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EMANATION AND DISPERSAL OF TRITIATED WATER FROM DISPOSAL SHAFTS[a]

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ABSTRACT. The source level of contamination induced by the presence of tritiated water (HTO) in the Bandelier tuff near Los Alamos, New Mexico has been seen to decrease in all directions at the same rate. This decrease in radioactivity with distance from the source has been measured around three different disposal shafts and found to be slower than the decrease in emanation rate with distance from the source.

Physical factors, suspected of influencing HTO emanation, were entered as independent variables in a regression equation including measurements taken over a 14-month period. The physical variables studied were of thermal, hydrological, and meteorological origin. Only four variables were retained as significant.

Air concentrations of HTO show a maximum during the warmer months and an overall minimum during the colder months. Emanation of HTO, shown to be strongly associated with daily heat flux amplitude, also at a maximum in summer and at noon, more than counterbalances higher dissipation due to increased turbulence occurring during the same period.

More than 76% of the variation in air concentration of HTO was associated with the regression on emanation, wind direction, wind velocity and turbulence.

INTRODUCTION

Determining the potential impact of buried radioactive wastes on the environment requires knowledge of the rates at which the radionuclides can enter the environment. Tritiated water (HTO) emanation is subject to the influence of pertinent meteorological, thermal and pedological variables. Air concentrations of HTO will in turn be influenced by atmospherically induced dissipation which is highly influenced by turbulence, windspeed and wind direction.

MATERIALS AND METHODS

This paper discusses, among other things, sampling and analysis of tritiated water in soils, the spatial distribution of tritiated water emanation and the physical factors influencing that emanation from contaminated soils, the physical factors influencing dispersion of triated water in the atmosphere and finally the atmospheric concentration of tritiated water in the atmosphere.

Soils suspected to be contaminated are collected at increasing distances around a potential source of tritium. The water is then distilled from 750 gram samples from each location. In the case of tritiated water emanation or tritiated water vapor in the atmosphere, the tritiated water is collected by a silica gel trap either through evaporation from the soil (emanation) or forced passage (for atmospheric concentration measurement). Tritiated water is then distilled from the silica gel. In each case, 10 ml of water are tentatively collected and added to 15 ml of a scintillation cocktail before submission for liquid scintillation counting.

TRITIATED WATER IN SOILS

Radionuclides disposed of at the Los Alamos National Laboratory include significant amounts of transuranic elements (mostly plutonium and americium), uranium, fission products, activation products, and tritium. Studies of tritiated water in soils were initiated by Purtymun (1) and Wheeler and Warren (2). All investigations of the radioactive waste disposal areas revealed that, with the exception of tritium, practically no migration of these radionuclides has been detectable. Because of the

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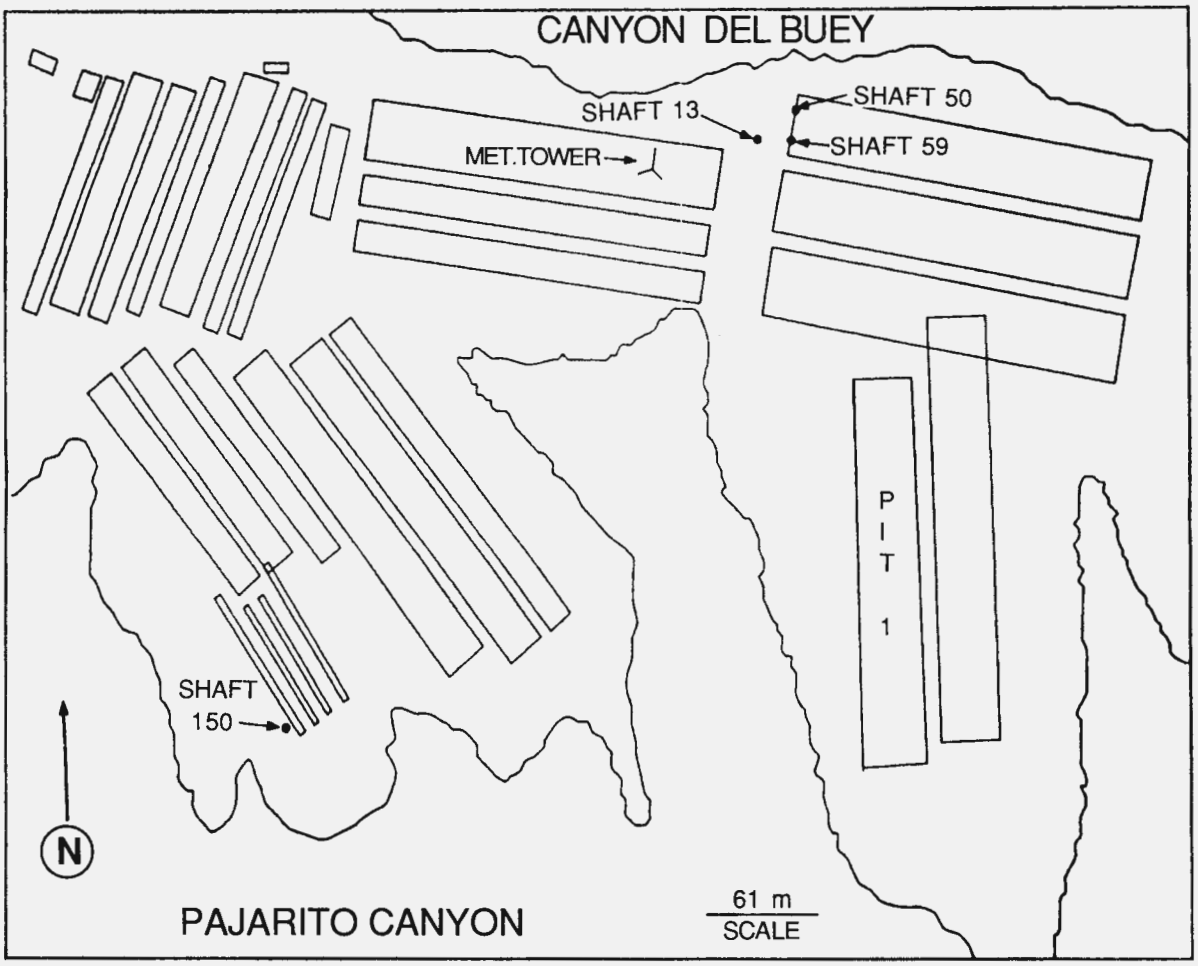

