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THE TRANSPORT, DISPERSION, AND CYCLING OF TRITIUM IN THE ENVIRONMENT (U)

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MASTER

PREFACE

This report was compiled in response to a request from the U.S. Department of Energy, Office of New Production Reactors to review the state of understanding of tritium transport and cycling in the environment. The ability to understand and predict the movement of tritium in the environment is basic to predicting the effect of tritium releases from nuclear facilities on humans and other potentially sensitive parts of the environment.

The sources of information used in this review include the published results of experiments done by government, academic, and industrial agencies. The motivation of many of these reports was to learn more about tritium as a biomedical and environmental tracer, as well as to specifically study the effect of tritium released to the environment from military and industrial activities. The record of tritium monitoring around known tritium sources is an additional source of information used in compiling this report. If the understanding of tritium transport and cycling is adequate, the physical and biological processes determined by detailed, controlled experiments should explain the patterns of tritium concentration found in the vicinity of sources that release tritium to the environment.

Many of the monitoring data used in this report were collected at the Savannah River Site (SRS). The SRS produces most of the tritium made in the United States and has a 35-year history of tritium monitoring in the surrounding environment. The environmental monitoring data were collected to ensure that SRS operations did not have a detrimental effect on the surrounding population and would meet all applicable standards for exposure of the offsite public.

The current annual limits for atmospheric tritium exposure to the public is 500 mrem (International Commission on Radiological Protection) and 100 mrem (Department of Energy). The atmospheric concentration guide (40 CFR 61) derived from the Department of Energy exposure limit is 150 pCi/m^3 of air. The EPA drinking water standard for tritium is 20,000 pCi/L of water. The concentration guides have been exceeded near facilities within the SRS boundary. However, the average tritium concentration in air at the SRS boundary (where the public is exposed to the highest concentration) is 7% of the concentration guide. The annual average concentration of tritium in the Savannah River (the only water drained from SRS which is used for drinking) has never exceeded 70% of the EPA drinking water standard. At no time has the worst case annual dose calculated for atmospheric or liquid releases exceeded 25% of the EPA exposure limit of 10 mrem (10 CFR 61).

ABSTRACT

The processes which determine transport, dispersion, and cycling of tritium are identified for atmospheric, terrestrial, aquatic, and groundwater systems. The processes are discussed in terms of the storage capacity for tritium in each component of each system and ranges of residence times are derived. The residence times of each component of the systems are discussed in terms of the residence time of the whole system for transient releases of tritium into different components of the systems. The role of the ocean as a sink for tritium is described. The concentration of tritium in the system at steady state is described in terms of the inputs and outputs to the components of the systems.

The analysis indicates that the key residence time for a specific release of tritium into the environment is dependent on both the residence time of the components and the means of introduction into the environment. The initial concentration and residence time of tritium in the terrestrial system after an exposure to tritiated water vapor are determined by the atmospheric and vegetative conditions at the time of the release. The dominant residence time is that of the vegetation. On the other hand, the initial concentration and residence time of tritium in the terrestrial system after an exposure to tritiated hydrogen are determined by the atmospheric and soil conditions at the time of the release. The dominant residence time is that of the soil. The initial concentration and residence time after a liquid release to the soil surface are determined by the diluting soil water content and the residence time for water in the rooting zone of the soil. Little tritium enters the organic fraction of terrestrial systems from transient releases of gases or liquid water.

The steady-state concentration of tritium in the different components of a terrestrial system exposed to atmospheric releases of tritiated water is generally somewhat less than the concentration in the atmosphere. The concentration of water in the soil is diluted by the relatively low concentration in rain water. The concentration in the vegetation water is generally between that in the atmosphere and that in the soil. The concentration in the organic fraction of the vegetation is close to, but probably slightly less than, that in the water of the vegetation. Animals exposed to the tritiated water of the atmosphere, rain, and food will tend to have a concentration of tritium in their body water close to that in their drinking water. The concentration in the organic fraction of animals will be influenced more by the organic tritium in their food. There is no indication of biomagnification of tritium in terrestrial systems.

Tritium introduced into an aquatic system will remain associated with the water in the system, diluted by water added from other sources. Aquatic organisms generally have an internal tritiated water concentration very close to that of the water in which they live. As in terrestrial organisms, the incorporation into organic components is much slower. Aquatic organisms may also incorporate measurable amounts of tritium from detritus, dead organic matter. This can dilute or elevate the organic tritium concentration of the system, depending on the concentration in the detritus. There is no indication of biomagnification of tritium in aquatic systems.

Tritium dynamics in groundwater systems are related to the movement of water. The large differences in the velocity of movement and residence times in groundwater systems require that each system be studied separately to understand the transport of tritium in that system.

Analysis of global tritium cycling indicates that most tritium released to the atmosphere or aquatic systems will ultimately accumulate in the ocean. The ocean has an extremely large capacity for diluting tritium and ocean water tritium concentrations will not be elevated greatly as a result of any tritium facility planned at the present time.

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INTRODUCTION

The safe and environmentally acceptable operation of facilities which produce or use large quantities of tritium requires an understanding of the effect of tritium on human health and the environment. The effect of tritium is the result of the absorbed dose of radiation, which, in turn, is determined by the tritium concentration and time over which an individual or component of the environment is exposed. The objective of this paper is to review our knowledge of the physical, chemical, and biological processes which determine the concentration and residence time of tritium in the various components of the environment.

Environmentally Important Characteristics of Tritium

Tritium is an isotope of hydrogen. The three isotopes of hydrogen, found in nature, are protium, deuterium, and tritium. Protium is the most abundant of the three, and the lightest, its nucleus contains one proton. The deuterium nucleus contains one proton and one neutron while tritium contains one proton and two neutrons.

Tritium is radioactive, decaying to helium-3 by emission of a beta particle. The half-life of tritium is 12.26 years. Based on the half-life and an atomic mass of 3.017, the specific activity of elemental tritium is approximately 9,670 Ci/g. The energy of the tritium-decay beta particle averages 5.69 keV with a maximum energy of 18.6 keV. The energy is small compared to most other radionuclides. The stopping distance in air is about 7 mm. The tritium beta is stopped by a 0.01 mm thickness of paper or similar material (such as the outer, dead layer of human skin).

Tritium is classified as a long half-life radionuclide because the 12.26-year half-life is long when compared to the time needed for tritium to disperse in the environment. However, because of the low energy, and subsequent low penetration of the tritium beta, there must be intimate exposure to the tritium atom for there to be any effect on biological systems. This means that only exposure through internal uptake needs to be considered in assessing radiation dose.

It is likely that hydrogen forms more chemical compounds than any other element. It is involved in almost all the compounds of carbon, plus numerous inorganic compounds. Tritium forms the same chemical compounds as any other isotope of hydrogen. The most important compound of hydrogen is water. Tritium may become incorporated in any hydrogen-containing compound.

Incorporation of tritium into hydrogen-containing compounds can take place in a number of ways (Peng 1973, Wesson 1973). Incorporation during synthesis from a labelled precursor is done naturally in photosynthesis and other biological processes (Moses and Calvin 1959, Belot et al. 1983). Exchange of tritium for other isotopes of hydrogen is another method of tritium incorporation. Exchange is usually associated with labile sites such as hydroxyl groups which exchange rapidly with compounds such as water and many organic compounds (Lang and Mason 1960). Hydrogen atoms held by carbon bonds are generally considered fixed. However, lability is a relative term since almost all hydrogen sites in compounds will exchange given enough time. Tritium can also be incorporated into compounds because of decay. The energy of decay, through ionization or rebound of the associated atoms, can lead to incorporation. In addition, the free radical of tritium formed when one of the atoms of molecular tritium gas decays is very reactive. In the natural environment, exchange with water and incorporation through photosynthesis and subsequent synthesis of other compounds in the food chain are the most import processes.

While tritium generally acts as any other isotope of hydrogen, there are differences between the physical and chemical characteristics of tritiated compounds and those of the other isotopes of hydrogen. The physical isotopic effects found in natural systems, while sometimes measurable, are small (Jacobs 1968). Tritiated water has a vapor pressure that is about 10% lower and a molecular diffusion coefficient in air that is about 5% lower than natural water.

Chemical reaction rate constants for tritium are lower than those for protium or deuterium. In the some reactions this effect can be large, on the order of 100 times slower reactions for tritium than protium. Equilibrium isotope effects appear to be much smaller, ranging from 0.7 to 3.3 for exchange of a variety of compounds with tritiated water (Wesson 1973). The integrated effect of chemical isotopic effects on biological systems appears to be even smaller than those for individual chemical reactions. It is rare to be able to measure these effects against the background variation in natural systems. Because the reactions involving tritium are slower than those involving the other isotopes of hydrogen it is reasonable to expect tritium to be left behind in a series of exchanges or reactions in biological systems. Therefore, effects such as biomagnification in food chains would not be expected.

The Distribution of Hydrogen in the Environment

Since tritium is a form of hydrogen and acts as hydrogen in all important ways, tritium will be transported by the same processes and will eventually exist in the same physical locations and the same chemical forms as hydrogen. The pool of hydrogen is large. It is estimated that 0.76% of the mass of the earth is hydrogen, ninth among all the elements. Hydrogen is third in the abundance of atoms in the earth's crust at 13.5%, behind oxygen and silicon. Most of the hydrogen is in the form of water and most of the water is in the oceans. Much of the remaining hydrogen is in the form of minerals such as hydrates, petroleum deposits, or in the organic material of organisms.

Sources of Tritium in the Environment

Tritium is a naturally occurring isotope of hydrogen. The source of natural tritium is bombardment of nitrogen by cosmic ray neutrons in the upper atmosphere. The reaction has the form

$$^{14}N_7 + {}^1n_0 \text{->} {}^3H_1 + {}^{12}C_8$$

The natural occurrence of tritium in rainwater is 1-10 atoms of tritium per 10^{18} atoms of hydrogen. The production rate has been calculated to be between 4 and 8 million Ci/year. This leads to a natural tritium inventory of 70-140 million Ci (UNSCEAR 1982).

Since 1954, the global inventory of tritium has been dominated by the tritium produced in testing nuclear weapons. Most of this tritium was injected into the stratosphere. Miskel (1973) has estimated that 8,000 million Ci of tritium were introduced into the atmosphere between 1955 and 1962. Extrapolation of Miskel's initial inventory to 1988 indicates an inventory of 1,600 million Ci of weapons- testing tritium still existing in the global environment.

Tritium is also produced by nuclear power reactors. A number of nuclear reactions are involved in tritium production in power reactors. The most important reaction in light water and gas cooled reactors is ternary fission of uranium and plutonium (Albenesius 1959). Tritium is also produced in reactions involving boron and cadmium in control rods and in the cooling water. Tritium production is estimated to be 0.75 PBq/[GW(e) a] for the various types of light water and gas cooled reactors (UNSCEAR 1982). In heavy water reactors, tritium is produced by neutron activation of deuterium in the moderator water. The tritium yield from deuterium activation is estimated to be about 30 times greater than the yield of ternary fission, leading to a heavy water reactor tritium production of 25 PBq/[GW(e) a] (UNSCEAR 1982).

Most of the tritium produced in power reactors is retained in the fuel elements. The exception is the tritium produced in the moderator or cooling water. Much of this tritium is released to the environment. Table 1 shows the releases to the environment that can be expected from the different power reactor types.

The tritium in power reactor fuels is released when the fuel elements are reprocessed to recover plutonium and unused uranium. Almost all of the undecayed tritium in the fuel is released to the environment.

Tritium is also produced for use in national defense, medical and biological research, and consumer goods such as watches and other luminous products. Tritium is produced by irradiating lithium targets with neutrons in a nuclear reactor according to the following reaction (Jacobs 1968).

$$^{6}Li_{3} + {}^{1}n_{0} \rightarrow {}^{3}H_{1} + {}^{4}He_{2}$$

Separation of the tritium from the targets and subsequent handling results in some releases to the environment.

Table 1. Tritium releases per unit of power production from nuclear power reactors (TBq/[GW(e)] (UNSCEAR 1982).

Type of Reactor	Atmospheric	Liquid	Total
Pressurized Light Water	7.8	38	45.8
Boiling Light Water	3.4	1.4	4.8
Gas Cooled	11	25	36
Heavy Water	540	350	890

Small additional amounts of tritium are released to the environment by the use and discard of consumer products and biomedical wastes. While it was once thought that backlit, tritium-illuminated watches would become a major source of tritium in the environment, a reduction in their popularity now seems to make this a remote possibility (Combs and Doda 1979).

In the future, fusion reactors may become a major source of tritium to the environment. Tritium is a fuel in the most promising reaction schemes for the production of fusion energy. However, it is still too early to make accurate estimate of the inventory and releases of tritium from fusion reactors (Dinner and Gulden 1986).

The remainder of this paper describes the factors that control the concentrations of tritium in the environment found around tritium-producing facilities. This will be done by dividing the environment into characteristic systems in which similar transport processes are active. Then the tritium transport processes, the chemical transformations, and hydrogen pools will be identified in each system. These factors will be used to determine the transport rates and residence times of tritium in the various components of the system. Finally, the residence and transport times will be used to determine the integrated response of tritium concentration to tritium inputs in the system.

TRITIUM IN THE ATMOSPHERE

There is a large volume of literature which quantitatively describes the transport of materials in the atmosphere (see Pasquill and Smith 1983 or Slade 1968). It is not the purpose of this paper to review this literature but a few examples will be presented to illustrate the effect of the atmospheric processes which are important in determining the concentration of tritium in the environment around a source of tritium. These examples will act as a framework into which the effects of other processes affecting tritium transport can be integrated.

The Effect of Atmospheric Transport on Concentration

In many cases the air concentration measured perpendicular to the wind direction at some distance downwind from a source will have a Gaussian (bell shaped) distribution. The width of spread will depend on both the size of the eddies in the atmosphere and the time over which the concentration is averaged. Figure 1 shows three different Gaussian cross-wind distributions that will be measured along the same transect, depending on the length of time the concentrations are average over. The narrowest distribution represents the time scale over which the smoke of a plume is seen, on the order of seconds. The second Gaussian transect is averaged over a time that integrated the loops in the release plume, usually in the order of minutes. Notice that at a sufficient distance downwind the size of the "visible" plume has grown to the point that the large loops of the meanders are now included in the visible plume. The plume path with the largest spread is for time periods when a steady shift in wind direction has spread the plume, successively, over an area downwind from the source.

