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ENVIRONMENTAL TRANSPORT AND CYCLING OF TRITIUM IN THE VICINITY OF ATMOSPHERIC RELEASES

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C. E. Murphy, Jr., and M. M. Pendergast

E. I. du Pont de Nemours & Co.
Savannah River Laboratory
Aiken, SC 29801

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ENVIRONMENTAL TRANSPORT AND CYCLING OF TRITIUM IN THE VICINITY OF ATMOSPHERIC RELEASES*

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ABSTRACT

Routine releases of tritium to the atmosphere from the Savannah River Plant (SRP) have averaged 5.12×10^5 curies per year during the period 1974-1977.[1] Patterns of tritium dispersion in the environment surrounding SRP indicate a power law relationship between air concentration and distance from the source. The relationship between the concentration of tritiated water in vegetation and distance is more complex. A model of the tritium dispersion and cycling in the environment explains the dispersion patterns in terms of the ratio of tritiated hydrogen to tritiated water, climate, and vegetation characteristics. The annual variation in the environmental patterns around the SRP are almost completely determined by the fraction of tritiated hydrogen in the release.

INTRODUCTION

The Savannah River Plant (SRP) is a major producer of nuclear materials for the U.S. Government. Inevitably, some of these

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materials and their byproducts find their way into the environment around the Plant. The SRP Health Physics Department and the Environmental Sciences Section of the Savannah River Laboratory (SRL) measure the distribution of these materials in the environment and assess the dose-to-man from the operation of the plant. We have collected data to demonstrate the patterns of distribution of tritium in the terrestrial environment. Experiments will be briefly described which have led to a better understanding of tritium movement in the environment. Simulations with a model of tritium movement were compared to the measured tritium distribution.

THE SAVANNAH RIVER PLANT SITE

The production facilities at the SRL include three operating heavy water reactors, a heavy water separations facility, a fuel fabrication facility, a product separations facility and waste disposal facilities. The production areas are located in a 77,831 ha reservation located in South Carolina, east of the Savannah River near Augusta, Georgia, in the southeastern United States.

The reservation is on the western edge of the coastal plain and has soils derived from coastal plain and piedmont parent materials as well as extensive areas of alluvial soils near the Savannah River and sandhill soil away from the river. Much of the upland area that had been farmed before the establishment of SRP are now managed pine plantations. The river swamp and areas next to the streams are in mixed broadleaf forests. The climate of SRP is relatively temperate with mild winters and long, warm summers. The temperature averages 18°C in Augusta and the average relative humidity is 70%. Rainfall is evenly distributed throughout the year and averages 120 cm/a.[2] Dry periods of 8-10 d are encountered almost every year.

Tritium Source Characteristics

Tritium releases to the environment are about equally divided between releases from the heavy water reactors and the tritium separations areas. 90% of the releases to the atmosphere are from 63 m high stacks. The average annual release to the atmosphere during the existence of the Plant is 7.82×10^5 Ci/a. Releases from the separations area average 40% tritium hydrogen gas (HT) and 60% tritiated water (HTO). The releases from the reactor areas are primarily HTO under normal operating conditions. Accidental releases of 4.79×10^5 Ci on May 4, 1974, and 1.82×10^5 Ci on December 31, 1975, were greater than 99% HT.

Patterns of Tritium Distribution in the Environment

The Health Physics Department routinely analyzes precipitation, and moisture in air, vegetation and soils for HTO content at a number of collection locations at the plant boundaries and at radial distances of 40 km and 161 km[3]. In addition, special studies are made to determine HTO in vegetation along transects in four directions from the plant center. Results of the routine measurements of air water and vegetation water are summarized in Table I for 1974-1977.

The air moisture samples are collected continuously on silica gel which are analyzed once a week. The vegetation samples are grab samples made during collection runs. The water is removed from the vegetation by vacuum distillation and counted by liquid scintillation[4].

A plot of the air concentrations against distance from the center of SRP on log-log paper is nearly a straight line. The slope of the line is a little less than the inverse of the distance which would result from radial advection. The vegetation samples are more scattered than the air concentration, and the ratio of air water concentration to vegetation water concentration varies between years. Results from transects of vegetation water taken in the four cardinal directions during June, July and August of 1973 have a logarithmic slope greater than the inverse of distance from the center of SRL. These transects were taken inside the boundary of SRP from distances of 1.6 to 16 km.

Tritium releases over 99% HT in 1974 and 1975 were sampled to help understand tritium distribution in the environment. The environmental samples have already been presented and discussed [5,6] so only a summary of main findings will be presented here.