FIGURE 1. Study area at Waste Disposal Area G in Los Alamos.

high mobility of tritium in biological systems, there is a legitimate need to define and minimize the quantities of this radionuclide entering the environment from disposal areas (2). This study shows that migration of tritium from a disposal site was detected as early as 1970, although dissolution and subsequent migration of solid radionuclides in the disposal area is minimal. This has been verified continually by analysis of innumerable samples collected during the excavation of additional shafts in the disposal areas. Tritium, on the other hand, occurring principally as tritiated water, is nearly as free to move as water itself. Small quantities of tritium gas, disposed of by burial, convert very quickly to HTO on exposure to soil oxygen.

It is shown that around an unlined shaft (number 13, Area G, Technical Area 54 on Mesita del Buey shown in Fig. 1), tritium concentrations in the surrounding tuff increased to a maximum between depths of 3 to 10 m and then decreased again with depth (1). Irregularities in the isotritium contours are seemingly influenced by the presence of open joints in the tuff, which provide for a much more rapid means of migration in the vapor phase (Fig. 2). Close but irregular isotritium contours or high tritium gradients are to be found between the source (shaft) and an eventual transversal joint. The contours are elongated to the west of the shaft, indicating regions where either a radial open joint was present or the outward (upward) movement of tritium was inhibited. High tritium gradients would consequently be an indication of swift, uninhibited aeration of tritium. A close and complete set of isotritium contour lines, has been chosen along a southwest axis. The presence of tritium radioactivity decreases logarithmically with distance from the shaft. Figure 3 indicates the rate of decrease at various depths of radioactivity as a function of distance from the shaft. The radioactivity is measured in Becquerels per cubic meter of water and is determined by liquid scintillation counting.

We performed an analysis of the data from Purtymun (1) which showed that a vertical plan along the shaft axis had a maximum tritium concentration near the bottom of the shaft with the radioactivity (A) decreasing upward according to $A = 1.11 \cdot 10^{10} - 6.10^{9} \ln d$ and $r = 0.97$, where d is upward distance from the bottom of the shaft. The similarity between