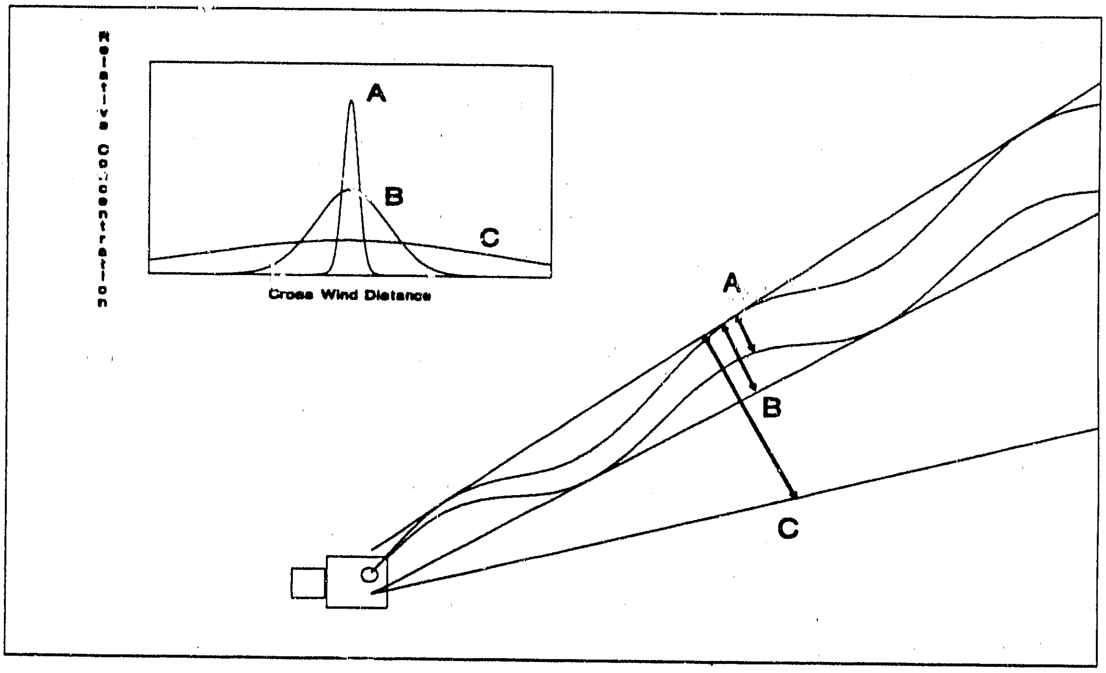

Figure 1. An example of the effect of sampling time on the measured cross-wind distribution of tritium following a release. Transect A is for a very short sample time. Transect B averages the concentration within the meanders of the plume. Transect C includes a wind shift during the sample period.

The fact that all of these processes lead to Gaussian or Gaussian-like distributions probably explains why this distribution is so common. On the other hand, it is easy to think of ways that non-Gaussian distribution can be formed from the same processes which produce Gaussian concentration distributions. Even if the turbulent diffusion process produces a Gaussian distribution on short time scales, a nonuniform wind shift or nonuniform release rate will produce a non-Gaussian distribution. Another factor, when a plume from an elevated source is descending to the surface, is the elevation of the surface. After the 1974 tritium gas release from Savannah River Site (SRS), the tritium concentration distribution of vegetation collected along a transect 12 km from the source was found to have two peaks. It was found that the two peaks and the intervening "valley" coincide with topography along the sampling transect (Murphy et al. 1977).

The distribution of concentration at groundlevel along transects beginning at the source and moving away in the direction of the wind will look like figure 2 (adapted from Turner 1969). If the source is elevated, the concentration will increase as the plume approaches the ground. The vertical distribution in the plume is often initially Gaussian as the plume moves away from an elevated source. The highest concentration at the surface is found where the center line of the plume touches the surface. Beyond the maximum point the concentration decreases with increasing distance from the source. At first the rate of decrease is determined by the vertical and horizontal spread of the plume. At some point downwind the vertical spread is limited by the height of the mixed layer of the atmosphere and the rate of concentration decrease is a function of only the horizontal spread. This pattern is the same along any transect measured downwind of a Gaussian plume. However, the concentration is lower at a given distance from the source as the transects are further off the centerline of the plume. The general pattern also holds when the concentration is averaged over the whole 360 degrees of the arc around the source. Figure 3 shows the average decrease in concentration measured in the vicinity of the SRS (Murphy et al. 1990). If the concentrations are averaged for relatively long periods of time (years), the average concentrations can often be characterized by the average decrease in concentration weighted by the proportion of wind blowing in a particular direction. Figure 4 illustrates this for concentration data collected over a five-year period in the vicinity of SRS (Murphy et al. 1990).

The Effect of Processes Removing Tritium from the Atmosphere

Removal of tritiated compounds from the atmosphere can take place by transfer to the surface or by chemical transformation into another compound. The latter process does not actually remove tritium from the atmosphere, it only transforms it into another compound. The only transformation that can accomplish actual remove is radioactive decay. However, chemical transformations are important because they can change the rate of exchange between the atmosphere and the surface.

Probably the most well studied atmospheric chemical transformation involving tritium is the oxidation of tritiated hydrogen gas to tritiated water. Tritiated water is readily removed from the atmosphere by a number of processes which do not as readily remove tritiated hydrogen. The results of a number of studies indicated that the oxidation of hydrogen gas in air takes place very slowly, with a half-life on the order of 100 or more years (Eakins and Hutchinson 1973, Easterly et al. 1985). Mason and Ostlund (1979) estimate that the half-life of tritium in the atmosphere is 6.5 years. The more rapid re-

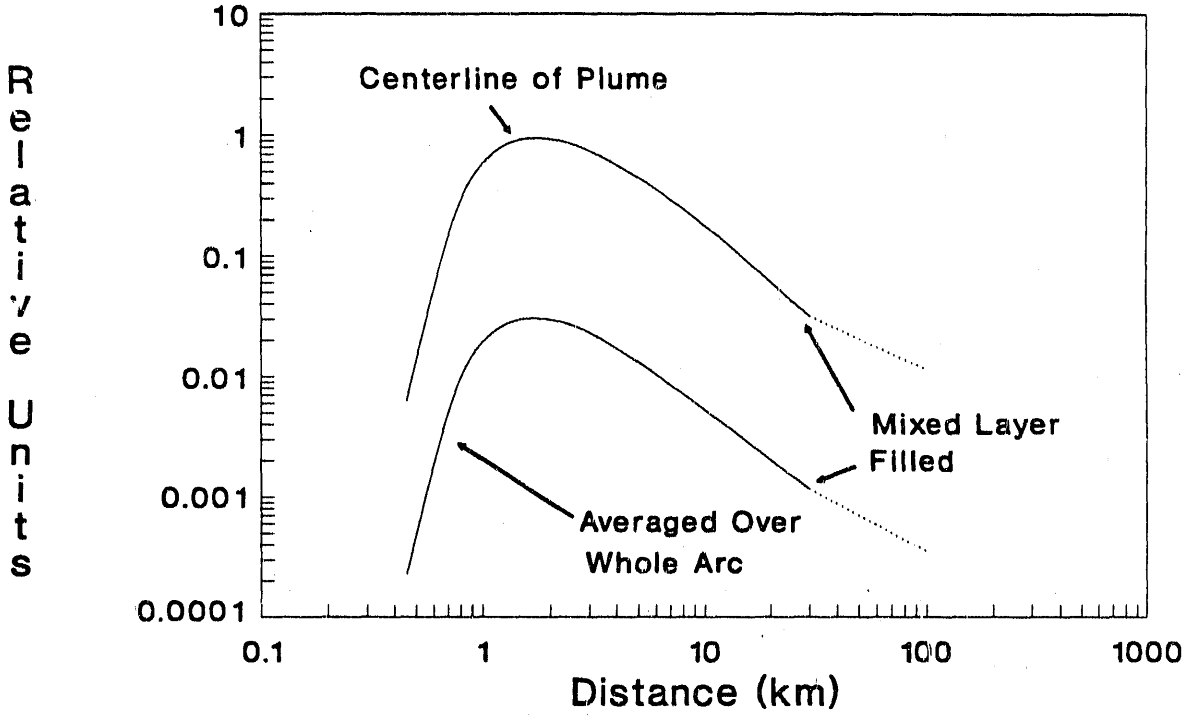

Figure 2. An example of the decrease in centerline and long-term average tritium concentration from an atmospheric release from an elevated stack (stack height 70 meters, with atmospheric moderately stable temperature stratification).

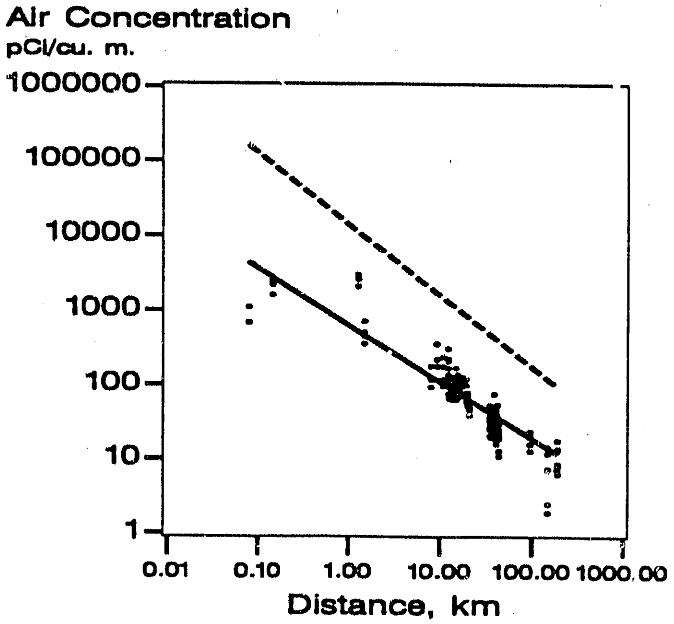

Figure 3.The decrease in average (1982-86) and maximum (all available data) air tritiated water concentration with distance from the center of SRS.

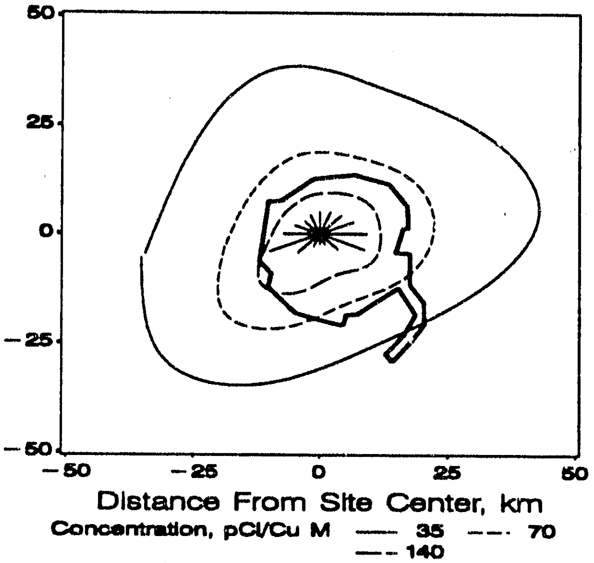

Figure 4. The directional distribution of tritiated water vapor in the vicinity of SRS (1982-86 data).

moval of tritiated hydrogen from the atmosphere suggests that some process other than oxidation in the free atmosphere is important in removing tritiated hydrogen from the atmosphere. These processes are the ones which transport atmospheric tritium to the surface.

Bardolie (1985) presented data which indicated that the oxidation of tritiated hydrogen in the atmosphere away from the surface might be more rapid than previous studies had indicated. His results led to two experimental releases of tritiated hydrogen gas to determine if the results could be duplicated. One experiment was held in France (Djerassi and Gulden 1988) and the other in Canada (Brown et al. 1988). The results confirmed the earlier work which showed very low oxidation rates under natural conditions (Paillard et al. 1988, Brown et al. 1990).

Surface removal processes are called, by analogy with particle removal from the atmosphere, deposition. The two types of depositional processes are identified as dry or wet deposition, based on whether the deposition process involves or does not involve precipitation. Deposition of tritium in precipitation can be the result of two processes. Rainout is the process of incorporation of tritium in the rain drops as they form in a cloud. Washout involves the incorporation of tritium into precipitation falling through a plume (Chamberlain and Eagleton 1964). The maximum tritium concentration in a raindrop or other form of precipitation is determined by the equilibrium concentration of the gas in solution. Once saturation is reached, no more tritium will be removed by the precipitation. Tritiated water is removed more rapidly by the wet depositional process than either tritiated hydrogen or methane because of its relatively high equilibrium concentration in water.

Figure 5 illustrates the effect of wet deposition of tritiated water from a plume under two different rainfall rates. The result, based on the calculations of Long (1978), are also shown in terms of the deposition of tritium on the surface. Notice that a higher rainfall rate causes the initial deposition to be greater than a lower rainfall rate. However, depletion of the plume near the source results in lower air concentrations and thus lower deposition at further distances from the source.

Since deposition is strongly influenced by the tritiated water concentration of the air, the spatial distribution of tritium around a source should be related to the spatial distribution of air concentration. Figure 6 shows this to be the case for the distribution around SRS.

The rate of dry deposition of tritiated gases is influenced by diffusion from the bulk of the atmosphere to the surface and by processes which are characteristics of the surface. In most cases it is the surface processes which predominate. These processes take place in the vegetation and the soil of terrestrial systems and at the water surface of aquatic systems. The individual processes will be discussed in detail in later sections of this paper. Figure 7 shows that the effect of dry deposition on the distribution of tritium in vegetation around a source is similar to that of wet deposition because of the relationship of the dry deposition to the air concentration (Murphy et al. 1990).

