-op survey, H3 in soil moisture around pad 1

Surface efflux measurements

- RADIAN - tritium ground efflux measured
 - throughout Area G and at 3 hotspots (1993)
 - at specific shafts, in canyons & weak data areas (1994)
- CST-LANL - to continue RADIAN type measurements for
 - routine monitoring of high level tritium shafts (1994+)
 - establish seasonal efflux variation (1994+).

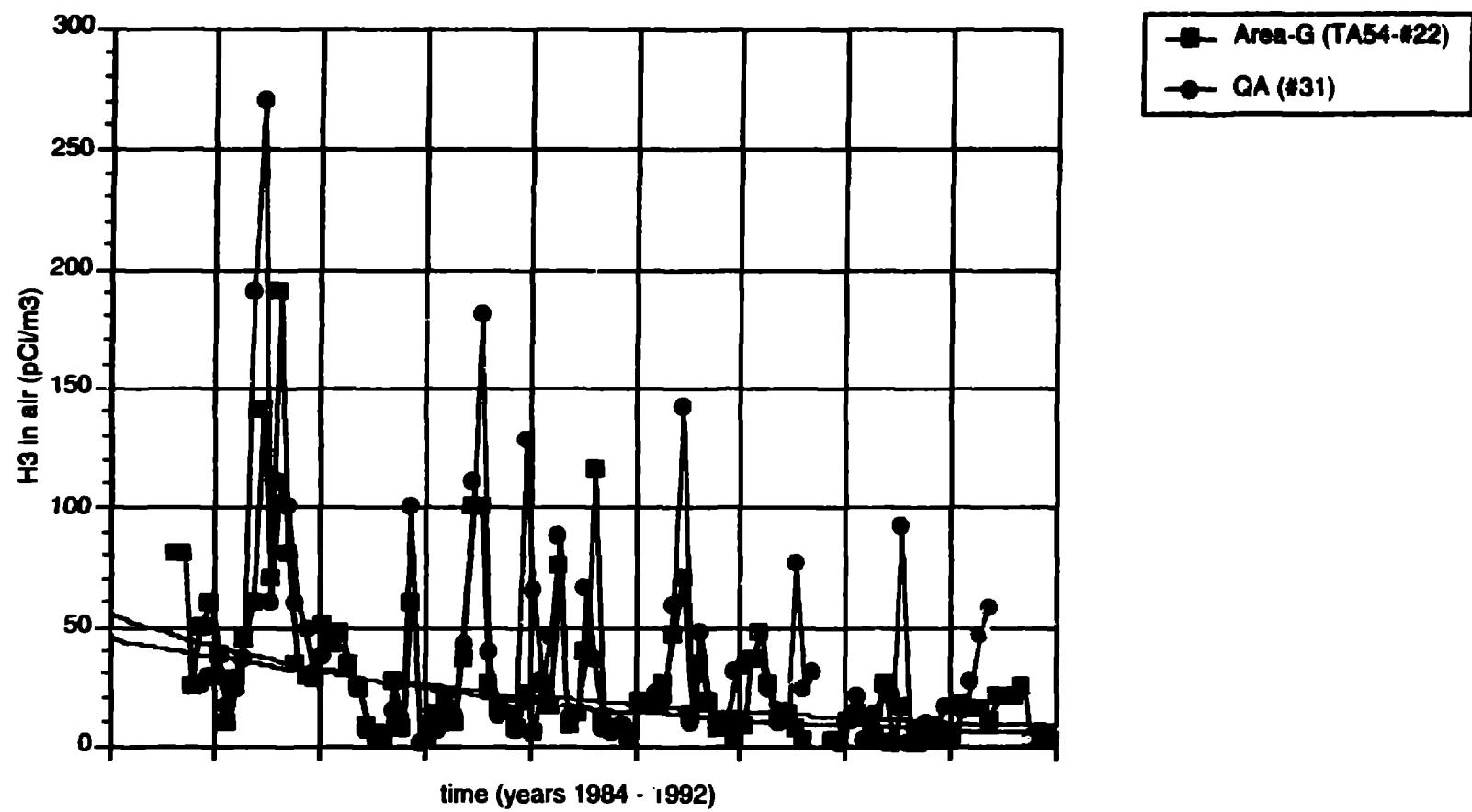


Figure 1. Tritium air concentration data for two stations at Area G for 1984 - 1992.