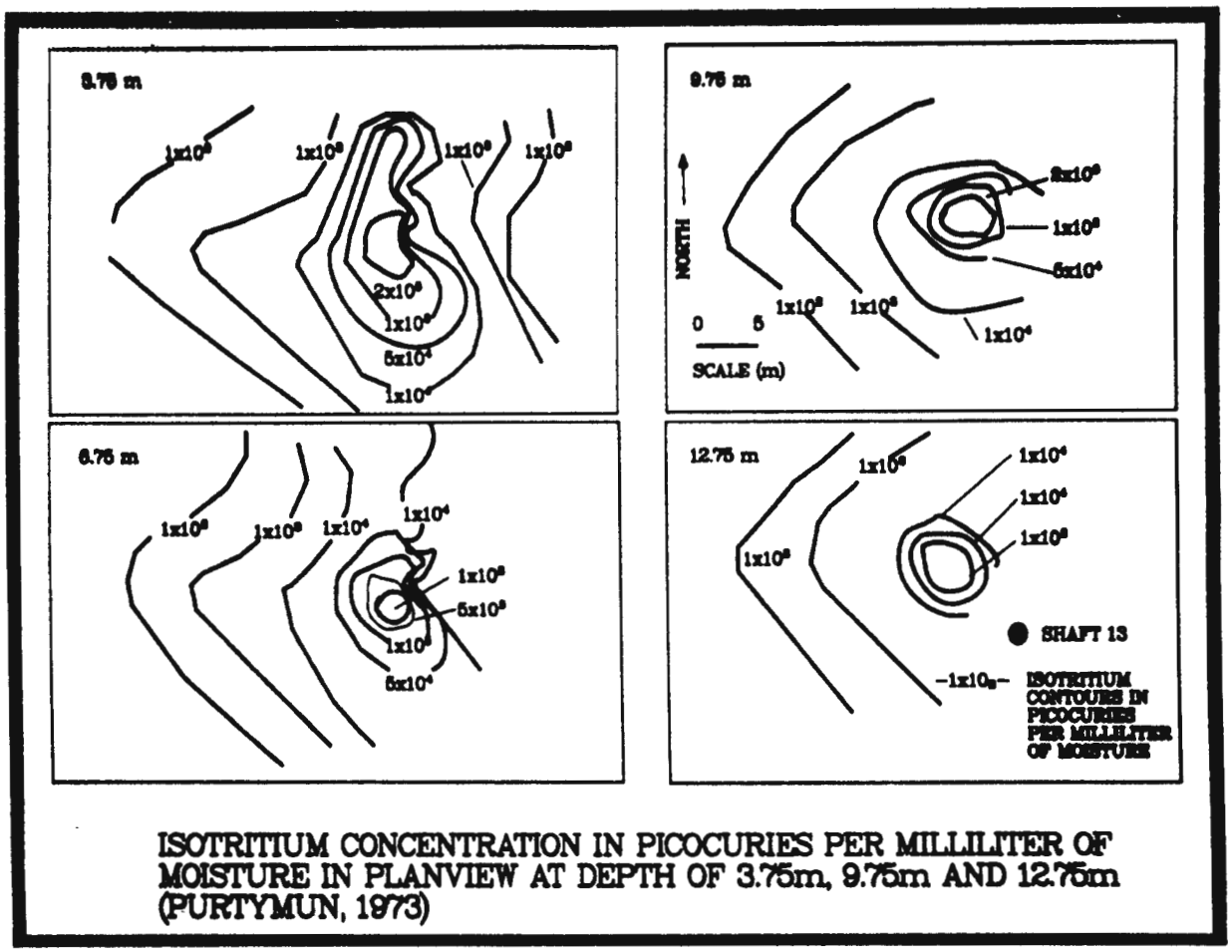

FIGURE 2. Isotritium concentration in picucuries per milliliter of moisture in plainview at depths of 3.75 m, 9.75 m and 12.75 m (1).

the rate of decrease in radioactivity as a function of distance is striking along the vertical and horizontal planes. (Compare with Table 1.) The closest match is between the horizontal rate of decrease as a function of distance from the shaft at 6.75-m depth and the vertical rate of decrease upward as a function of distance from the bottom of the shaft. Figure 3 shows that at equidistance from the bottom of the shaft (3.75 m and 9.75 m depth), the rates of decrease are very similar.

The vertical rate of decrease is 14% faster than the horizontal rate of decrease away from the bottom of the shaft. This is to be expected if the medium is isotropic. However, normally the medium is anisotropic with a higher horizontal diffusivity due to stratification. The higher vertical rate of decrease is due to a stronger gradient pull brought about by the free atmosphere at the horizontal interface.

Shafts 50 and 59, also located in Area G, Technical Area 54 on the Mesita del Buey at Los Alamos, NM, have received tritium contaminated waste since 1973 (Fig. 1). Approximately the same time period elapsed between the placement of the waste and the collection of samples in shafts 50 and 59 as in shaft 13. Both shafts 50 and 59 were asphalt lined and asphalt containment was applied to the packaging of wastes placed in the shafts. Yet, on a percentage basis, significantly more tritium has migrated out of shafts where asphalt containment was used than from shafts where no asphalt was applied. Undoubtedly, many factors contribute to this difference, including different waste forms and packaging before asphalting, variabilities in permeability of the tuff, and differences in the size and number of fractures intersected by the various shafts. However, the asphalting was intended to counteract and override these variabilities. It is concluded that the asphalting techniques did not significantly reduce the migration of tritium away from the disposal shafts. The shafts also appear to serve as a moisture source to the tuff, particularly at the bottom of the shaft (2). This is probably, as the authors state, because of the entrance of precipitation into the shafts during and between waste deliveries and before they are sealed. The asphalted bottom serves as an inhibitor to the flow of moisture out of the shaft, and this moisture in turn increases the total amount of water vapor to be diffused out of the shaft after its closure. Asphalt lining of the bot-

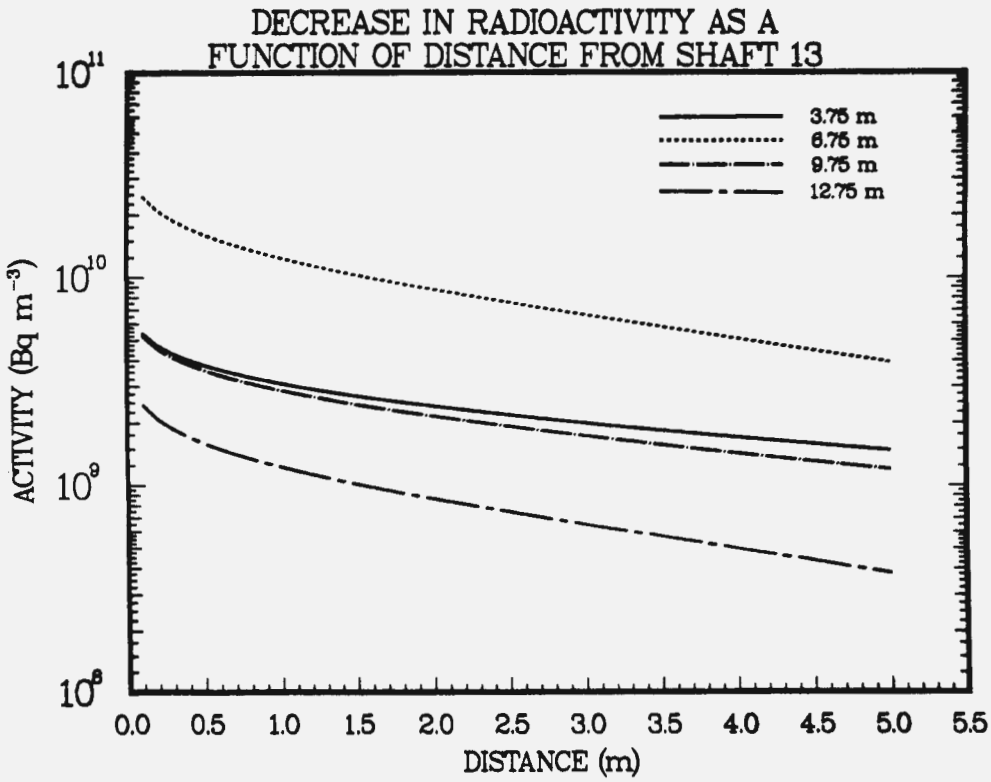

FIGURE 3. Decrease in radioactivity as a function of distance from shaft 13.

tom of the shaft may also induce a higher horizontal vapor flow instead of a downward liquid flow, thereby causing higher horizontal contamination levels around the asphalted shafts.