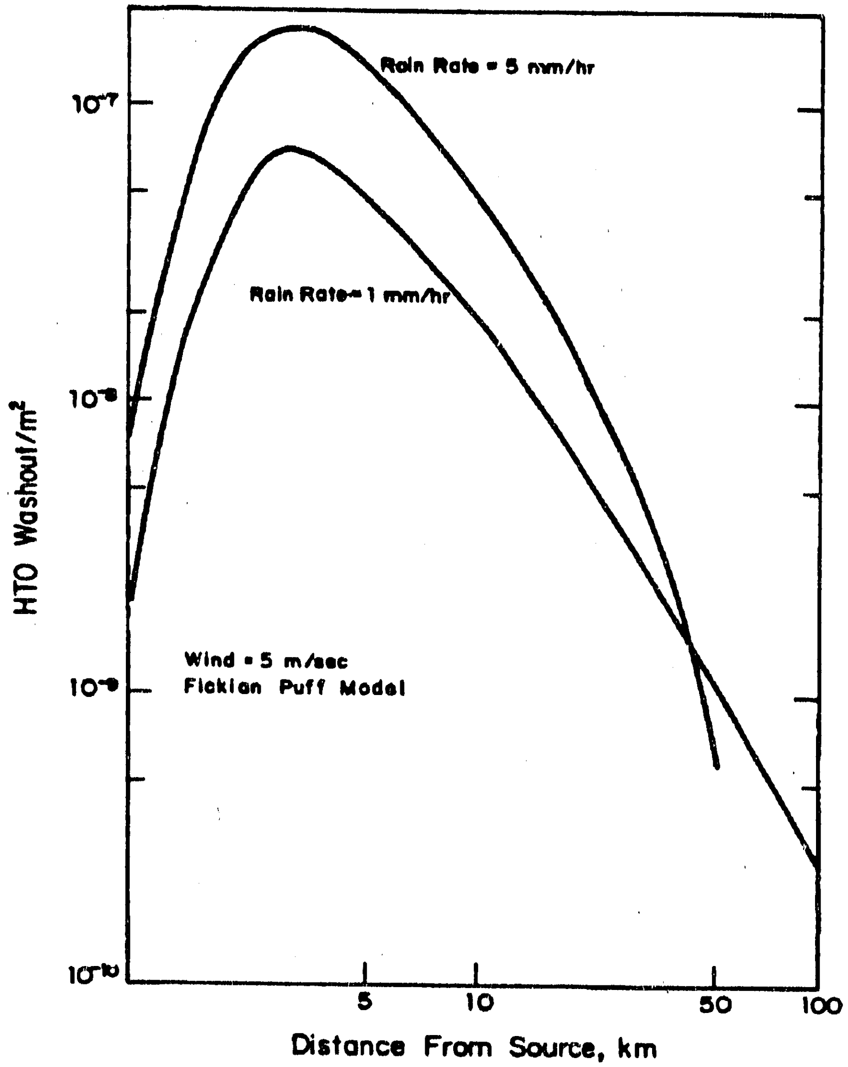

Figure 5. The effect of rainfall on the tritiated water vapor washed out of a plume.

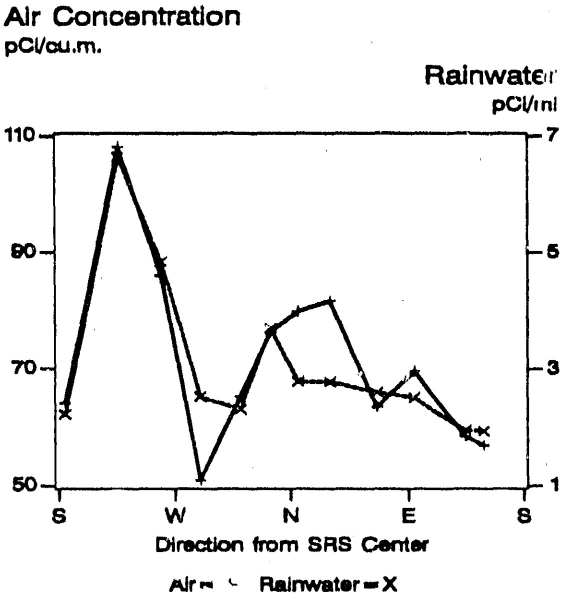

Figure 6. A comparison of the concentration of tritium in air and rainwater in different directions from the center of SRS.

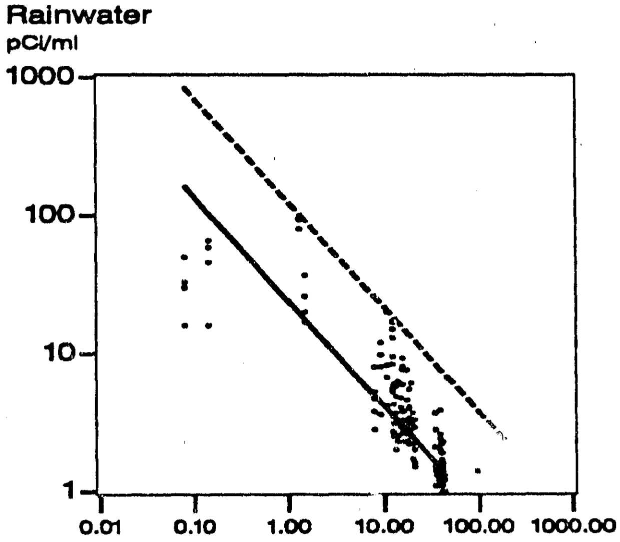

Figure 7. Variation of average (1982-86) and maximum (all available data) tritium concentrations in rainwater with distance from the center of SRS.

Exposure to Atmospheric Tritium

The exposure of an individual to atmospheric tritium is proportional to the air concentration integrated over the period of exposure. As indicated previously, the concentration will vary with the type and amount of release and meteorological conditions, such as wind speed. In a short release the exposure time will depend on the speed and path the release takes when passing the individual.

When the release extends over a longer period of time, the average exposure will be proportional to the average concentration patterns shown in figure 2-6. The general pattern will result in greater concentrations in the directions of more frequent winds. There is normally a general decrease in exposure with distance along any radial direction from the source.

Deposition removes tritium from the air, leading to lower exposure from contact with the air. This effect is not large near the source; however, the eventual removal of tritium from the atmosphere is largely through deposition.

NCRP Report Number 62 (NCRP 1979) summarizes the results of a number of experiments which estimate the half-life of tritiated water in the atmosphere to between 21 and 41 days. This is equivalent to a deposition velocity (the amount of deposition per unit area divided by the concentration in atmosphere) of between 0.4 and 0.8 cm/sec if the height of the tropopause is taken to be in the range of 12 to 15 km.

The deposition velocity of tritiated hydrogen can be calculated in the same way. Mason and Ostlund (1979) have estimated that the residence time for HT is 6.5 years. Again, using the height of the mixed layer as beteen 12 and 15 km, the deposition velocity is between 0.04 and 0.05 cm/sec.

The deposition of tritiated water is not irreversible. Tritiated water that is absorbed by vegetation or soil as a plume passes over a location will re-enter the atmosphere after the plume has moved and the concentration of tritiated water at the surface becomes greater than that in the atmosphere. When integrated over time periods greater than a few days, only that part of the tritium that is removed by percolation into soil below the root zone or that part which is incorporated in organic matter is removed from the atmosphere. Deposited tritiated hydrogen is oxidized at the surface (largely in the soil) and in this sense the deposition is irreversible (Ehhalt 1972, Murphy et al. 1977, McFarlane et al. 1978). However, the tritiated water produced by oxidation becomes part of the soil water and in this form re-enters the atmosphere (Kirchmann et al. 1986a, Belot et al. 1988). Depositional processes are important because of this influence on atmospheric concentrations and because deposition is the primary path for tritium to enter terrestrial systems after an atmospheric tritium release.

TRITIUM IN THE TERRESTRIAL ENVIRONMENT

In this discussion, the terrestrial environment is defined as the layer at the land surface of the earth that contains the majority of the living organisms. This includes the atmosphere within plant canopies, the soil down to the rooting depth of the vegetation, and the vegetation, animals, and other organisms living in this layer. Tritium transport and cycling in the terrestrial environment is determined by physical and chemical processes taking place in each of the three parts and the transport of tritium between the parts.

Soil Tritium

Tritium can enter the soil from the atmosphere in precipitation or by diffusion of gaseous tritiated compounds. The diffusion of tritiated compounds into the soil is impeded by vegetation and any vegetative litter on the soil surface. For this reason the transport of tritiated water to the soil is usually much lower than the transport to the vegetation at sites having a vegetative cover (Sweet and Murphy 1983, Kirchmann et al. 1986). Under similar conditions, transfer of tritiated water to bare soil is higher than transfer to soil at vegetated sites. Once tritiated water has reached the soil, it diffuses into the soil through the air-filled pores and mixes with the water in the soil. Gaseous diffusion in the soil is a function of the gas-filled pores. Course, sandy soils have a more open structure than clay soils to gas diffusion. Diffusion is lower at high soil water contents because the amount of diffusion is limited by water filling of the soil pores. Garland (1979) measured tritiated deposition velocities between 0.09 and 0.91 in bare soil.

Tritiated hydrogen and tritiated methane also diffuse into the soil where they are dissolved in the soil water. Degradation and production of these gases is possible in soils. The chemical reactions are mediated by soil microorganisms. The direction of the reactions depend on the soil environmental conditions (Bohn et al. 1979). Under aerobic conditions these compounds will be oxidized to tritiated water. Under reducing condition these same gases can be produced from degradation of organic material.

There has been considerable interest in the oxidation of tritiated hydrogen because the radiotoxicity of tritiated hydrogen is 20,000 times greater than that of tritiated hydrogen (ICRP 1979). As indicated previously, the oxidation takes place slowly in the atmosphere. It is thought that the main site of the conversion of tritiated hydrogen to tritiated water is in terrestrial soils. Once tritiated hydrogen has penetrated the soil, it goes into solution in the soil water. Soil microorganisms, producing the enzyme hydrogenase, are able to break the bonds in the diatomic hydrogen molecule (Schlegel 1966). This is the first step in oxidizing tritiated hydrogen to tritiated water. At the low tritiated hydrogen concentration normally found in the environment, the oxidation reaction is very efficient. However, because of the comparatively slow diffusion into the soil and the low solubility of hydrogen in the soil water, the conversion rate of tritiated hydrogen from an atmospheric source to tritiated soil water is not rapid. Figure 8 shows the range of deposition velocities measured by Dunstall et al. (1985) in a variety of natural soils.

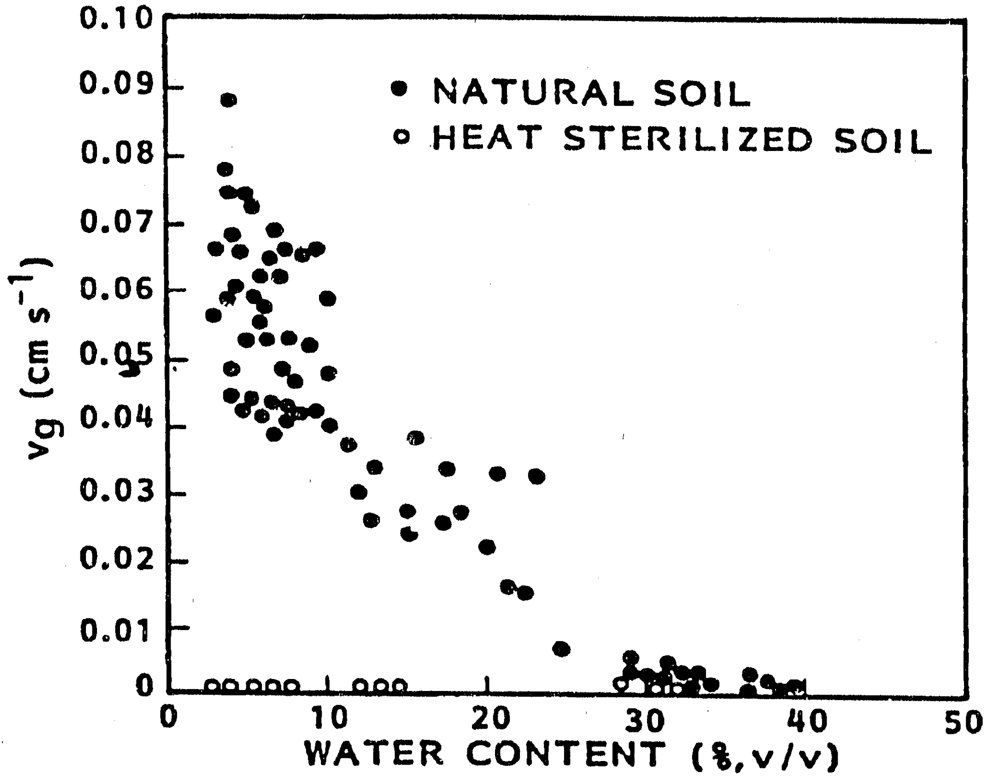

Figure 8. The deposition velocity of tritiated hydrogen to soil (from Dunstall et al. 1985)

Tritium can also enter soil through direct application of tritiated water, either from rainfall, a spill of contaminated water, or an outcropping of contaminated groundwater. In general, the transport of tritiated water in the soil will involve the same processes and follow the same paths as other water. Liquid water moves through the soil in response to gravity and capillary tension (see Hillel 1971 for further discussion and references on soil water movement). Capillarity causes soil water to be held more tightly as the size of the pores containing the water decreases. The water held in the small pores will be under a greater tension and water will tend to move from larger to smaller pores. Given a constant distribution of pore sizes in a particular soil, the water will move from the soil region having a greater water content, that is having water in large pores, to a soil region having a lower water content, that is soil containing water only in the smaller pores. Since the pore size distribution in soils is very variable, in general, water does not move in response to water content but to water capillary tension. As a result of the combined influence of capillarity and gravity, water will move vertically in soil by miscible displace. Tritiated water entering the surface of the soil will move downward in a layer as it is displaced by the water entering the soil from above. Figure 9 shows the resulting distribution of tritiated water added to the surface after displacement by subsequent rainfall (Jordan et al. 1970). The movement of water due to capillarity and gravity is by mass flow and the concentration remains the same in the water-filled pores as the water moves downward. However, there is some mixing of water within the soil because the water is dispersed among the different pores in the soil when it is transported. Some water ends up having a longer flow path than other water. Thus some water moves ahead and some lags behind and is mixed with water at either edge of the layer containing tritiated water.

Tritiated water can also move by vapor transport. Under constant temperature conditions, vapor transport will take place along the same tension gradient as liquid water because the equilibrium vapor pressure in the pores is a function of the tension the water is held under. The vapor pressure is also a function of soil temperature and the vapor pressure gradient will tend to move water from regions of high temperature to regions of low temperature. Vapor transport is by diffusion through the air-filled pores of the soil. Since vapor transport in soil is by diffusion, tritiated water will move in response to the difference in tritiated water concentration between two locations in the soil, independent of the movement of water vapor. This means that tritiated water can diffuse from a region of high tritiated water concentration to a region of low tritiated water concentration even when there is no net transport of water vapor.