The highest tritium concentration was found in the soil water (9000 pCi/mL) after the May, 1974 release. The highest vegetation concentration in the vegetation samples was in grass (1600 pCi/mL) and the concentration in pine trees had a maximum of 700 pCi/mL. Measurements made following the release indicated that a band of tritiated water was formed at the soil surface and then displaced downward by rain. The concentration in the trees decreased exponentially with time with half lives of 2.45 and 22.7 days. The high concentration in the soil, compared to the vegetation, suggests that HT may be converted to HTO in the soil. Conversion of HT to HTO at a slower rate in the vegetation is also possible; however, the concentrations in the vegetation can be explained by the vegetation coming into equilibrium with the small percentage of HTO in the release.

Another example of the spatial distribution of HTO is the concentration found in the needles, branch wood, stem and soil near the tritium separations facility (Table II). Sampling was made possible by pulpwood cutting in the area. These samples are likely to be characteristic of the terrestrial ecosystems under normal SRP operating conditions. The highest concentrations are found in the needles and the soil surface and are the likely points of entry from the atmosphere by diffusion. The lower concentrations in the deeper soil could be the result of water of lower concentration entering during rainfall. Rain gauge data indicate that tritium concentration in rain is usually lower than the weekly average concentration in the air. The concentration in the stem and branches should be a weighted average of the concentration in the soil. Since most of the tree roots are near the surface, the stem concentration should be nearer the surface concentration than the deeper soil concentration.

Tritium Uptake and Cycling Experiments

The patterns of tritium distribution observed in the vicinity of SRP suggest which experiments are necessary to understand how the patterns developed. The major mode of entry into terrestial ecosystems seems to be from the atmosphere into the vegetation leaves and into the surface soil. The forms of tritium which are important are HT and HTO.

A number of investigations of the uptake of HTO by plant leaves have been conducted[7,8]. The experiments all show that HTO enters and leaves the plant by the same paths as water. However, the gradient for HTO diffusion is from the atmosphere to the leaf water and thus, HTO may diffuse at a rate very different from the diffusion of water vapor in the same leaf[9]. HT uptake by leaves has not been studied as much as HTO exchange[10]. Therefore, we conducted experiments to determine the rate of uptake of HT by pine needles. Groups of pine needles were exposed for one minute to different concentrations of HT[11]. The needles were slipped into a glass test tube while still attached to the tree. The base of the needles were sealed with a split test tube stopper and air was circulated through the tube at a rate of two test tube volumes a second. HT was added to the entrance air stream.

While there is a good deal of scatter, it appears that tritium uptake is rate limited at the higher concentrations (Figure 1). If we calculate diffusion resistance (assuming a zero internal HT concentration) we find an internal leaf resistance of 2.0 s/cm at the lower concentrations which gets larger as concentration increases. The low internal resistance at lower concentrations suggests an efficient internal uptake mechanism which is saturated at higher concentrations. Because of the low solubility of HT the uptake rate is not high compared to HTO even at the low concentrations.

HTO uptake by the soil is likely to be governed by the same principles as water exchange. However, the high values of HTO concentration found in the soil after releases which were largely HT indicated that the soil was a significant sink for HT. This has since been proven[12,13,14]. The action of micro-organisms has been implicated in the conversion of HT to HTO in soils. More work is necessary to make estimates of the conversion rate over a variety of conditions, but our calculations suggest a tritium deposition velocity (flux/concentration) of .4 cm/s which is equally divided between soil and vegetation uptake[15].

An understanding of tritium cycling in terrestrial ecosystems also requires an understanding of tritium movement in soils and plants. Tritium spiking of soil water has demonstrated that HTO movement in soils is dominated by mass flow with some hydrodynamic dispersion[16,17]. Spiking experiments in trees also indicate that HTO moves up tree stems by mass flow[18,19]. Dispersion in tree stems is greater than in most soils, because of the large amounts of exchangeable hydrogen in the tree stems.

The results of these experiments help explain the tritium distribution patterns we see in the environment of SRP. However, they do not prove that the distribution patterns result from exactly these processes. A further, although not conclusive, test is to formulate the processes as a mathematical model and see if the model can simulate some of the patterns observed.

A Steady-State Model of Tritium Dispersion and Cycling

Model Structure and Implementation

A mathematical model of tritium dispersion and cycling must contain mathematical statements which express the processes of 1) atmospheric dispersion of HT and HTO, 2) wet and dry deposition (uptake) of HT and HTO by vegetation and soil, 3) movement of HTO in soil, and 4) HTO absorption by vegetation roots from soil and flow through the vegetation. A steady-state model has been developed in this case because it seemed appropriate for comparing averages of environmental samples taken over a year's time. Transient models have been developed to examine conditions after large accidental releases[20,21]. However, transient models require environmental data at sampling rates of the duration of the release (15 min in the releases mentioned above).