In this case also, tritium activity decreases logarithmically with distance from the shaft. Figure 4 indicates, at two different depths, the rate of decrease of radioactivity as a function of distance from shafts 50 and 59. The fastest rate of decrease away from shaft 50 is along a SSW axis, and from shaft 59, the fastest rate of decrease is along the eastern axis. However, the tritium data are incomplete along the eastern axis, so that the next steepest gradient, along the northern axis, was chosen for analysis. For the tuff surrounding shafts 50 and 59, the radioactivity was measured using as basis a unit volume of tuff instead of water, as was used for the analyses surrounding shaft 13. Therefore, the constants in the regression equation, expressing radioactivity as a function of distance from the shaft should be substantially higher for shaft 13 for equal degrees of contamination. Because this is not the case, it can be concluded that tritium contamination is substantially higher around shafts 50 and 59. The possible reason for this has been described in the preceeding paragraph. The rates of decrease as a function of distance from the shafts are strikingly similar in both cases and higher activities again prevail near the bottom of all shafts.

EMANATION OF TRITIATED WATER — SPATIAL DISTRIBUTION

Burial Pit 1

Soil borings in the cover material overlying the waste from Burial Pit 1, Area G, Los Alamos (Fig. 1) showed tritium concentrations increasing from $7.4 \cdot 10^7$Bq m^{-3} of soil moisture at 15 cm to $4.6 \cdot 10^9$ Bq m^{-3} at 3.5 m. This gradient suggested an upward diffusion of tritiated water from the waste (at a depth below 3.5 m) to the surface soils. This tritiated water mixes with meteoritic soil moisture and is evaporated. Experiments were designed to quantify the tritium flux as a function of the distance from the source. Amounts of evaporated soil moisture were measured on silica gel collectors. Collection was at night, or on cloudy days, to reduce thermally-induced lateral moisture fluxes. The average moisture flux and tritium flux were determined for the collection period. Measured tritiated water fluxes varied from $2 \cdot 10^{-3}$ Bq m^{-2} s^{-1} to $2 \cdot 10^{-1}$ Bq m^{-2} s^{-1}, with a minimum in winter and a maximum in summer. The actual surface area contributing to this flux was not established, but measurements of the tritium distribution in the soil suggest that only a portion of this particular pit is contributing. Pit dimensions are approximately 30 m by 200 m ($6 \cdot 10^3$ m^2). If the entire pit were a source at the observed rate, atmospheric releases would

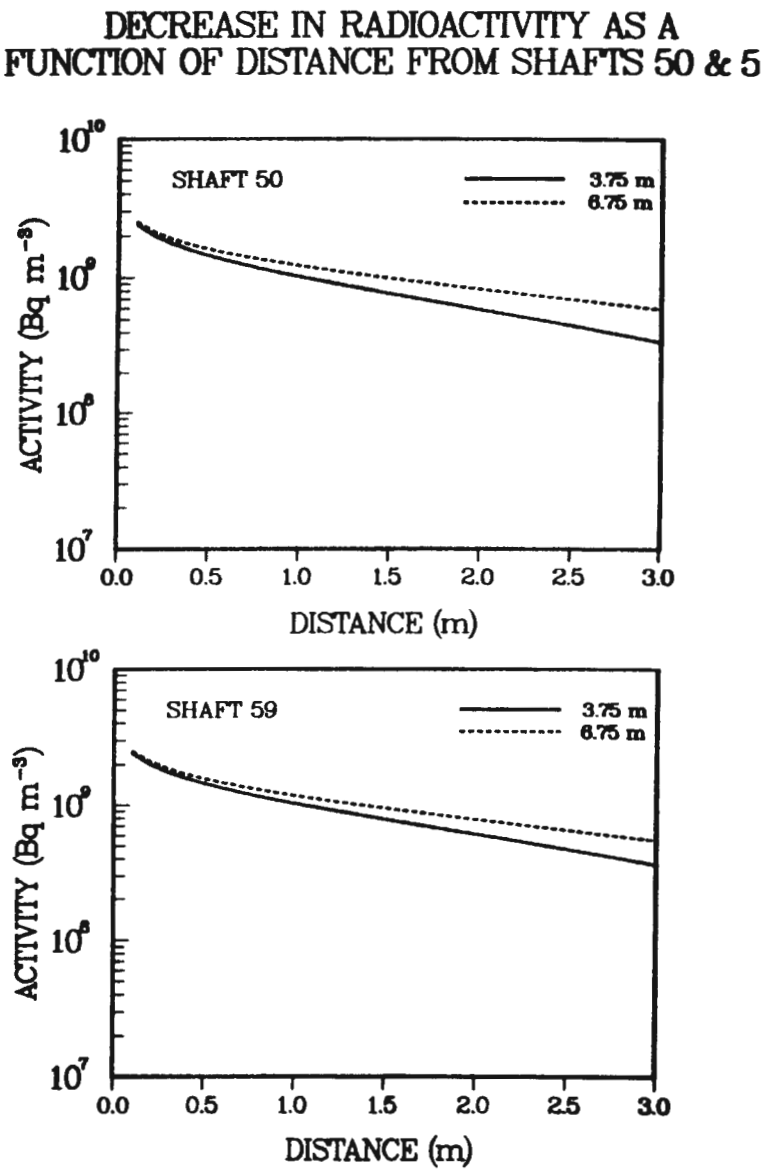

FIGURE 4. Decrease in radioactivity as a function of distance from shafts 50 and 59.

range from 13 Bq s^{-1} to $1.1 \cdot 10^3$ Bq s^{-1} during the measurement period. Because only some fraction of the total pit is actually contributing to tritium releases, the actual tritium released is less than this amount.