As more water is added to the surface, the tritiated water will be moved to below the depth where it will interact with the vegetation. Water moving at these depths is discussed in the section on tritium in groundwater. However it is important to remember that tritiated water that has been transported below the root zone during periods of high rainfall can re-enter the rooting zone by upward movement when the soil in the rooting zone is dry enough that the soil water tension gradient in the upward direction is greater than the gravity force pulling the water downward. Water can also be held in the rooting zone by impeding soil layers of low hydraulic conductivity which channel the water in a horizontal direction (Jordan et al. 1970)

Absorption of soil water by plant roots supplies the majority of water for terrestrial vegetation. The extraction of water is through the action of capillary tension formed in the plants by removal of water

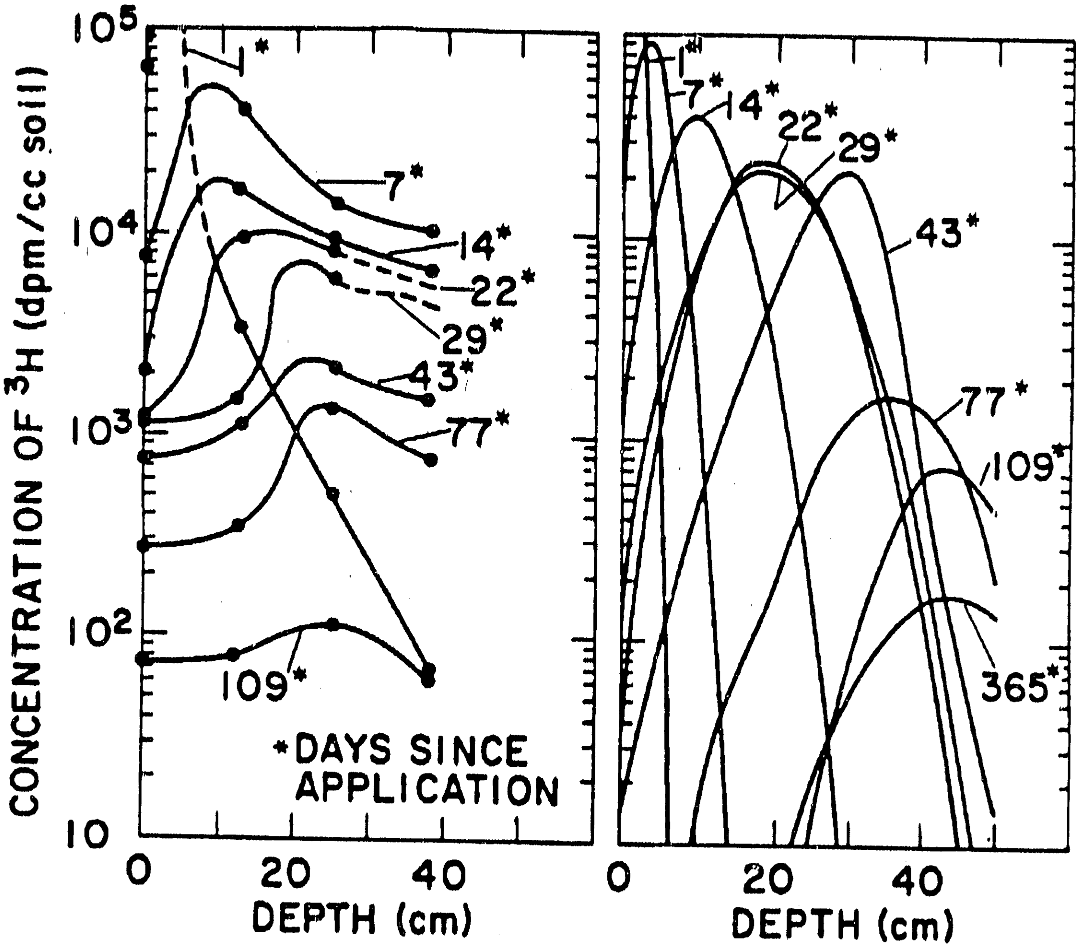

Figure 9. The movement of soil water with time after application of tritiated water at the surface (from Jordan et al. 1970)

from the leaves by evaporation (Kramer 1969). The tension draws water from cells in the water conducting organs and other cells of the plant which are then under tension. Water moves out of the soil into the plant roots along this tension gradient. Extraction of water from the soil is also a bulk flow process and the water moves into the plant at the same tritium concentration as the tritiated water concentration in the soil pores near the roots. Rooting depths vary greatly between different types of plants, and between the same type of plant grown in different soils and climates. In many soils of the Southeastern U.S., plant roots are impeded by heavy clay subsoils. In arid regions with highly permeable soils, plant roots have been found in a mining excavation at a depth of over 300 feet (Phillips 1963).

The soil is also a site of incorporation of tritium into organic compounds. Much of the organic tritium in soil is derived from plants growing in the soil. However, small amounts of tritiated organic matter are produced by autotrophic soil microorganisms that use tritiated hydrogen as an energy source. Soil microorganism participate in a large number of metabolic, chemical reactions which transform one compound containing tritium into other compounds. However, the net effect of the soil organic tritium transformations on tritium transport is probable very small compared to transport by water. In practice, the most important transformation of tritiated compounds taking place in soils is the oxidation of tritiated hydrogen to tritiated water (Watts and Murphy 1978, Ogram et al. 1990).

The residence time of tritium in soils is affected by all of the processes that move tritium in and out of the soil and the storage capacity of the soil for tritium. The storage capacity of the soil in the terrestrial environment, as defined above, is very dependent on the depth of the rooting zone. Given the same input of water from above and the same root uptake, the time necessary for tritium to leave the rooting zone will be proportional to the depth of the rooting zone.

Vegetation Tritium

Tritium transport between the atmosphere and vegetation is a diffusional process, usually called dry deposition. Dry deposition is the result of a number of processes acting in parallel and series which result in tritium being transferred from the atmosphere to the sinks for tritium at the surface. Deposition begins with turbulent diffusion in the fluid boundary layer near the surface as eddies of air carry the tritiated compounds from the atmosphere to the surface of the vegetation and the soil.

After the tritium has reached the surface of the vegetation, it must cross the fluid boundary layer of the surface elements. The waxy surfaces and bark of vegetation are relatively inert to tritiated compounds, such as hydrogen, water vapor, or methane. The only significant sinks for these compounds are inside the vegetation. The route into the vegetation is through the stoma on the leaf surfaces. The size of the stoma can change in response to the environment. Plant leaves have evolved to limit water loss while allowing transport of CO_2 to the leaf for photosynthesis. Under conditions of adequate moisture, the stoma will be open during the day and closed at night. If water stress develops in the leaf, the stoma will close to conserve water. The rate of diffusion of any of the tritiated gases will be controlled by the size of the stoma.

After diffusion through the stoma, the gas will go into solution in the cell sap in the leaves. The diffusion is in the direction of the gradient of the tritiated compound. Tritiated water vapor diffuses in response to its own gradient and it is possible for water to be evaporating from the leaves while tritiated water vapor is entering the leaves. Tritiated water vapor will mix with the water in the leaves and will participate in all the physical and physiological processes taking place in the vegetation. As the tritiated water concentration increases, the leaf water will come to a steady state with the atmosphere and net uptake will stop. Leaves can come into equilibrium with the atmosphere in from 15 minutes to more than 2 hours, depending on the uptake rate of tritiated water as controlled by the leaf stoma. The comparatively large initial deposition velocity for tritiated water to vegetated surfaces will decrease to nearly zero as the leaves of the vegetation approach equilibrium (Mason et al. 1973).

Tritiated methane and tritiated hydrogen will go into solution in the leaf cell sap (Cline 1953, Mason et al. 1973). Since the solubility of both of these gases is low, the cell sap will tend to saturate at low internal concentrations. The fact that tritiated water is detected in vegetation soon after exposure to either of these gases suggests that there are physiological processes which promote absorption and conversion. However, the rates are small compared to either the deposition rate of tritiated water to vegetation or the conversion rate of tritiated hydrogen to tritiated water in soil (Sweet and Murphy 1984, Spencer and Dunstall 1986, Murphy 1989).

As explained above, the other source of tritium in plants is from tritiated water taken up from the soil. This is a bulk flow process and the concentration entering the vegetation will be a weighted average of the tritium concentration of the soil layers from which water is being withdrawn. Most plant roots are within a few feet of the soil surface. Under conditions of equal soil moisture tension at all depths, most of the water will come from the surface layers of the soil. However, as water is removed from the surface layers, the majority of water will come from successively deeper in the soil. The driving force for movement of water from the soil into the plant is evaporation from the plant leaves (Kramer 1969).

The green leaves of vegetation are the primary site for incorporation of tritium into organic matter. This is accomplished through the process of photosynthesis, where carbon dioxide and water are used to synthesize sugar. The sugar is the building block for the synthesis of all other plant organic matter. Tritium incorporation is from the tritiated water in the leaf (Moses and Calvin 1959, Guenot and Belot 1984). The amount of tritium incorporated into organic matter is small compared to the amount of tritium moving through the plant from the soil to the atmosphere as tritiated water. The fraction of tritium converted to organic matter varies from 0.3 % to 0.06 % for a rapidly growing crops (Kramer 1969). The percentage is likely to be less by a factor of ten in many natural vegetation types. Therefore incorporation into organic matter has little effect on the amount of tritium moving through the environment. However, tritiated organics need to be considered when dealing with tritium concentrations in organisms feeding on the vegetation.

A number of other mechanisms for incorporation of tritium into the organic matter of vegetation have been demonstrated (Moses and Calvin 1959, Peng 1973). However, all of these mechanism are much less efficient in incorporating tritium than photosynthesis. Their effect on tritium transport is extremely small and the main interest in these processes has been to explain anomalies in data collected from vegetation exposed to tritium.

The mass of tritiated water in the leaves of vegetation ranges from a few grams to a few tens of grams. This is a small fraction of the water moving through the leaves in most vegetation types. The residence times are short enough, in the order of minutes, that the leaves approach a steady state concentration of tritiated water.

The steady state tritium concentration of water in the leaves of vegetation is the result of the processes by which tritiated water enters or leaves the vegetation (Raney and Vaadia 1957, Belot et al. 1979, Murphy 1984). Liquid water enters the leaves from the soil, by way of the roots and stems. Water leaves the plant by evaporation from the leaves. Tritiated water vapor can also enter the leaves from the atmosphere.

If the soil water contains tritiated water, the water will enter the roots and move to the leaves at about the same concentration as the water in the soil. When the tritiated water reaches the leaves it will evaporate along with the other water. The rate of evaporation of each type of water, tritiated or normal, depends on the difference in the concentration between the air and the leaf surface for that type of water. The relative rate of evaporation also depends on the fraction of the water in the leaf of each type of water. If the air has no tritiated water or other water vapor, that is, the relative humidity of the air is zero, then the evaporation of both types of water is the proportional to the fraction of each in the leaves, and the concentration in the leaf remains the same as that in the soil.

However, if the relative humidity is greater than zero, the difference in the water vapor concentration between the leaf and the air will be less than in the case with zero relative humidity. The evaporation of tritiated water will not change if there is still no tritiated water vapor in the air near the vegetation. In this case the evaporation of tritiated water is faster, relative to its concentration in the leaf water, than the evaporation of the other water. The higher evaporation of tritiated water will lower the tritiated water concentration in the leaf. The lower concentration will, in turn, lower the evaporation rate of tritiated water. This continues until a point is reached where the evaporation of tritiated water is equal to the amount of tritiated water being transported from the soil. The result is that the concentration in the leaf is generally lower than the concentration in the water of the source, that is the soil water.

The source of tritium in the vegetation can also be tritiated atmospheric water vapor. If the soil water contains no tritiated water, tritium will enter the leaves by diffusion from the air and exchange with the other water in the leaves. If the difference in the water vapor concentration between the atmosphere and the leaf is zero (100% relative humidity), the tritium will enter the leaf until the concentration in the leaf is approximately the same as in the atmospheric moisture. However, if there is a positive difference between the water vapor concentration in the leaves and the air, water will be evaporated from the leaves. When the water evaporates it will be replaced by soil water pulled up through the roots and the stem. This water will dilute the tritiated water entering from the atmosphere. In time, a steady state tritium concentration will become established in the leaf which will depend on the rate of entrance of tritiated water and evaporation of other water. The result is the tritium concentration of the leaf water will generally be less than that of the atmospheric source.

One would expect that both the atmospheric and soil water in the vicinity of a source of atmospheric tritium would contain tritium. In this case, the steady state tritiated water content would be between the concentration of the soil and air water. Environmental monitoring data collected at SRS during the period 1977 to 1982 show an average ratio of tritiated water in vegetation to tritiated water in the air of 0.77 (Murphy et al. 1986). This ratio compares well with the value of 0.82 calculated for the steady state leaf water to air water ratio, under the assumption that the soil water tritium has the same concentration as rain water tritium. This ratio of leaf tritiated water concentration to air tritiated water concentration was greater than 1.0 during 1976 when the concentration in the soil was elevated by tritium from oxidation of tritiated hydrogen in the soil following releases of this compound.

The water content of whole terrestrial plants varies from less than a gram to thousands of kilograms. Residence times in vegetation is related to both the mass of water in the vegetation and the flow of water through the vegetation. The mass of water, relative to the rate of flow of water, in forests or other large vegetation types is great enough that the turnover time for tritium in this type of vegetation is in the order of days (Jordan et al. 1970, Kline et al. 1970, Luvall and Murphy 1982). Water flow through plant organs is not uniform. Some organs, such as fruits, have slower water turnover than the root, stem, leaf conducting system leading to longer turnover times (Dinner et al. 1980). The same situation of comparatively large storage mass and low water flow would seem to be the case for succulent, arid-land vegetation such as cactus.

The amount of hydrogen in the organic matter of large vegetation can also be in the order of hundreds of kilograms. The turnover of this material is complicated by the many types of physical and physiologic processes that take place in the natural environment. Exchange processes take place in the order of hours or days, metabolic turnovers can be in the order of years, and decomposition of some organic compounds after death can be in the order of hundreds of years. While the amount of organic tritium moving through the terrestrial system is small compared to the amount of tritium associated with water, the longest retention times in these systems are likely to be associated with organic compounds (Stewart et al. 1972, Sanders 1976, Brown 1979).

Animal Tritium

Tritium concentration in animals is similar to that in vegetation in the sense that the concentration is the result of tritium transport to and from the animal through a number of paths (Robertson 1973 Yousef 1973). The majority of the water entering the animal is through drinking water. A major second source is the water in food eaten by the animal. The water in food is from the moisture in the food and from the water produced by digestion and catabolism (burning for energy) of the food. Water leaves the animal by excretion in urine and feces. Water can also enter or leave the animal in the breathing air and by absorption or excretion from the skin.