Annual wind direction is nearly equally distributed at SRP, although seasonal trends do exist. The annual dispersion by wind is nearly radial and the concentration should vary as the inverse of the radial distance from the plant. The expression used in our model is:

$$X = Q/(\mu L 2\pi r) \tag{1}$$

where

X = concentration in air (pCi/m^3)

Q = source strength (pCi/s)

μ = wind speed (m/s)

L = mixing depth of the atmosphere for HT and HTO (m)

r = radial distance from SRP (m)

A correction for stack height and vertical diffusion is made to the air concentration at the ground using the equations of Novak and Turner[22]. This correction does not seem to be important at distances greater than 3 km.

Dry deposition is expressed by the equation:

$$F = V_d\ (X - C) \tag{2}$$

where

F = flux density of material (pCi/m^2-d)

V_d = deposition velocity (m/d)

C = surface concentration (pCi/m^3)

The classical use of deposition velocity assumes that the surface concentration is zero. This is appropriate when HT is converted to HTO at the surface. However, a zero surface concentration is not appropriate for HTO which may accumulate at the surface. Equations of the form of equation (2) are used for deposition of HTO and HT to both the soil and the vegetation.

Wet deposition of HTO is calculated by assuming that rainfall is in equilibrium with air HTO concentration. This probably over estimates wet deposition because much of the rain water comes from heights in the atmosphere above the direct influence of SRP emissions. Wet deposition of HT is ignored because of the low solubility of hydorgen

Soil-movement is described by a mass balance equation for a surface soil layer and a deep soil layer. Water entering the soil is partitioned between vertical displacement to lower soil depths and absorption by vegetation on the basis of annual evaporation/rainfall ratios. The steady-state assumptions of the model smooths out the transient banded structure seen in field soils. The soil HTO concentrations calculated must be looked at as long-term averages. The balance equation for the surface soil is:

$$V_{ds}\ (X_H - C_s H) + V_{dTs}\ X_T + R_R\ X_H/H - T\ C_s - (R_R - T)\ C_s = 0 \quad (3)$$

where

V_{ds} = deposition velocity of HTO to soil

X_H = concentration of HTO

C_s = soil HTO concentration

H = Henry's law constant for HTO

V_{dTs} = deposition velocity of HT to soil

X_T = concentration of HT

R_R = rainfall rate

T = transpiration rate

The equations were coded in the IBM simulation language CSMP and solved using annual average climatic and deposition parameters. Table III summarizes the inputs to the model.

Model Simulations

Figure 2 shows the calculations of X_{HTO}/Q_{HTO} for a mixing depth of 700 m. The experimentally determined values of X_{HTO}/Q_{HTO} for the years 1974-1977 are plotted on the same figure.

The plot is not an exactly straight line due to incomplete mixing through the surface layer near the stacks and the addition of HTO converted from HT which increases the amount of HTO as the air moves downwind. The effect of changing HT/HTO at the source on the ratio X_{HTO}/Q_{HTO} is shown as dashed lines in Figure 2.

The percent of HT in the releases is known for 1977 only (16% HT). However, an estimate of the HT percentage can be made for 1974 and 1975. In 1974, 479,000 Ci of the total release were more than 99% HT because of an incident at the tritium separations area. If we assume that the chronic releases contained 16% HT as in 1977, then 40% of the 1974 release was in the form HT. Similar calculation for the 182,000 Ci inadvertently released in 1975 indicate that 47% of the released tritium for the year was HT. We must assume 1976 was a normal year. The inclusion of the HT/HTO ratio brings the dispersion estimates into a better agreement with the measurements, but some unexplained variation is left. The variation is to be expected because the 1974 and 1975 releases were 15 min puffs which do not meet the long averaging period of the steady-state, radial diffusion model. A strategy for a best estimate seems to be selecting a mixing height of about 700 m. This is a little lower than expected from the climatic data which indicate a range of 350-1500 m. The low value of mixing height may be explained by the fact that the higher values of mixing height occur for only short periods in the middle of the day.

It might be expected that the variation in vegetation HTO concentration is also related to the percentage of HT in the source release. This can be studied by looking at the ratio of vegetation/air HTO versus percent HT in the source air. Figure 3 shows the model calculation and the average ratios from data for the years 1974-1977. The ratios calculated above have been changed so that the 1975 inadvertent release will be credited to 1976 since the release took place late on December 31, 1975, and the vegetative effects would be expected over the first months of 1976. The results are surprisingly good. It seems most of the variation in the ratio of leaf HTO to air HTO is due to the HT percentage.