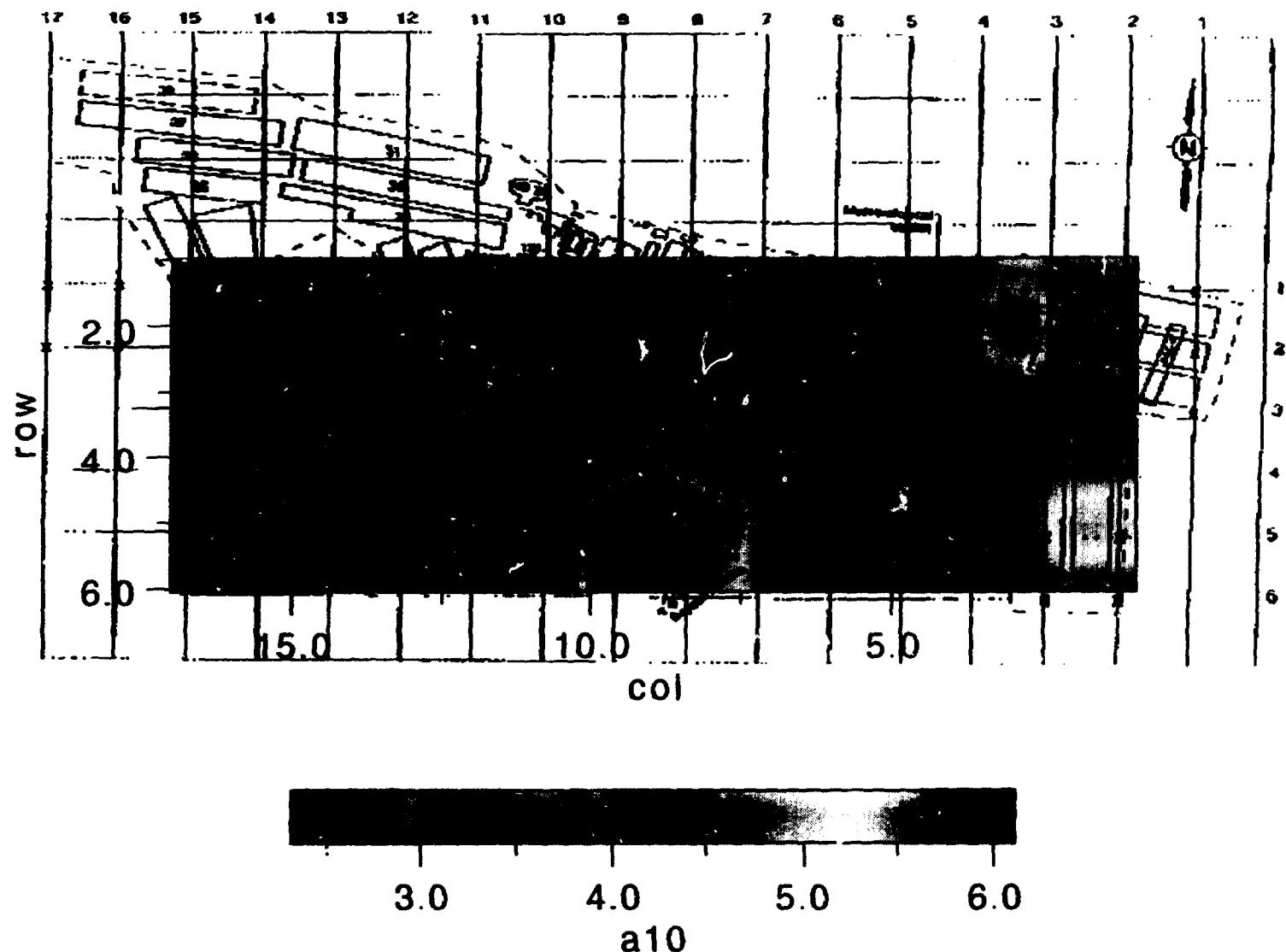


Figure 2. Tritium in near-surface soil moisture, 1989 survey. The scale a_{10} indicates \log_{10} exponent, or tritium concentration in units of ($10^{a_{10}}$ pCi/liter).