The major path by which water leaves the animal is through excretion. The relationship between the volume of water in the animal and the rate of excretion is the major determining factor for the turnover time of tritium in the body once the animal has been contaminated. Water turnover rates have been found to be closely related to body weight for a wide variety of animals (Figure 10, Yousef 1973). An exception to this trend is the dry-land kangaroo rats which have low water looses compared to their body weight. The water turnover time for most species is less than 10 days. This suggests that most

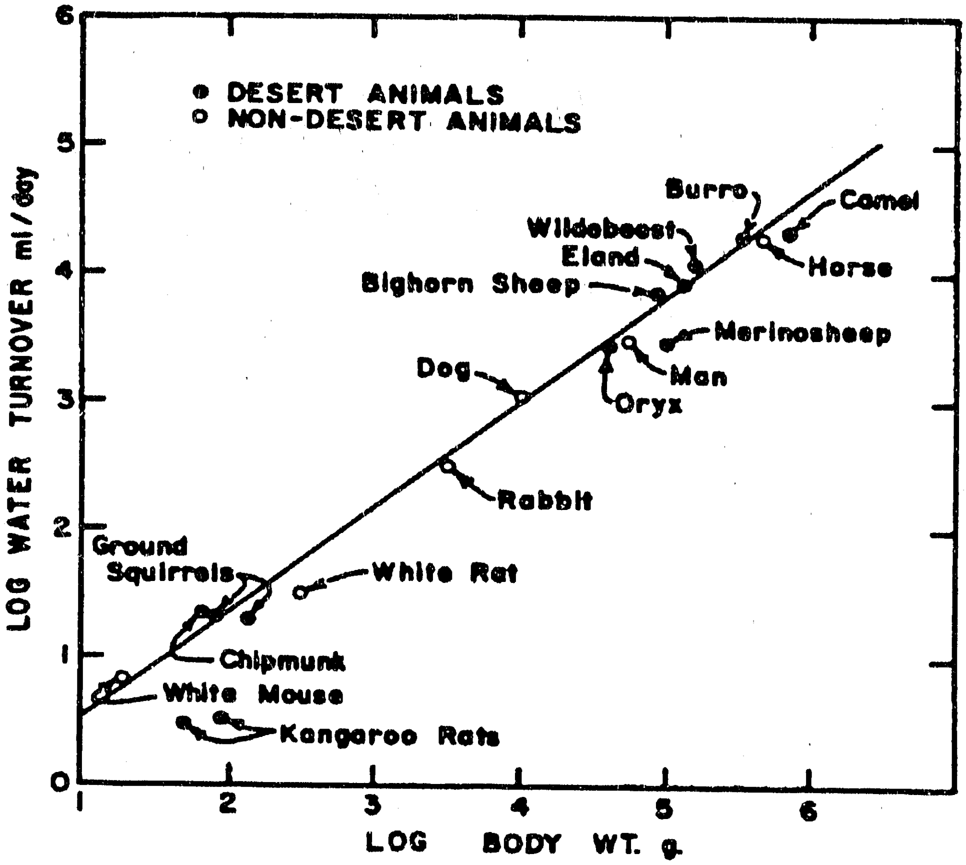

Figure 10. The water turnover in animals is closely related to the body weight except in a few desert rodents (from Yousef 1973).

animal species, like vegetation, will respond to the day to day changes of tritium in their environment. However, unlike vegetation, animals are mobile and will contain water that is averaged over the area in which they range.

The fate of tritiated organic material which is transported to animals in food chains depends on the chemical form of the compounds containing the tritium. The fraction of exchangeable sites varies with the compound. In addition, the metabolism of digestion and assimilation varies. Carbohydrates have comparatively large numbers of exchangeable sites and even complex carbohydrate are digested to basic units before assimilation. Proteins have more nonmobile sites but are metabolized to amino acids before being used for assimilation. Lipids are either completely metabolized as energy or assimilated with few changes. This is reflected in the studies of van den Hoek et al. (1985) which show that when milk is produced from tritiated feed, fats retain better than 90% of the tritium when assimilated into the consuming animal. In contrast, proteins retain around 50% and carbohydrates around 25%. Since the hydrogen content of the non-aqueous, animal matter is in the range of 40% of the size of the aqueous pool, the tritium content of the non-aqueous matter is a significant portion of the total tritium content of an animal under equilibrium conditions (Evans 1969).

Residence times for tritium in the organic fraction of animals has not been widely studied. Studies of humans (Sanders and Reinig 1969), marine animals (Harrison et al. 1973), and milk cows (van den Hoek et al. 1985) exposed to tritiated water indicate that there is one or more "slow" turnover components, usually attributed to organic tritium, that are in the order of 100 days to 1 year.

Tritium Concentrations and Residence Times in Terrestrial Systems

Terrestrial systems are most likely to be exposed to tritium by exposure to tritium from the atmosphere or tritium introduced into the soil. The initial concentration in the system is determined by the rate of uptake, the time over which the uptake takes place, and the mass of water diluting the uptake. Uptake of tritiated water from the atmosphere will be proportional to the concentration of tritium in the atmospheric water. Given the same amount of tritiated water per volume of air, if the atmospheric humidity is high there will be more water to dilute the tritiated water and the concentration will be lower than if the humidity is low. Uptake by vegetation will also be affected by the size of the stoma limiting diffusion into the leaves. Vegetative uptake will be lower at night and for water-stressed plants at all periods of the day. Since water stress is more common in dry terrestrial ecosystems, vegetative uptake should, on average, be less in these systems.

Given the same uptake, a system with more water, i.e., moister soil, more moist vegetation, or more succulent plants, will have a lower initial concentration. The initial concentration must be calculated by determining the concentration in the atmosphere from the dispersion and atmospheric conditions. The initial concentration in the terrestrial system must be determined by integrating the uptake by each of the processes described above (wet and dry deposition of all important compounds) and calculating the dilution by the known amount of water in the system. Figure 11 shows that tritium concentration in vegetation at 25 km from the release point after short duration tritium releases at SRS were proportional to the time-integrated tritium exposure from atmospheric tritium (Murphy et al.

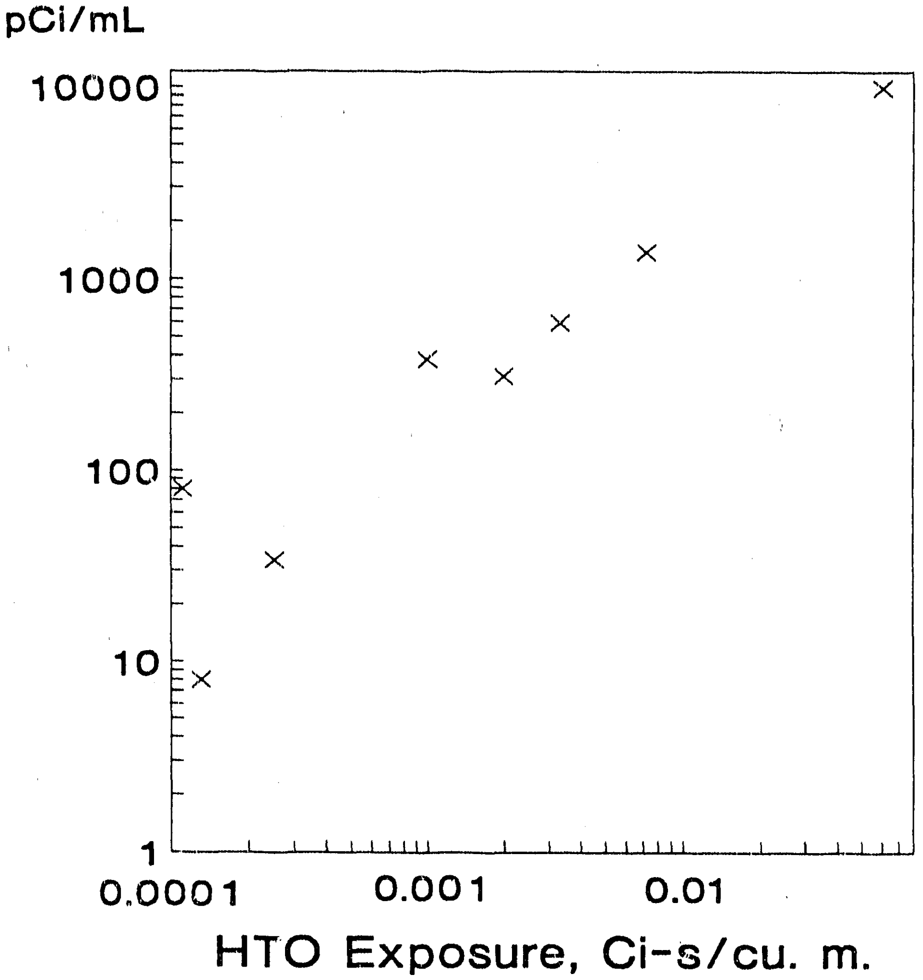

Figure 11. The relationship of exposure to tritiated water vapor in an atmospheric release plume and the tritium measured in vegetation immediately after the release (Murphy et al. 1990).

1990). A similar integration of tritium exposure and dilution by system water must be made for direct releases of tritiated water to the soil.

It is a general property of systems made up of a number of connected components that the residence time of the system will be determined by the longest residence time among the components (Sasser et al. 1973). However, in order to make use of this principle one must be very careful to identify the components of the system that are important to the transport of interest. The determination of importance is subjective. If the interest is in where the majority of tritium is going after release into the system, then an inspection of the relative transfer rates can be helpful. Under unstable atmospheric conditions, less than 10% of the tritiated water released from an elevated tritium source (a stack) will be deposited within 25 km of the site of release. Less than 0.3% will enter into the vegetation organic fraction. If 0.03% of the total released is a trivial amount of tritium, there is no reason to include the organic components in the determination of the system residence time. Some other examples will be discussed.

Sasser et al. (1973) present data from a series of terrestrial systems exposed through irrigation with tritiated water. The results (Figure 12) show that the residence time of the systems, as measured by the residence time in the vegetation water, was determined by the residence time in the soil. In these systems , after initial exposure, the other system components were supplied tritiated water from the soil water. The short residence time in the grassland compared to the desert is a result of the greater amount of rainfall moving tritium out of the rooting zone of soil of the grassland. The residence of the tritiated water in the tropical rain forest is controlled by a soil layer of low permeability which retards drainage of water from the soil. Miettinen (1979) has presented a summary of soil tritiated water residence times measured in the IAEA coordinated program. The half residence times vary from a few hours to 14 days for the shortest component of tritium loss. Longer components can be as long as several years.

A somewhat different result was found after exposure of forests to tritiated water vapor following an atmospheric release. The short duration release resulted in tritium entering the vegetation but not the soil (Sweet et al. 1983). The residence time of all measurable tritium in the system was less than one day. On the other hand, after a release of tritiated hydrogen most of the tritium remaining the system was in the form of tritiated water, oxidized from tritiated hydrogen entering the soil. The residence time in the vegetation after the tritiated hydrogen release was found to be on the order of 10 days, which is the residence time in the soil (Murphy et al. 1977).

Tritium concentrations in system components resulting from chronic releases to terrestrial systems will be determined by the balance of input and output tritium transport regardless of the capacity of the component (Murphy 1986). For systems exposed to atmospheric tritiated water vapor, the air concentration will depend on the atmospheric condition and the release rate. Soil water will tend to be lower than the atmospheric water vapor because of dilution by rainfall. The higher the average rainfall rate, the greater the dilution. The vegetation will have a tritium concentration between the soil water and atmospheric tritium concentration (Murphy 1984). The vegetation organic concentrations will be close to the vegetation water which is the source of water used in photosynthesis (Guenot and Belot 1984). The animals living in this system will have a tritium concentration in their body water that is a balance between intake by drinking water and vapor absorption, with the drinking water being most important.

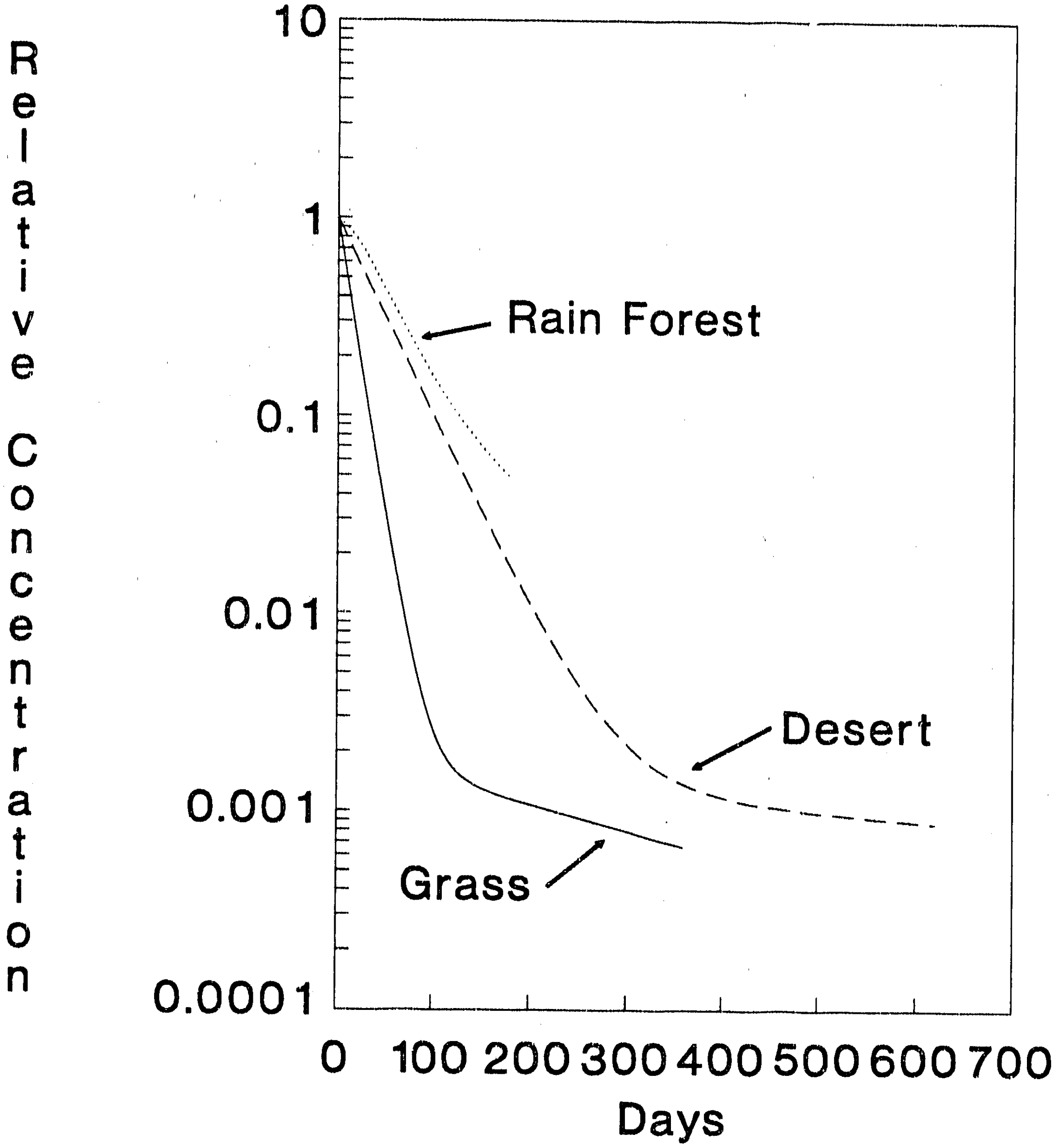

Figure 12. A comparison of the response of three terrestrial systems to tritiated water additions to the soil. The concentration of the vegetation is controlled by the response time of the soil (redrawn from Sasser et al. 1973).