SUMMARY

The results of analyzing the tritium releases by simulation, based on a model constructed from controlled experiments, corroborate the hypothesis that the processes of dispersion, wet and dry deposition, and ecosystem water cycling are dominant in producing the tritium patterns seen in the vicinity of SRP. The results also emphasize the importance of knowing the form of the tritium in the atmospheric releases.

The model used was developed to investigate chronic releases where the wind direction and speed were uniformly distributed. However, the dispersion is linear enough that long term averages of samples taken uniformly around a point source should also show very similar radial distribution patterns. This may explain why the puff releases of HT in 1974 and 1975 do not seem to cause a large deviation from the radial pattern.

Dose estimates with radial population distributions are possible when the distribution of wind speed and direction are uniform. It is possible to make dose estimates for non-uniform distributions; however, the population must be weighted by the wind speed and direction.

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TABLE 1

Tritiated Water Concentration in Air and Vegetation Water (pCi/ml)

Year	*1974*	*1975*	*1976*	*1977*
Plant Perimeter (10km)				
Air	11.8	8.8	5.0	6.5
Vegetation	15.2	14.0	7.1	2.9
(40km)				
Air	4.9	4.5	1.8	1.8
Vegetation	12.3	6.0	2.6	0.6
(160km)				
Air			0.5	0.7
Vegetation	8.4	4.0	1.0	1.4
Source Strength pCi/s	2.87×10^{10}	1.55×10^{10}	8.43×10^{10}	1.24×10^{10}

TABLE 2

Tritiated Water Distribution in a Forest Ecosystem (pCi/ml)

Ecosystem Part	*Concentration*
Pine Needles	160
Pine Branch	114
Pine Trunk	127
Forest Floor	166
Soil	
0.0 - 8.0 cm	150
8.0 - 15.0	92
15.0 - 23.0	76
23.0 - 30.0	61

TABLE 3

Constant Input to Model Simulations

Deposition Velocities (cm/s)

Vegetation		*Soil*	
HT	*HTO*	*HT*	*HTO*
0.2	0.6	0.2	0.1

Precipitation (cm)	*Evaporation (cm)*	*Temperature (°K)*	*Humidity (%)*
119 *cm*	89 *cm*	18.0	71.0

Wind Speed (cm/S)	*Stack Height (m)*
6.0	63.0

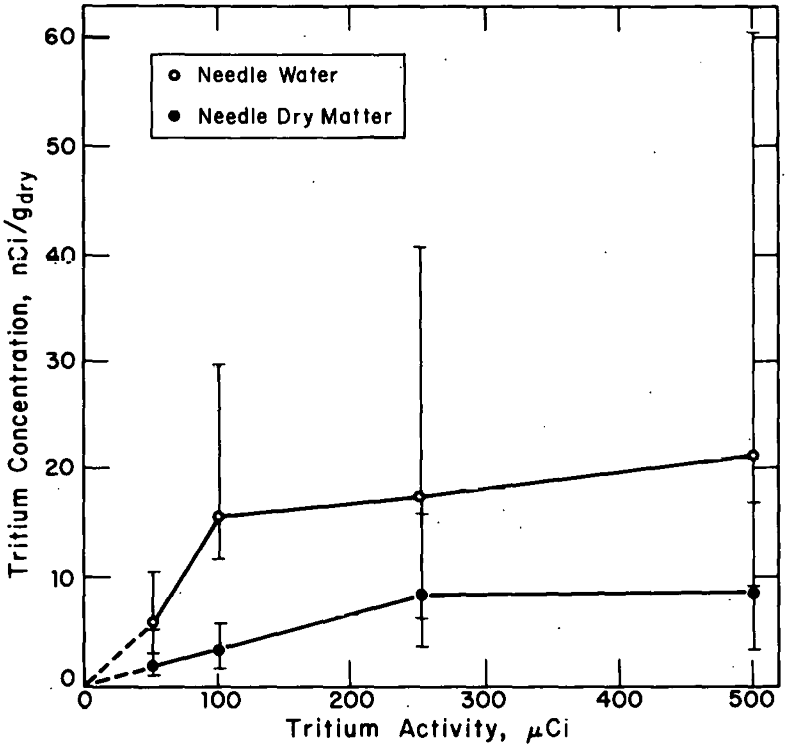

FIGURE 1. Activity of Tritium in Water and Dry Matter After Exposure to Gaseous Molecular Tritium