Area G Flux Study

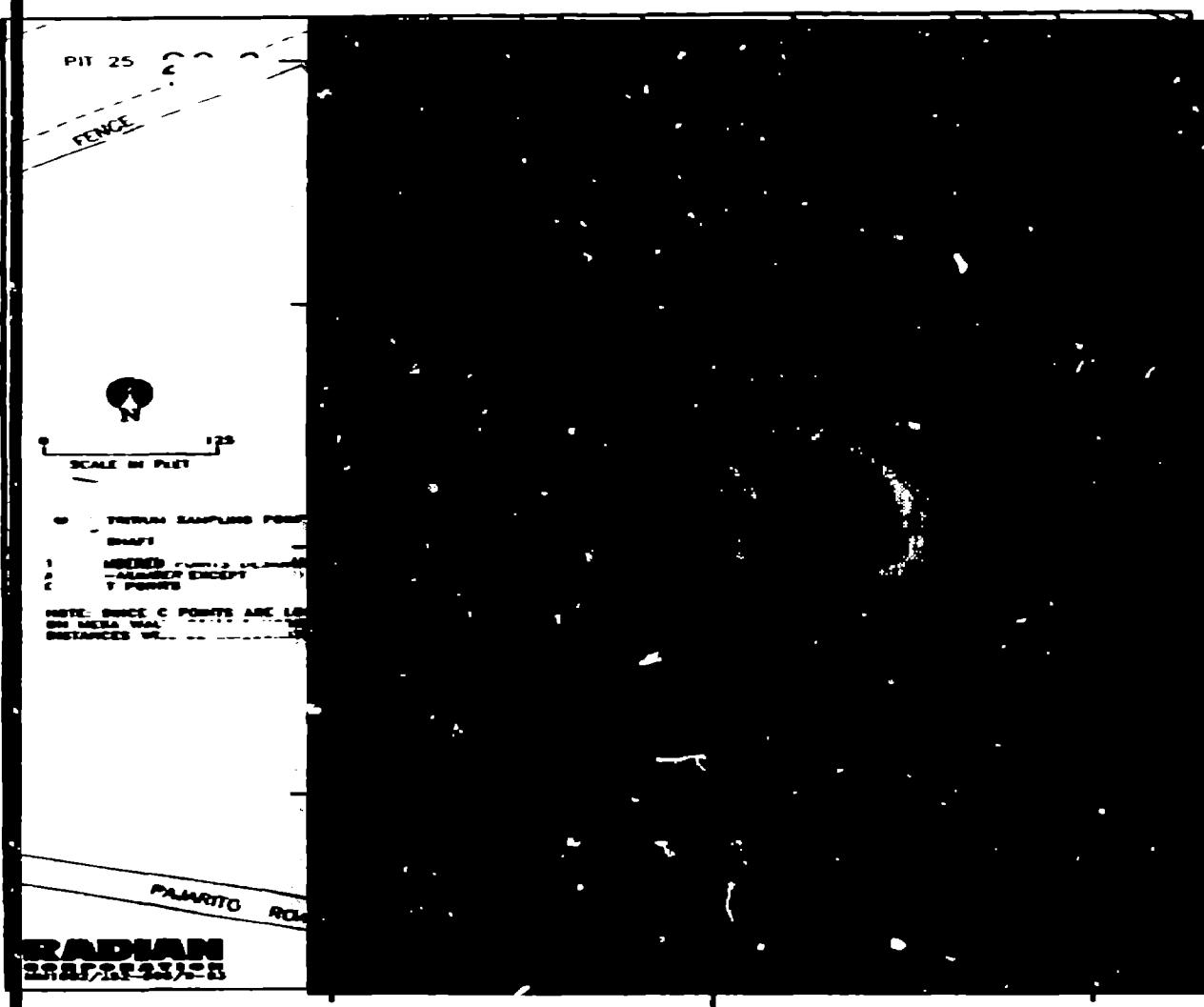


Figure 3. RADIANT results for tritium soil surface efflux

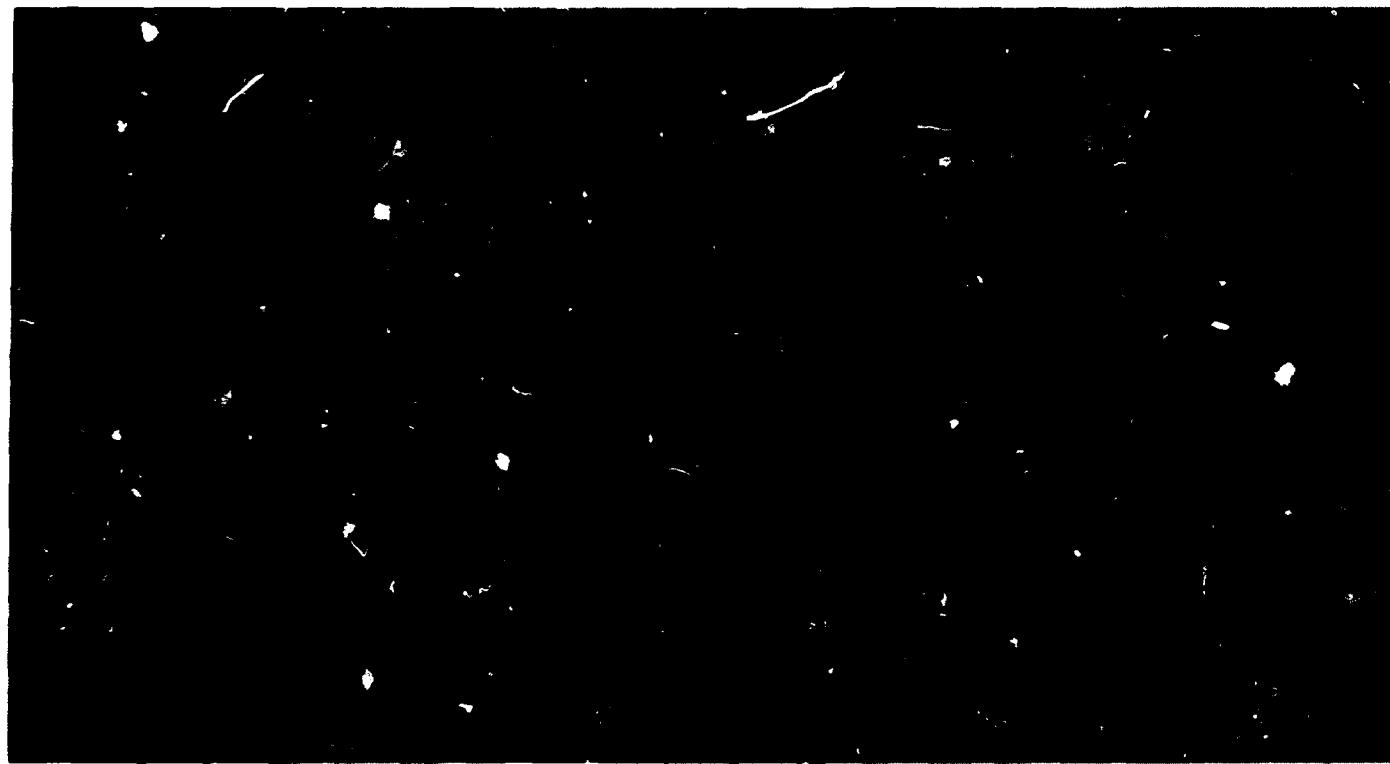


Figure 4. Model results for tritium concentrations in vicinity of tritium disposal shafts. Left figure shows the isocontours on the surface of the entire region with the upper surface corresponding to the soil surface as shown in Fig.3. The smaller figures to the right show relatively higher isocontour levels, indicating the three shafts with the greatest inventory and the two of those shafts contributing to the highest levels of tritium.