The concentration of tritium in the organic material of the animals will reflect the concentration of the food and the body water. Monitoring data taken at SRS suggest that the average tritium concentration in farm animals and plants grown near the facility is very similar to the average value of measured tritiated water vapor concentrations in the air in the same locality (Figures 13 and 14).

A number of studies have reported concentrations of tritium in the organic fraction of vegetation or vegetable food that exceed the concentration of the water in the same vegetation (Stewart et al. 1972 , Murphy and Pendergast 1979, Belloni et al. 1984, and Belot et al 1986). In some cases these studies have been used to suggest that tritium will accumulate in vegetation or be actively concentrated. However, this has not been observed in carefully controlled experiments where plants grown under equilibrium with tritiated water have slightly lower concentration than the water (Garland and Ameen 1979). In addition, the tritium concentration of the organic fraction of plants grown in areas far removed from tritium sources are nearly in equilibrium with the water in their environment (Hisamatsu et al. 1989). The latter results suggest that the elevated tritium levels are either from the slower turnover of organic matter than water following higher tritium exposure in the past or by uptake of tritium from another sources which is at high concentrations (Murphy and Pendergast 1979) rather than from bioaccumulation.

TRITIUM IN THE AQUATIC ENVIRONMENT

In this review, the aquatic environment includes streams, ponds, and lakes. The marine environment will be addressed in a separate section. Sources of tritium into the aquatic environment can be by direct discharges or through transport of tritium discharged into some other component of the environment. Tritium concentrations and residence times in these systems are functions of the mass of water involve and the processes that transport tritium through the system.

Streams

Tritium can enter a stream system through direct discharge of tritiated compounds into the stream, through exchange with the atmosphere through rainfall or gas deposition, or through input from the groundwater system. In a natural stream system, the majority of the water in the stream is derived from rainfall that has entered the soil and passed through the groundwater system. Second in importance to stream flow is water that has moved laterally through the soil without reaching the water table or has run off the surface of the ground during storm events. In some watersheds, the majority of water reaching the streams may be from the soil water system above the water table. One would expect the tritiated water concentration of streams near an atmospheric tritium source, but not receiving direct tritium discharges, to have a tritium concentration similar to the concentration in the precipitation, appropriately averaged in space and time over the watershed. However, some of the water moving through the groundwater system may have transport times in the order of several tritium half-lives. .In this case, some of the stream water will be relatively depleted in tritium. This will tend to decrease the stream water tritium concentration relative to the precipitation average. Figure 15 shows the relationship between the tritium concentration in stream water and atmospheric releases in a stream at the SRS which received no direct tritium releases through the period of measurement (Murphy et al. 1990).

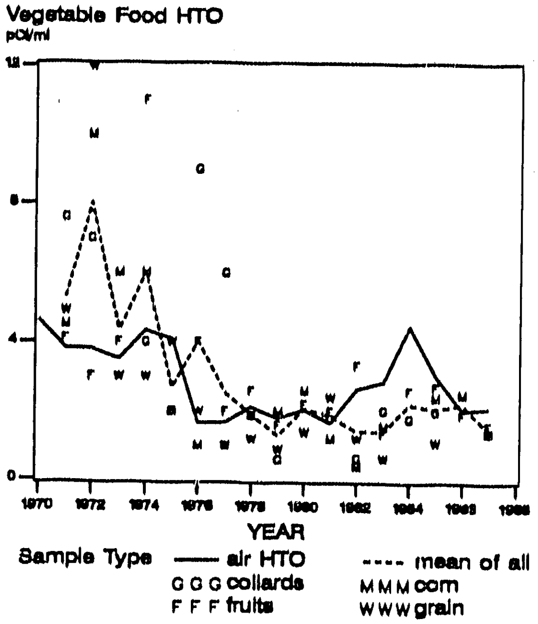

Figure 13. Tritium concentration in the water of selected vegetable food grown in the vicinity of SRS. The annual average of all foods is compared to the annual average air moisture concentration at an arc 40 kilometers from the center of SRS (Murphy et al. 1990).

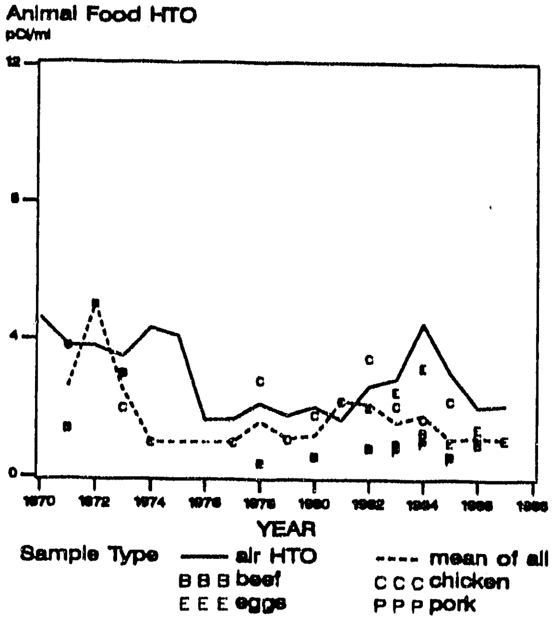

Figure 14. The tritiated water concentration of animal foods collected in the vicintity of SRS compared to the annual average tritium concentration of air moisture (Murphy et al. 1990).

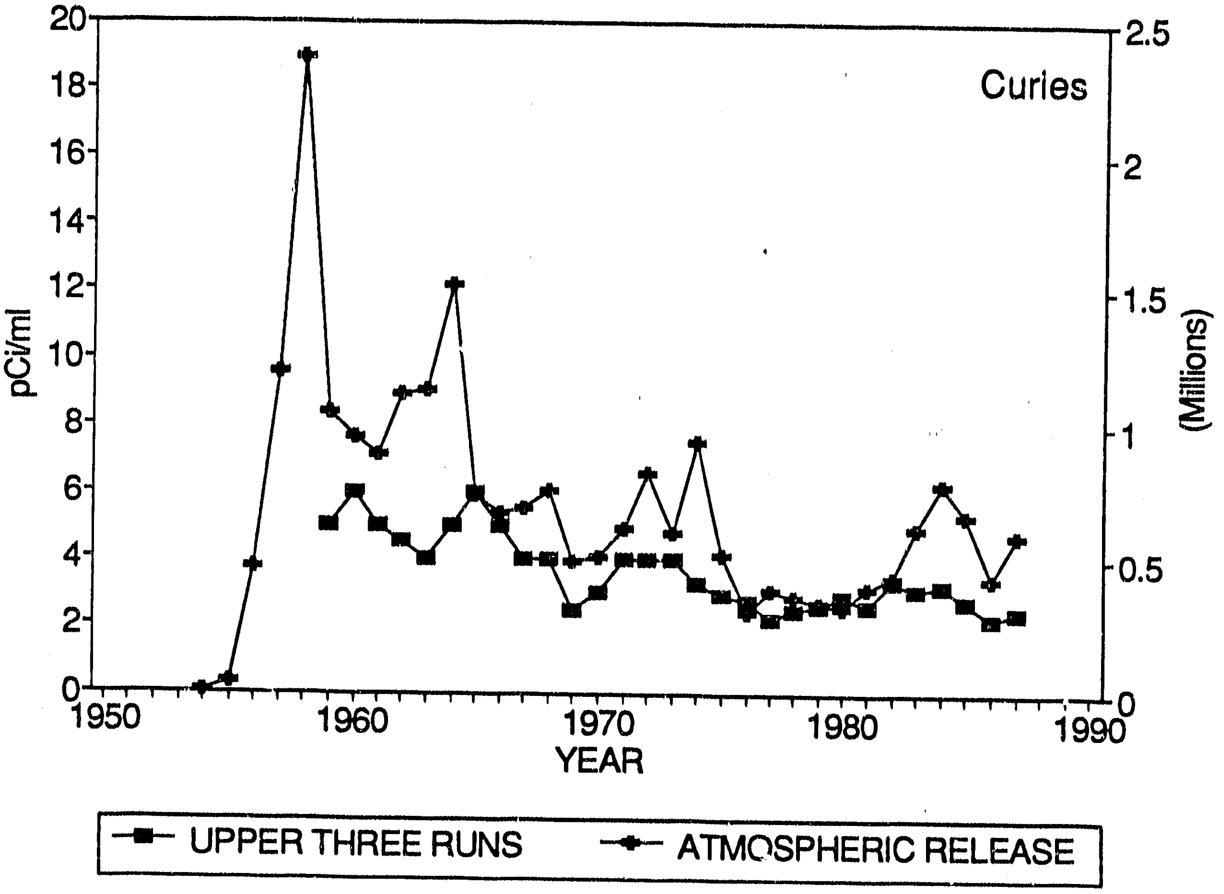

Figure 15. Tritium concentrations measured in Upper Three Runs Creek compared to atmospheric releases from SRS facilities (Murphy et al. 1990).

The transport and concentration of tritium as tritiated water in streams is determined by the processes of volumetric dilution, advection, dispersion, and turbulent diffusion (Thibodeaux 1979). Volumetric dilution of tritiated water occurs when there is an increase in water flow volume in the stream from an addition of water with a low tritium concentration. This can happen from addition of uncontaminated groundwater or addition by combination with another stream. Volumetric dilution is the rule in streams flowing away from a tritium source in humid climates. In arid climates the volume of water may actually decrease by evaporation. In this case, the tritium concentration should remain constant or decrease somewhat because of differential evaporation. Evaporation is discussed in the section on ponds and lakes.

Advection, as discussed previously with respect to the atmosphere, is the movement of tritiated water with the bulk fluid flow. Tritiated water will be carried along at the same general speed and direction of the bulk water flow. The concentration of tritium in the water is not changed by advection.

Dispersion is the mixing and dilution of the tritiated water resulting from parts of a stream traveling different paths at different speeds. This can be from the slowing of water near the edges, bottom, and surface of the stream relative to the water at the center of flow or it can result from streams with multiple channels which move at different speeds.

Turbulent diffusion in surface waters is the process in which mixing and dilution of tritiated water takes place through the action of eddy water currents, similar to those already discussed in atmospheric transport. Turbulent diffusion is responsible for the initial mixing and dilution of tritium discharged directly into a stream or when two streams combine. Dispersion and turbulent diffusion contribute to the mixing of tritiated water with water at the leading and following edges of releases. These process are important in the initial dilution of short period releases and are less important as the width of a peak increases. These processes are generally not very important in the dilution of releases that are longer than a day.

The combined effects of volumetric dilution, dispersion, and turbulent mixing are illustrated in Figure 16. The measurements were made at two locations on the Savannah River following an accidental spill of heavy water contaminated with 4,200 curies of tritiated water. The peak tritium concentration of 280 pCi/ml had decreased to 120 pCi/ml in 87 miles of river transit (Buckner and Hayes 1975).

Ponds and Lakes

For the purpose of this review, ponds are defined as small water bodies with small throughflow of water compared to water volume. Lakes are larger bodies of water which have a range of volume to flow characteristics. Some lakes undergo a characteristic seasonal cycle of stratification.

Mixing in ponds and lakes is through the action of wind on the pond surface or, if the flow through the lake is large compared to the volume, by stream-like advection (Hutchison 1957). Complete mixing of tritiated water added to the pond usually takes place in a matter of days while mixing may take longer for some lakes. The tritiated water concentration is determined by the balance of tritium entering the pond through direct discharge, precipitation, etc. and leaving the pond through evaporation, seepage

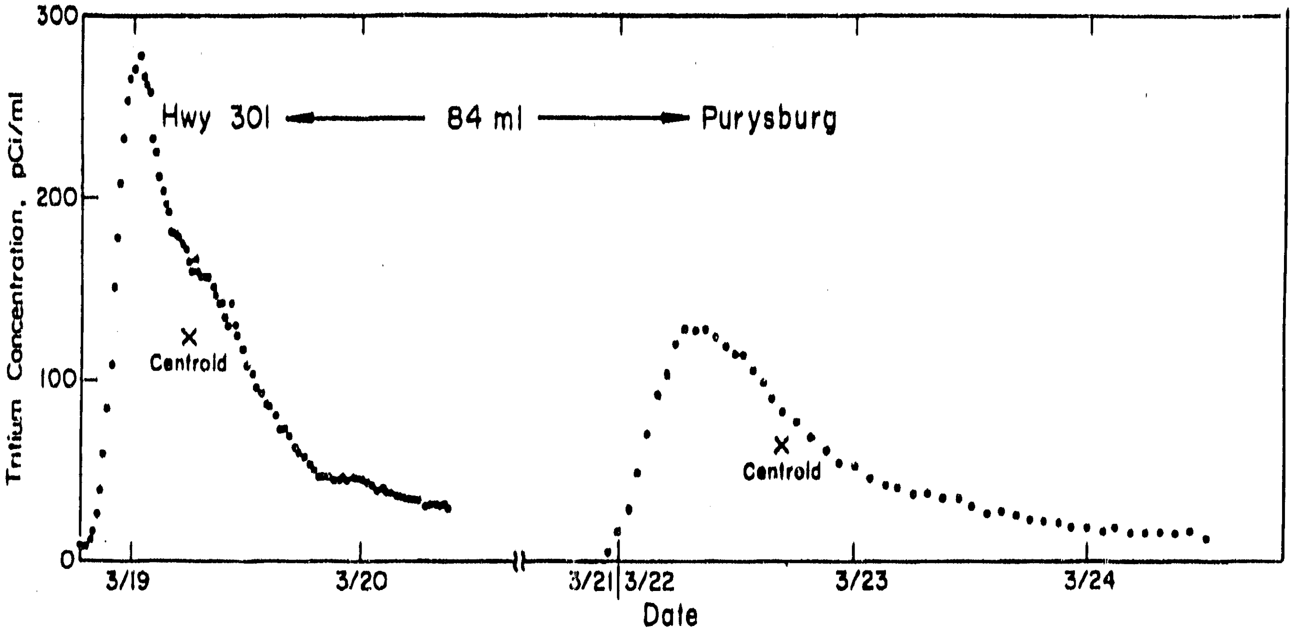

Figure 16. Dispersion of tritium in the Savannah River following a release through Beaver Dam Creek (Buckner and Hayes 1975).

into the soil under the pond or ponds, and flow from the outlet. If the addition of tritium has stopped, the residence time will be determined by the output processes and the amount of water in the pond.