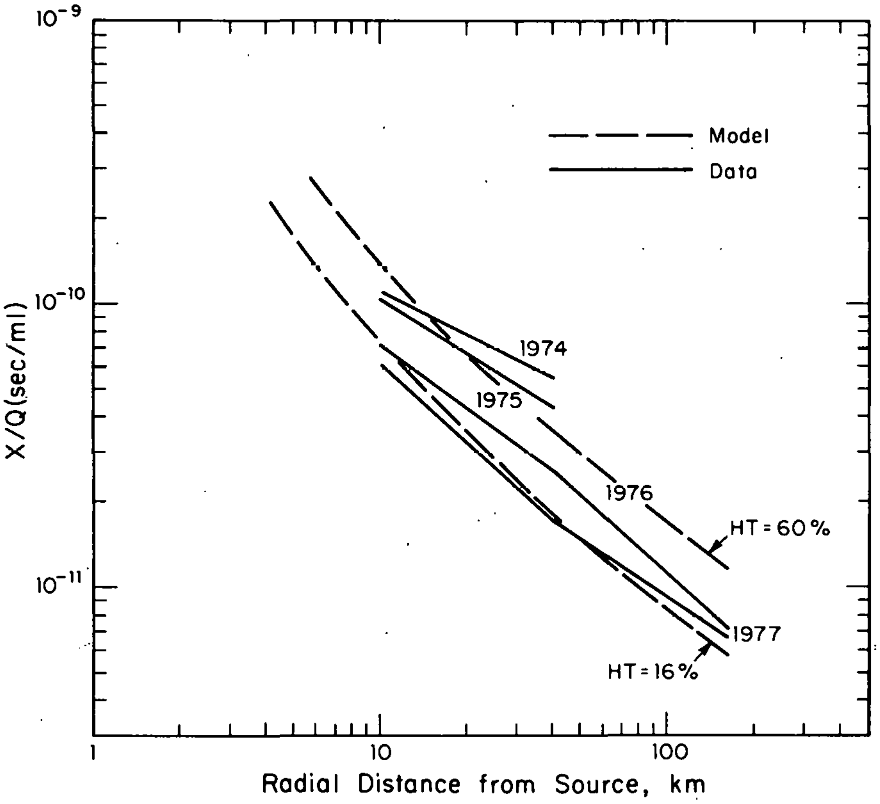

FIGURE 2. Model and Measured Tritium Concentrations in Air Moisture. Mixing Layer is 700 m.

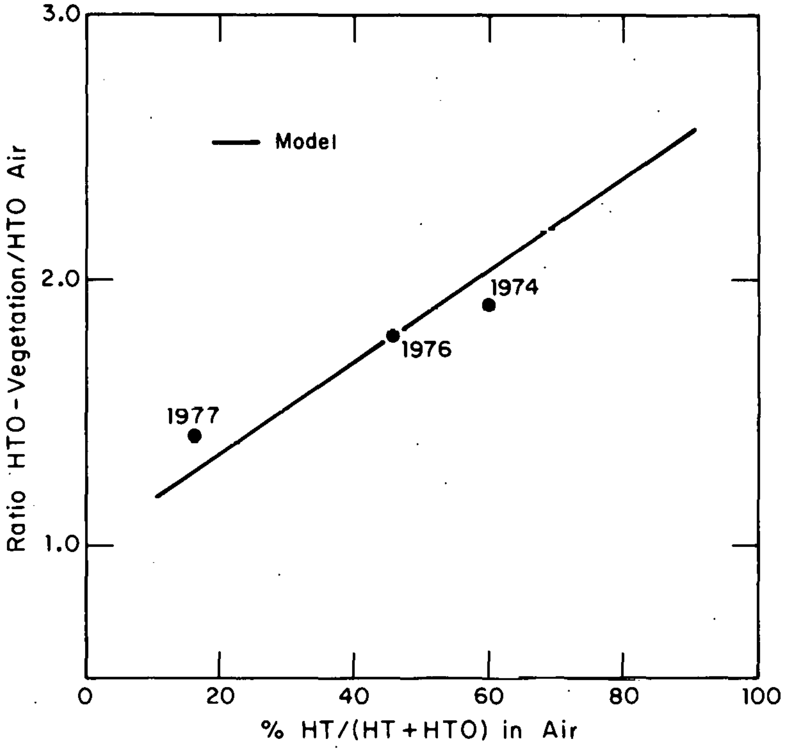

FIGURE 3. Comparison of Model and Measured Tritium Concentration in Vegetation Water as Effected by Tritiated Hydrogen Content.