Shaft GS 150

A new disposal shaft, No. 150, located in an area where no prior tritium disposals had occurred, was used for further investigation (Fig. 1). The extent of tritium migration into the tuff surrounding the shaft could then be related directly to the contents of the shaft. Disposal shaft 150 was drilled in the spring of 1976, with the first disposal occurring on 12 May 1976. In the winter of 1977, after more than $1 \cdot 10^{15}$ Bq of tritium had been disposed to the shaft, nine sampling holes were processed to remove the contained water, and the water was analyzed for tritium. The sampling holes were provided with surface casings and then sealed. Subsequently, in the fall of 1979, samples of the water vapor in the access holes were collected using silica gel as an absorbing medium. Water was distilled from the gel and analyzed for tritium. Also, the moisture content of the tuff surrounding the access holes was determined using a neutron moisture probe. The average water content in the tuff surrounding the shaft was about 4% by volume. This value agrees well with other measurements made in the area (3). Moisture distribution in Bandelier Tuff is presented in Fig. 5. Nakayama and Jackson (4) report that the apparent diffusion coefficient for tritiated water in soils increased rapidly below a volumetric water content of 10 percent with a maximum at 4 percent water con-

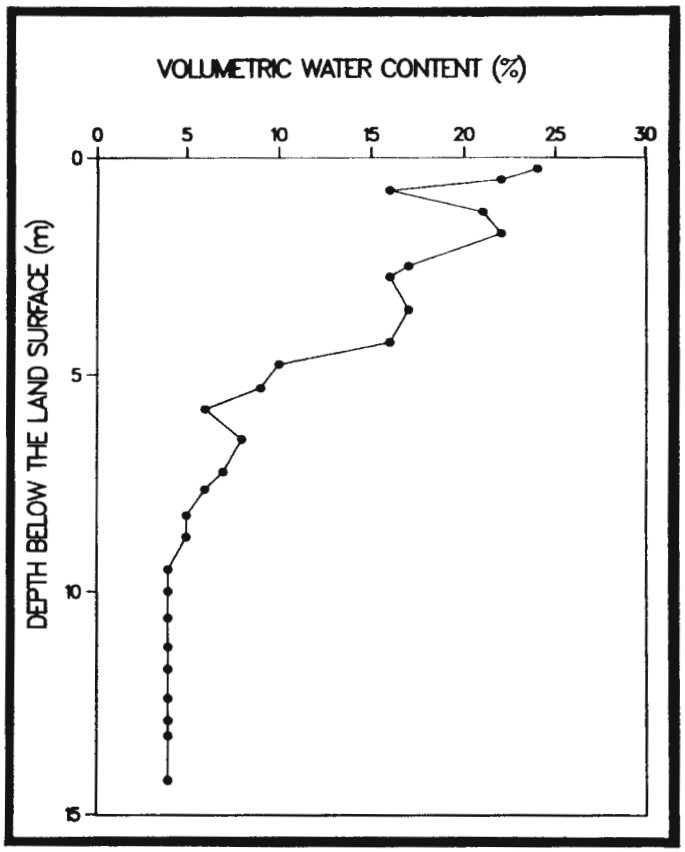

FIGURE 5. Moisture distributions in Bandelier Tuff as a function of depth.

tent, beyond which the diffusion coefficient decreased rapidly. As can be seen in Fig. 5, the "equilibrium" moisture content in the tuff tends to level off around 4 percent by volume.

Emanation of tritiated water was measured in December 1979 along three axes, A, B, C, 120° apart, around GS 150. The measured average nightly concentration read $6.1 \cdot 10^8$ Bq m^{-3} of soil water for an area including 113.10 m^2. Because the nightly evaporation rate was averaging $7.75 \cdot 10^{-9}$ m s^{-1}, the total nightly release of tritiated water was $6.1 \cdot 10^8$ Bq $m^{-3} \times 7.75 \cdot 10^{-9}$ m $s^{-1} = 4.75$ Bq m^{-2} s^{-1}, or within the area under consideration (6 m radius), 535 Bq s^{-1}. Along each of the three axes, the rate of tritium emanation drops off with distance from the shaft according to a power function.

With d, the distance, expressed in meters, and E, the tritiated water emanation flux, expressed in Bq m^{-2} s^{-1}, the regression analysis for the three different axes yields:

A-axis: $E = 95.5\ d^{-4.7546}$ with $r = 0.9902$
B-axis: $E = 104\ d^{-4.4050}$ with $r = 0.9972$
C-axis: $E = 133\ d^{-5.0313}$ with $r = 0.9881$

The average regression analysis for the three axes yields

$$E = 110\ d^{-4.7000} \text{ with } r = 0.9932$$

This is an indication of an extremely fast decrease (Fig. 6) of tritium release with distance from the source (GS-150).

By the end of December 1979, along the B axis originating from GS-150, the regression formula $E = 104\ d^{-4.405}$ then obtained, yielded 0.11 Bq m^{-2} s^{-1} at 4.7 m from the shaft. This result seems to be quite compatible with actual measurements made by the end of December of the following year, when at 4.7 m from the shaft, the tritiated water emanation flux was 0.14 Bq m^{-2} s^{-1}.