Stratification of lakes has the effect of decreasing the transport between layers of water within the lake. This may exclude part of the lake from tritium released into the lake. It may also restrict tritium transport out of the lake by some processes (Thibodeaux 1979). Stratification is caused by heating of the surface water of the lake. The warm surface water (epilimnion) is separated from the cooler deep water (hypolimnion) by a layer of water having a large decrease in temperature with increasing depth (thermocline). Turbulent mixing will be damped by the density difference between the two layers. In many temperate region lakes, stratification takes place in the summer. The epilimnion and hypolimnion are mixed during the winter. Stratification develops during the spring and is abruptly broken during the fall. This leads to one or more periods of sudden mixing of the upper and lower layers. Calculation of tritium concentrations in stratified lakes requires a knowledge of the mixed volume into which the tritium is released. Releases below the thermocline will lead to longer residence times than calculated on the basis of lake throughflow and volume. Similarly, the residence times for surface releases will be shorter. This situation will change in some reservoirs in which water is released from the below the thermocline.

The atmospheric transport from the pond or lake will be proportional to the tritiated water content of the water as limited by the mass transfer into the atmosphere. While the mass transfer processes into the atmosphere are almost identical to those for evaporation of water, the transfer of tritiated water will be proportional to the concentration difference between the pond or lake and the atmosphere and not proportional to evaporation. As in the transfer of tritiated water between the atmosphere and vegetation, tritiated water may be transferred to the atmosphere when no evaporation is taking place.

Measurements of tritium transport to the atmosphere from a small pond (used as a seepage basin before plugging of the soil) at SRS indicate that the half-residence time of tritium was about 1.0 year (Horton et al. 1971). The equation presented by Horton et al. can be adjusted to estimate tritium losses from ponds with different surface area to volume ratios. Evaporation and tritiated water vapor transport vary with the size of the lake. For large lakes, the transfer coefficients used by Horton et al. should be replaced by those from evaporation studies of a comparable lake , i.e., Lake Hefner for large reservoirs (U.S. Geologic Survey 1952).

Tritium in Aquatic Organisms

Since aquatic systems are almost totally immersed in water, the transport of water into aquatic organisms is very rapid. The turnover time for tritiated water in the tissues of aquatic organisms is in the order of a few minutes to a few hours (Harrison et al. 1973, Morgan et al. 1973, and Blayblock et al. 1986). Therefore, the tissue water concentration is the same as that of the surrounding environment for all conditions other than releases lasting less than a few hours. The exception is aquatic macrophytes which are often only partially immersed and characteristically have lower tritium concentrations than other aquatic vegetation (Blaylock and Frank 1979).

As discussed for terrestrial systems, a major source of organic tritium in aquatic systems is the incorporation of tritium from tritiated water by photosynthesis. In addition, some aquatic systems have the potential for receiving significant amounts of organic tritium from detritus of either aquatic or terrestrial origin. Algae and other aquatic plants are the source of photosynthetically fixed tritium. As in terrestrial plants, under carefully controlled laboratory conditions the tritium to hydrogen ratio in the organic material of aquatic plants grown in tritiated water is near 0.8 (Kirchmann et al 1979). However, algae can incorporate organic material from the environment as well as make it by photosynthesis. In this case, the tritium in the organic material of the algae will be an average of the two sources. If the tritiated organics have higher concentrations of tritium than the water, the organic material of the algae will also have higher tritium concentrations than the water. This has been shown to be the reason for the relatively high tritium concentration found in small effluent streams containing waste water with reactor water purification resins (Kirchmann et al. 1986b).

The transfer of tritium from photosynthetic or detritus inputs through the food chain to consumer organisms depends on the nature of the organic material consumed (Bruner 1973 and Blaylock et al. 1986). Aquatic consumer organisms raised in tritiated water but fed untritiated food have tritium to hydrogen ratios that are from 10 to 50% of the ratios in the water. Some of the tritium may be the result of isotopic exchange and some may be from the consumption of food inadvertently grown in the water. When consumers are grown in tritium free water but fed tritiated food, the tritium-hydrogen ratio of the consumer will approach 50% of that in the food. This suggests that approximately 50% of the hydrogen sites in these organisms are readily exchanged with the water and the remaining sites are not easily exchanged once incorporated in the organism. Few experiments have been run long enough or controlled well enough to ensure that all the organisms in a particular aquatic system are in equilibrium with the environment; however, under conditions which most closely approach this ideal situation, the tritium to hydrogen ratios in the organisms of these systems generally range from 80 to 130% of the ratio in the environment. None of these systems, either laboratory or natural, show any sign of bioaccumulation of tritium in the food chains.

Studies of the release of tritium from aquatic organisms after exposure to tritium show that the decrease in tritiated water content is very rapid, with a turnover time of less than a few hours for most of the tritium released (Harrison 1973, Morgan 1973). Smaller amounts of tritium, usually less than 5% of the total, are released more slowly with turnover times of 100 to 300 days. It is usually assumed that the smaller components of release are from the turnover of tritiated organic compounds. It is likely that at least some small component of tritium is released even more slowly, perhaps being retained until the death of the organism.

Tritium Concentrations and Residence Times in Aquatic Systems

In stream systems, the dominant factor affecting the residence time is the duration of the release of tritium to the system. The turnover time of water in most streams is very short relative to the water or organic turnover of the organisms living in the streams. If the release is of short duration, i.e., less than an hour, the water in many organisms will still approach equilibrium and then drop quickly to the background concentrations. The organic fraction of the the organisms will not incorporate very much tritium under these conditions. If the release period is longer than a few hours, the water of most aquatic organism will reach equilibrium. If the release period is longer than a year, most of the organic matter of living organisms will approach equilibrium with the water. However, the organic matter in sediments has even lower turnover rates. Bogen and Welford (1976) suggested that the high tritium concentration measured in Hudson River sediments could be explained by the relatively high tritium concentrations in the environment when the organic matter in the sediments was synthesized immediatly following the introduction of weapons testing tritium into the environment and the long turnover time of tritium associated with the organic residue in the sediments.

As discussed previously, the residence time of tritium in lakes is determined by the balance of input and outflows relative to the volume of water. The residence time of tritiated water in lake systems is generally determined by the physical residence time. The residence time of tritiated organic components can be longer than that of the tritiated water. In this case, a relatively small amount of tritium can remain in the lake system for extended periods of time. Figure 17 shows the concentration of tritiated water in crayfish living in a natural aquatic ecosystem which received a single release of tritiated water. The water concentration in the animal muscle followed the water concentration in the environment while the organic matter concentration lags behind the concentration in the water (Adams et al. 1979).

In most cases when an aquatic system has reached a steady state with its tritium input, the concentration of tritium in all components of the aquatic system will be in equilibrium with the tritium concentration of the water. The exception would be systems which have high inputs of organic matter or high inputs of tritium in organic matter (low amounts of highly labelled organic material). In these systems the steady state tritium concentration in organisms would be intermediate to the concentrations in the organic matter and the water (Kirchmann et al. 1986b). In no case is there evidence of bioaccumulation of tritium in the food chain.

TRITIUM IN THE GROUNDWATER ENVIRONMENT

In this section, the groundwater system is defined as the ground, or earth, beginning below the rooting depth of the vegetation and extending down to bedrock. In some cases it is not necessary to consider the deepest parts of the groundwater system which do not exchange water with the surface. The movement of water through the ground below the rooting depth of the vegetation is driven by the forces of capillary tension, gravity, and hydraulic pressure. The movement is impeded by friction as the water moves through the soil or rock matrix. The rate and direction of water movement is strongly influenced by the characteristics of the matrix which determine the hydraulic conductivity (de Wiest 1965). Hydraulic conductivity is a measure of the ability of a material to transmit water.

In the absence of layers which restrict water movement, the general direction of flow will be downward from the upper boundary of the surface. When a retarding layer is encountered, the water will saturate the pore space in the soil or rock above the retarding layer. The upper surface of this saturated zone will be connected to the atmosphere by the unsaturated pore space above it and the upper surface of the saturated zone will be at atmospheric pressure. This unconfined, saturated surface is called the water table. Water in the pores above the water table is held in the pore space by capillary force and is

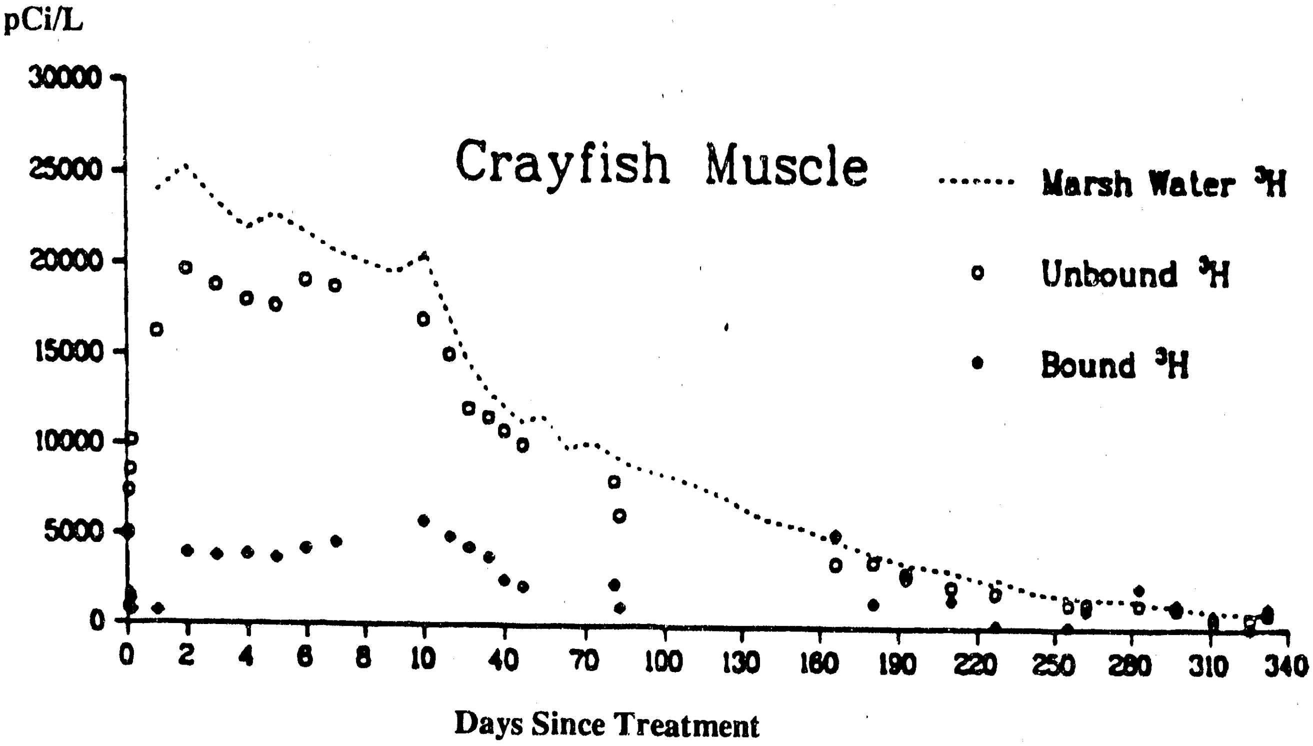

Figure 17. An example of an aquatic organisms response to a single addition of tritium to a lake system (Adams et al. 1979).

under tension with respect to the pressure at the surface of the water table. Water in the water table is under hydraulic pressure from the weight of the water above it (de Wiest 1965).

If the impeding layer producing the water table is inclined from the horizontal and/or the impeding layer reaches the surface (e.g., through the down cutting of a stream) the water table will also be inclined, leading to differences in the hydraulic head (the sum of the pressure and elevation geopotential) in the direction of the inclination. The water in the water table will flow, both vertically and horizontally, from areas of high hydraulic head to areas of low hydraulic head.

In many areas, the region below the water table will be made up of a succession of layers of material that conduct water (aquifers) or retard water flow (aquitards). Aquifers can be saturated or unsaturated with water but are often saturated. Some of the water in an aquifer may come from leakage through an aquitard. In many cases, the water enters the aquifer at some point where there are no confining layers between it and the surface. In this case, the geologic layer containing the aquifer is part of the water table and water enters directly by flow from the surface. These locations are often referred to as the source areas for the aquifer. Aquifers can also be fed by source areas under rivers or lakes.

Almost all of the tritium entering the groundwater system is in the form of tritiated water. Tritiated water in the groundwater system is transported from areas of high hydraulic head to areas of low hydraulic head. As in the atmosphere and stream systems, this type of passive movement within the groundwater flow system is referred to as advection (or convection). Other mechanisms also influence the movement of tritium through the flow system. Hydrodynamic dispersion is a combination of mechanical mixing and molecular diffusion. These two dispersive processes cause a spreading of the tritium in the groundwater over a larger area than the advection alone would produce, resulting in a dilution of the tritium concentration away from the source area. The mechanical mixing results from velocity differences produced by the length and flow velocity in different paths the water travels in the groundwater system. Molecular diffusion also occurs in groundwater but is usually much less dominant than the effect of mechanical mixing.

Most of the tritiated water in the saturated zone is associated with the bulk water in the pore space and moves as a nonreactive, nonabsorbed constituent within the groundwater system. Under some conditions, tritium in the groundwater can become associated with water of hydration on mineral surfaces. The water of hydration has a number of components varying in their mobility in the groundwater. At ambient temperatures, little tritium is associated with water of hydration. Incorporation at higher temperatures is associated with underground detonation of nuclear explosives which can produce tightly bound tritium (Stewart 1973). The tightly held component of the water of hydration has little significance to tritium movement in most groundwater systems. However, Koranda (1965) found relatively high tritium concentrations in bound water of hydration in the soils of islands in the Pacific Proving Grounds where the tritium had become fixed under the high temperatures produced by the thermonuclear detonations.

The velocity of groundwater moving through aquifers varies through a large range, normally from 1.5 meters/year to 1.5 meters/day. In highly permeable aquifers, the velocity can be as high as several hundred meters/day. With this wide range of permeabilities, which are often exponentially distributed in a given groundwater system, it is difficult to forecast the dispersion or residence time of tritium in a particular groundwater system without knowledge of the characteristics of that system (de Wiest 1965). Some examples of tritium transport in groundwater systems will be given to illustrate how groundwater analysis can be used to determine tritium transport.

Tritium Transport at the Savannah River Site Low-Level Waste Burial Ground

The Coastal Plain groundwater system at the SRS is very complex. The geology indicates a past environment where many interbedded clay, silt, and sand layers were deposited in an intricate three-dimensional flow system. Upon detailed inspection, however, the groundwater system can be divided into six main units, which are composed of several separate geologic layers (Siple, G.E. 1967 and Langley and Marter 1973).

At the base of the groundwater system is a dense clay aquitard. Overlying this layer is the region of loosely packed sediments laid down during the Cretaceous period. The Cretaceous unit is an excellent aquifer that can sustain yields of 1,000 gallons per minute (gpm).