High release rates during the summer are expected because the observations follow a seasonal pattern. Atmospheric concentrations are only partly influenced by soil releases (wind speed, and atmospheric convection will also play an important role). Nonetheless, air concentrations also show a maximum during the warmer months. At 4.7 m from the shaft center (GS-150), the nightly water flux averages $7.3 \cdot 10^{-9}$ m s^{-1}, with an average HTO concentration of $2.6 \cdot 10^8$ Bq m^{-3}, resulting in an average nightly tritiated water flux of 1.84 Bq m^{-2} s^{-1}. HTO air concentrations are influenced by both emanation rates and atmospheric movements, vertical and horizontal. The last factors are best esti-

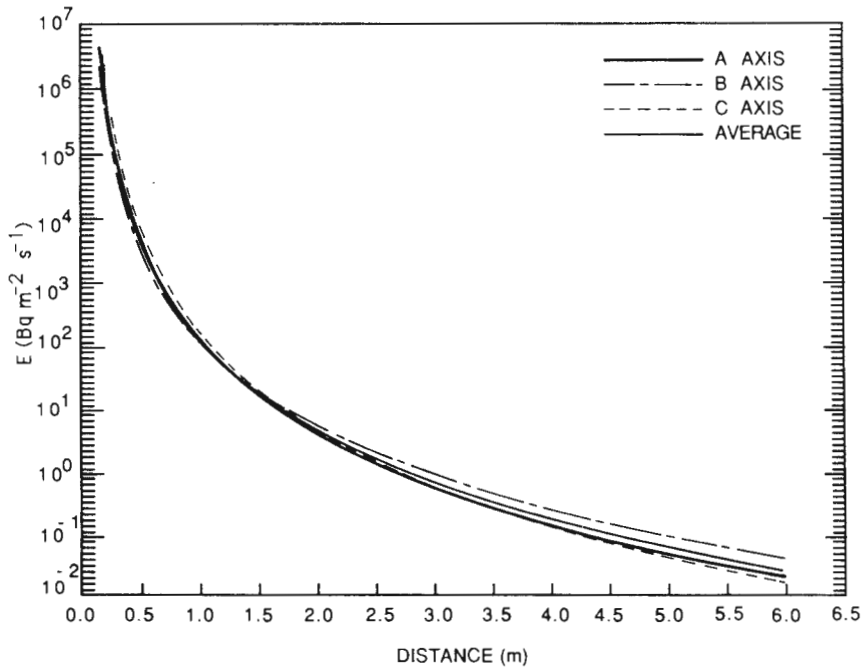

FIGURE 6. Regression analysis of tritium emanation fluxes for the three axes in shaft GS 150.

mated from the wind velocity and its increases in velocity with height. The specific activity of the emanated tritiated water, averaged over a 13-month period (n = 52), equals $2.75 \cdot 10^8$ Bq m^{-3} with a large standard deviation that follows a seasonal pattern. The maximum specific activity of tritiated water reached $1.088 \cdot 10^9$ Bq m^{-3} in the summer (September 3), while the minimum specific activity of tritiated water dropped to $1.7 \cdot 10^7$ in the winter, four months later (December 30). We thus see a large variation in the specific activity of the emanated tritiated water (close to two orders of magnitude), an indication that the emanation of tritiated water is more strongly subjected to seasonal variation (high emanation in summer) than is evaporation of meteoritic water, leading to a higher specific activity during the summer.

PHYSICAL FACTORS INFLUENCING EMANATION

As mentioned previously, disposal shaft, No. 150, was located in an area where no prior tritium disposals have occurred.

It has been suggested that the thermal characteristics of a medium and of the adjoining boundary layer play a role in the exhalation of the components of the vapor phase (5,6).

The termal diffusivity (D) of the tuff at GS-150 was derived in situ from the measurements of daily temperature fluctuations (ΔT) at different depths (z) using

$$D = -\left(\frac{\Delta Z}{\ln \Delta T_1/\Delta T_2}\right)^2 \frac{\pi}{864OOs}$$

The values for D averaged $8 \cdot 10^{-7}$ m^2 s^{-1} during an extremely dry summer period, which kept the soil moisture content low, resulting in a low specific heat by volume and a higher thermal diffusivity. Representative tuff debris was taken to the laboratory, where the bulk density was accurately determined to be $1.395 \cdot 10^3$ kg m^{-3}.

The specific heat (C_s) of the tuff was measured in the laboratory using the standard method. The test was repeated four times and averaged C_s dry = 866J kg^{-1} K^{-1} or a heat capacity of $C_v = 1.208 \cdot 10^6$ J m^{-3} K^{-1}. Because the moisture ratio by volume in situ turned out to be 0.04, the actual heat capacity of the tuff under consideration is $C_v 0.04 = 1.375 \cdot 10^6$ J m^{-3} K^{-1}, with a standard deviation of $2.8 \cdot 10^4$ J m^{-3} K^{-1}. The thermal conductivity, γ, was computed to be $D \cdot C_v 0.04 = 1.1$ W m^{-1} K^{-1}.

The thermal characteristics will help explain the magnitudes of the temperature and consequent vapor pressure gradients in the tuff. These will be among a series of physical variables that will be studied as potential influences on tritiated water vapor emanation. For that purpose, recording thermometers and heat flow sensors were emplaced at depths of 0.10, 0.35, and 1.00 m. Fourteen meteorological and pedological variables or properties were measured or computed over a period of 14 months. After preliminary screening, eight variables were retained for further analysis. Multiple linear regression analysis identified four independent variables as having a significant influence on HTO vapor emanation. This criterion was based on the value of lowest squared error given by Mallows (7). More than 71% of the variation in HTO flux was associated with its regression on

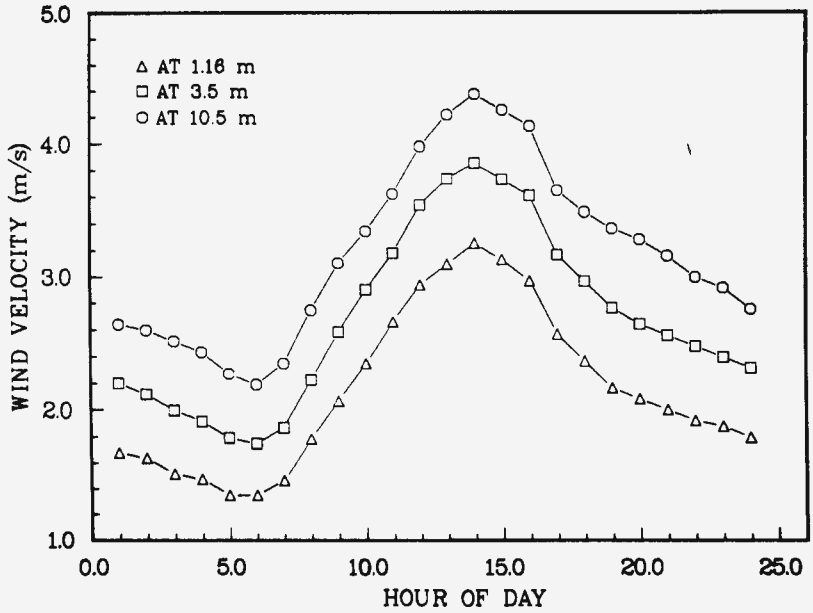

FIGURE 7. Wind velocity at Los Alamos.

- daily heat flux amplitude (in W m^{-2}).
- meteoritic water exhalation E or evaporation (in m s^{-1}),
- maximum soil vapor pressure gradient dP/dx (in Pa m^{-1}), and
- atmospheric pressure variation Δp (in Pa).

The independent variables are listed above in decreasing order of significance.

Because the *t*-test for Δp does not exceed the tabular value at the 0.05 level for the degrees of freedom under consideration, the hypothesis that the coefficient of Δp in the regression equation is equal to zero cannot be rejected at the 95% confidence level. This finding only adds to the controversy involving the influence of atmospheric pressure changes on gaseous emanation from the ground. Some authors were unable to detect any direct influence of the variations in atmospheric pressure on emanation rate (6), whereas others suggest its effect is significant (8).

The influence of the heat flux amplitude has been implied by Styra *et al.* (5) when the maximum rate of exhalation is linked to convective streams in the soil, generated by intensive surface cooling. Emanations have also been found to be a function of seasonal variation, with little variation and low emanation occurring during the winter months (6). It is worth noticing that heat flux amplitude remains extremely low in absolute value and variation during the winter months. It has been suggested that diurnal surface heating of the soil (which is at the origin of soil heat fluxes) may be responsible for increased transport velocities (9). It is also to be noticed that the daily heat flux amplitude is stabler and lower in winter; the resulting temperature gradient may be conducive to the formation of ice lenses, which will trap HTO and further inhibit its emanation.

The total nightly upward meteoritic water flux (evaporation) amounts to 0.11 m/yr. Because there is very little, if any, water seepage into deeper tuff layers, the actual evapotranspiration very closely matches the annual precipitation average of 0.46 m/yr. This tends to indicate that approximately 75% of the evaporation out of the tuff occurs during the day. Because the coefficient of determination (r^2) between HTO flux and H_2O flux is 0.59, there is reason to believe that the HTO flux is likely to be about three times higher during the daytime.

PHYSICAL FACTORS INFLUENCING DISPERSION

Wind and turbulence are strong pollution dispersers. The vertical wind profile is of great interest for the studies of turbulent mass exchange, energy, and water budget of the atmosphere, as well as for air pollution studies. Because turbulence has a vertical component, it will inhibit differential windspeed patterns.

The simplest way to express the relationship between wind velocity and height is by a power law of the form

$$U_2 = U_1 \left(\frac{Z_2}{Z_1} \right)^a$$

where U, is the wind velocity at height Z.

Consequently, if a log-log graph is used, the plot of wind velocity against height will be a straight

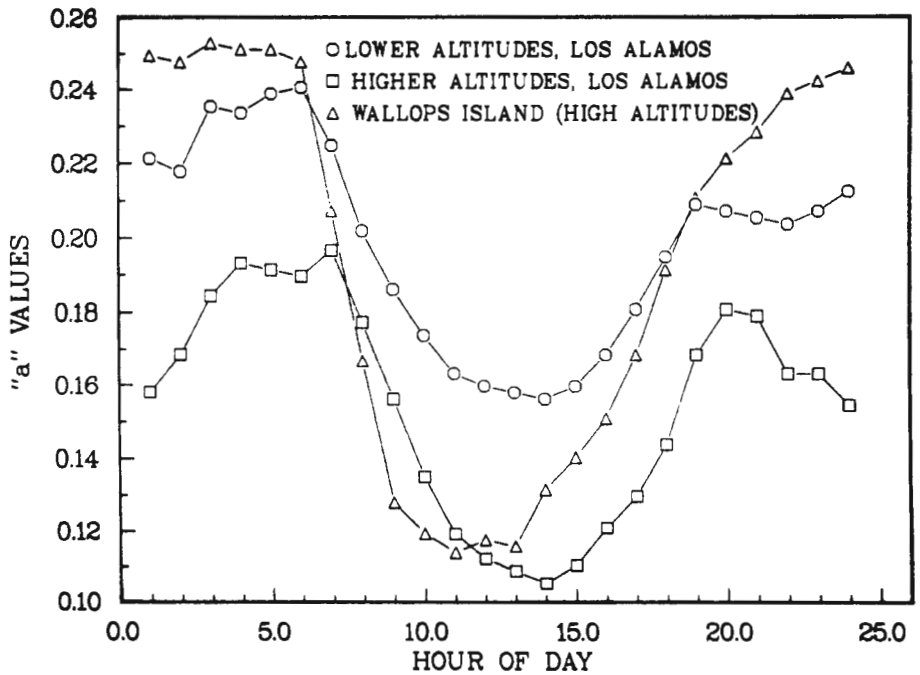

FIGURE 8. Wind profiles at Los Alamos.

line. A low value of "a" will generally be indicative of a strong turbulence.

Wind Velocity

Wind velocity data were collected by the Environmental Surveillance Group at the Los Alamos National Laboratory. The readings were collected during 1981 and 1982 at Area G (Fig. 1). The heights chosen followed a geometric progression: wind velocitities were measured at 1.16 m, 3.5 m, and 10.5 m above the ground surface. (See Fig. 7.)

Turbulence

Turbulence follows a daily sequence with well-marked maxima about noon and minima by night. Because of different rates of change of wind velocity with altitude, a different wind "profile" can be expected throughout the day. Plotted against hour of the day, the values for "a" go through a minimum during the time of the day that radiation is most intense, and maximum values are reached during the early morning hours. This is demonstrated in Fig. 8.

The strongest turbulence as indicated in Fig. 7 seems to be present between 11:00 and 16:00 hours, while windspeed attains its maximum during the same time period. Monthly distributions are not as well established. We used T = 1/a as an indication of turbulence.

Patterns for wind velocity and turbulence are much more obvious on a diurnal basis than on an annual basis (month by month). Therefore, it is possible to conclude that if atmospheric concentrations of HTO could have been taken on an hourly basis and the regression analysis run accordingly (instead of on a monthly basis), the dispersion action of wind and turbulence could have shown to be a lot more prominent and the relationship between emanation and atmospheric concentration less prevalent.

ATMOSPHERIC CONCENTRATION

The atmospheric concentrations of HTO, measured during a period ranging from 1980 to 1982 were compared with emanation measurements, wind-

TABLE 1

Decrease in Radioactivity Near Shaft 13 as a Function of Distance from the Shaft Along a S–W Axis

Depth (m)	Rate of Decrease (Bq m^{-3})	Coefficient of Correlation
3.75	$A = 3.1 \cdot 10^9 - 1.01 \cdot 10^9 \ln d$	r = 0.96
6.75	$A = 1.24 \cdot 10^{10} - 5.27 \cdot 10^9 \ln d$	r = 0.97
9.75	$A = 2.87 \cdot 10^9 - 1.04 \cdot 10^9 \ln d$	r = 0.98
12.75	$A = 1.23 \cdot 10^9 - 5.28 \cdot 10^8 \ln d$	r = 0.96

TABLE 2

Monthly Tritium Emanation, Wind Turbulence, Wind Direction, D and Atmospheric Concentration at Los Alamos.

Month	Emanation ($Bq\ m^{-2}$)	Wind (m/s)	Turbulence	Wind Dir (SSE) (%)	$\frac{D}{WT}$	Atm. Conc. ($Bq\ m^{-3}$)
J	0.13	2.9	4.3	1.4	0.11	0.70
F	1.26	2.6	4.5	1.2	0.10	0.11
M	0.67	3.5	5.2	1.6	0.09	0.14
A	1.11	4.3	5.3	1.8	0.08	0.78
M	2.02	3.6	4.9	2.2	0.12	1.18
J	2.65	4.2	4.0	3.8	0.23	1.26
J	2.60	3.1	3.9	5.6	0.46	1.67
A	4.44	2.6	3.9	3.2	0.32	1.63
S	6.24	3.2	6.0	2.0	0.10	0.81
O	1.88	3.1	6.7	2.0	0.10	0.70
N	0.76	2.7	5.3	2.0	0.14	0.70
D	0.27	2.3	5.3	1.8	0.15	0.63

speed and wind direction measurements, and turbulence calculations that were all taken or made within the indicated time interval. A composite factor, integrating wind direction D, wind velocity W and turbulence T was also included in the regression analysis. The wind direction that influenced the correlation most positively was SSE or a strongly upslope wind, most likely to occur during a warm sunny day or when the daily heat flux amplitude and consequently the emanation, were at a maximum. The data are summarized in Table 2.

CONCLUSIONS

The spatial distribution of HTO in the tuff and emanation at the soil-atmosphere interface revealed that HTO release into the atmosphere decreases faster with distance from the source than does HTO dispersion in the tuff. Lateral diffusion in the tuff may occur after a heavy rainstorm, for example, when surface pores are clogged and upward diffusion of HTO is inhibited, thus enhancing lateral distribution and entrapment of HTO. Both distributions indicate fast decrease of activity with distance, however.

More than 71% of the variation in HTO flux was associated with the regression on four independent variables, but the daily heat flux amplitude alone was accountable for 61% of the variation.

More than 76% of the variation in atmospheric HTO concentration was associated with emanation, wind direction, turbulence and wind velocity. It should be noted that the wind direction having the highest positive input in an upslope wind most likely to occur when heat flux amplitude and emanation are at a maximum.

Possible reasons for the atmospheric concentrations not being more closely related to the studied parameters is probably related to the periodicity of the measurements. While measurements and computations for windspeed, wind direction and turbulence were averaged over 15 minute periods, emanation measurements covered a period integrating 15 hours of tritium exhalation, while atmospheric concentrations were covering periods of no less than 15 days. It is possible to conclude that if atmospheric concentrations of HTO and emanation rates could have been taken on an hourly (or better: 15 min.) basis, the correlation between atmospheric concentration and the parameters influencing it would have been much higher.

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