A 100-foot layer of thick silty clay material forms a leaky aquitard unit above the Cretaceous aquifer. Overlying this aquitard is a tertiary-age aquifer. This unit is approximately 100 feet thick and consists predominantly of fine to medium and medium to coarse, well sorted sands. The Tertiary aquifer is not nearly as prolific as the Cretaceous aquifer below it, but it can sustain yields up to 100 gpm in water supply wells.

Overlying the Tertiary aquifer is a depositional layer which acts as a leaky aquitard unit about 50 feet thick. Overlying this aquitard is the water table unit which extends to the surface. The water table unit is not an aquifer by strict definition because of its low water-producing capabilities. Several wells in the water table at SRS cannot produce 1 gpm for a substantial period of time. Nevertheless, it is a distinct and important groundwater unit because any aquifer contamination from tritium or other materials entering at the surface must flow through the water table unit before entering other groundwater units or discharging to surface waters. The thickness of the water table varies greatly across the site due to the changing topography. It can be as low as zero where eroded away by a stream or as deep as 200 feet.

The Low-Level Radioactive Waste Burial Ground is a solid radioactive waste storage site centrally located at the Savannah River Site (Jaegge 1987). The Burial Ground used to store all radioactive solid waste processed at the site, as well as periodic shipments from other U.S. Department of Energy facilities. In 1988, it was estimated that 4,200,000 Ci of tritium had been interred in the Burial Ground since the beginning of SRS operations. After decay it is estimated that 1,500,000 Ci remain which are leachable and could get to the groundwater system. Tritium concentrations averaged over all grid monitoring wells in the Burial Ground suggest that greater than 40,000 Ci are accounted for in the groundwater system. Some of the remaining tritium still is associated with the waste and some is in the groundwater system under active parts of the Burial Ground that cannot be sampled at present.

The Burial Ground is located in an interstream area between two tributaries of the Savannah River: Upper Three Runs Creek to the north and Four Mile Creek to the south. The sedimentary layers below the Burial Ground are saturated with groundwater beginning at a depth of approximately 12 m. Part of this groundwater flows to Four Mile Creek and part to Upper Three Runs Creek. Measurement in cluster of wells show that the hydraulic head in sediments in the Tertiary aquifer is lower than head both above and below. Thus, water flows to the Tertiary aquifer from both above and below, limiting the depth of circulation of water from the Burial Ground. Of particular hydrogeolgic significance are the confining beds. These aquitards retard the vertical movement of the groundwater beneath the Burial Ground.

The difference in elevation between Upper Three Runs and Four Mike Creeks is of importance in the flow of groundwater. The bed of Upper Three Runs has eroded approximately 16.8 meters deeper than that of Four Mile Creek at the Burial Ground area. This difference in streambed elevation has displaced the water table divide about 300 meters toward Upper Three Run Creek. The water table gradient from the southern edge of the Burial Ground to Four Mile Creek is 0.01 m/m and that from the northern boundary of the Burial Ground to Upper Three Runs is 0.02 m/m. The gradient in the Tertiary aquifer from the Burial Ground to Upper Three Runs is approximately 0.003 m/m.

Though sediments in the Burial Ground area are highly heterogenous, flow over a distance apparently tends to reduce the effects of this variability. Sixteen groundwater velocity tritium tracer tests were made throughout the area over the years. Simultaneously, water table gradients were observed. A least square linear regression analysis of the data shows a strong correlation between water table gradient and groundwater velocity. The average velocity varies at the rate of 14.5 m/yr/1% gradient, with a correlation coefficient of 0.988.

Water flowing through porous media such as the Burial Ground sediments exhibits a distribution of velocities caused by the variability of the hydraulic conductivity. As a result, a tracer released into the system tends to assume a normal distribution in the longitudinal direction as flow proceeds. The leading edge of the distribution will precede the centroid by some multiple of the centroid depending on the distribution of hydraulic conductivities.

Results of a flow experiment in the southwest corner of the Burial Ground are shown in figure 18. Three tritium sources (residues of irradiated lithium-aluminum targets after the thermal extraction of tritium) were buried in 1957 and observed at intervals until 1970. The centroid was 76 m downgradient, but the leading edge had not arrived at detection wells 80 m beyond the centroid. Therefore the conservative estimate of the displacement of the leading edge is twice the rate of average displacement.

Applying these rates to observed water table gradients on flow paths originating in the east, middle, and southwest parts of the Burial Ground produces travel times for the leading edge of the plume of 11, 30, and 50 years for tritium to outcrop into Four Mile Creek. The average velocities for three flow paths are 12 m/yr, 18 m/yr, and 24 m/yr. These velocities are consistent with the detailed horizontal (and vertical velocities) that result from calibrated groundwater models. Extrapolation of these results by a computer models indicate a peak concentration of tritium equal to 6.71 pCi/L entering the Savannah River from the Burial Ground in the year 2085 (Jaegge 1987).

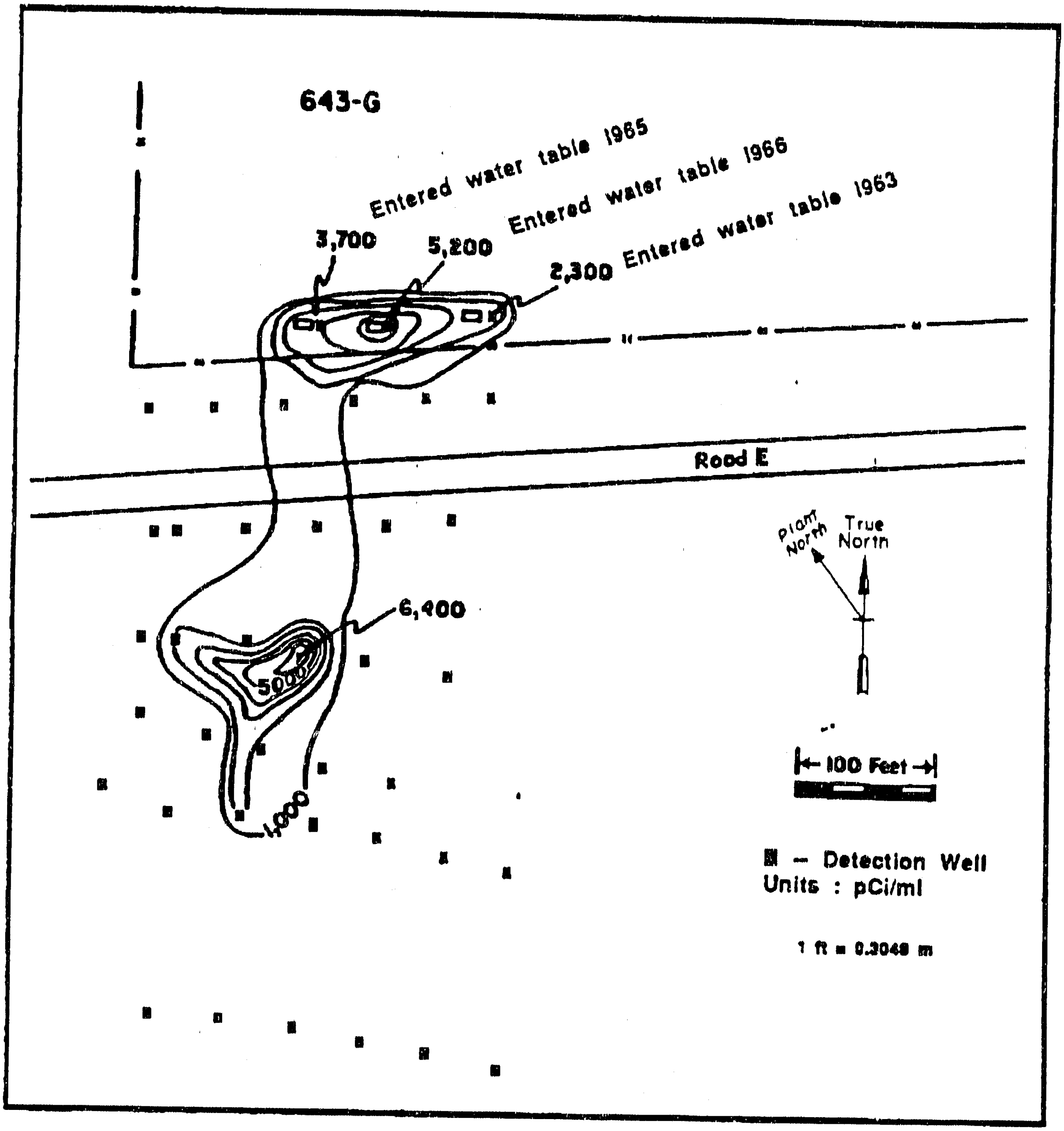

Figure 18. Tritium from spent melt test in groundwater at the SRS Low-Level Waste Burial Ground measured in October 1970.

Tritium in the Groundwater System at the Idaho National Engineering Laboratory

The Idaho National Engineering Laboratory site covers about 890 square miles. It is located on the eastern part of the semiarid Snake River Plain. The eastern Snake River Plain is a 12,000 square mile basin filled by a thick, interbedded sequence of basalt lava flows and sediments. The depth of the basins is not known but is estimated to be as much as 5,000 ft. The Plain is underlain by a large body of groundwater known as the Snake River Plain aquifer. The groundwater is contained within the cracks in the basalt or in the pore space of the courser sedimentary layers. Perched water tables are found in areas where the comparatively low permeability of some of the layers retards the downward movement of water. Natural perched water tables are associated with water entering the ground from the flow of streams which flow out into the Plain.

Aqueous wastes containing tritium have been discharged to shallow ponds and wells since 1952. The discharges from the ponds have produced perched water tables above retarding basalt and sedimentary layers under the ponds. The boundaries of the perched water tables, as defined by the tritium concentrations, have changed little between 1962 and 1970, suggesting that a steady state had been reached. Tritium outflow from the perched water tables has entered the Snake River Plain aquifer under the area.

Since 1953, low levels of tritium have been released directly into the Snake River aquifer from the Idaho Chemical Processing Plant disposal well. By 1978, a well defined tritium plume covered an area of 28 sq mi south of the well. The leading edge of the plume had travelled 7.5 miles from the source (Lewis and Goldstein. 1982). The velocity of the leading edge of the plume is about 1.5 m/day. This compares to velocities of 2 to 3 m/day measured in wells near the source (Hawkins and Schmalz 1965, Robertson et al. 1970). Groundwater velocities are much higher in the volcanic sediments of the INEL groundwater system than in the marine sediments of the SRS system.

From these two examples, it is easy to see that the geology of an area will dominate the velocity of tritium movement away from a source. Furthermore, the location of the source in the geologic setting will determine the residence time of tritium in the groundwater. In most, but not all, cases the residence time in geologic systems is large compared to those in atmospheric , terrestrial, and aquatic systems. In many cases the residence time is several times the half-life of tritium and can lead to substantial reduction of the tritium entering the surface environment. For this reason, the greatest concern with contamination of groundwater by tritium involves the contamination of aquifers that are used as sources for irrigation or drinking water .

TRITIUM IN THE OCEAN

The ocean is, by a large margin, the major global reservoir of hydrogen and, as such, has an enormous capacity for dilution of tritium. Given time, most of the undecayed tritium released to the environment will finally reside in the ocean. Tritium transport into the ocean from continental sources is through river water discharges, advection of continental air over the ocean and deposition, and discharge of groundwater directly into the ocean (Weiss et al. 1979).

River discharge takes place first through the flow out of the river estuary onto the continental shelf. Estuarine transport is partially controlled by the efflux rate of the river and partly by the tidal and coastal water currents in an area. The characteristic turnover time of water in most estuarine system is in the order of days or weeks. This can be of local importance but has little effect on the tritium concentration in the ocean.

The mixing of water between the continental shelf and coastal bays and inlets is driven by the local system of currents. In many cases, the mixing is not continuous but driven by events such as storms. Blanton found that the residence time of water in the south Atlantic coast of the U.S. to be about 3 months. The characteristic mixing in most coastal areas seems to be on the order of months (Hayes 1979, Hetherington and Robson 1979).

Deposition of tritium in the ocean is governed by both dry and wet deposition processes, as was shown to be the case for land areas. The average rainfall over the ocean is greater than that over land, in terms of both rainfall per area and the integrated total. For this reason, the deposition through wet processes should be somewhat greater. Dry deposition over water is generally less efficient than dry deposition over land because of the stable atmospheric conditions over water. However, the long fetches, high wind speeds, and rough surface over the ocean will allow considerable dry deposition of gases, such as tritiated water vapor.

Most of what is known about the transport of tritium in the ocean is from the study of the transport of tritium injected into the stratosphere by the nuclear test detonations in the early 1960's. Initial efforts to reconcile measurements of tritium entering the ocean and tropospheric air concentrations indicated that the deposition over the ocean must be greater than that over the continents. This was supported by the fact that the concentration of tritium in the atmosphere over the ocean was less than that over land, suggesting that it was being removed at a higher rate over the ocean. At first this was justified on the basis of a higher rainfall rate over the ocean. However later calculations suggest that a large portion of the tritiated water deposition into the ocean is by dry deposition (Weiss et al. 1979). The large wet and dry deposition rates for tritiated water are the reason for the relatively short residence time of tritiated water in the atmosphere when released into the atmosphere from the surface of the earth.

Tritium is initially deposited into the surface "mixed" layer of the ocean. This layer is about 100 meters thick and mixing to this depth is relatively rapid. While the mixed layer has a very large dilution volume for tritium, an even greater volume exists below the mixed layer. Studies of the transport of tritium in the ocean have shown that mixing of the surface layer with the deep ocean takes place at a few locations where cold surface water sinks into the depths (Michel 1976, Ostlund and Fine 1979). At these locations, tritium transport is fairly rapid; however, the average tritium transport from the surface layer to the deep ocean is comparatively slow. The turnover time of the deep ocean is on the order of 100 years or more. This means that tritium reaching the deep ocean is likely to decay to extremely low levels before the water reappears on the surface.

TRITIUM EXPOSURE IN ENVIRONMENTAL SYSTEMS AS RELATED TO TRANSPORT, DILUTION VOLUMES, AND RESIDENCE TIMES

When tritium is released into the environment, the exposure of any part of the system will depend on the magnitude of the transport into that part of the system and the residence time of the tritium after initial introduction. The processes that transport tritium in the environment and the storage capacities and residence times of components of the environment have been identified and described. Table 2 summarizes the range of residence times and the importance of different components of the environment following a short duration release of tritium, such as might follow a process breakdown in a nuclear facility. Several release modes are presented because the exposure to different parts of the environment is also affected by the type of release.

For a short duration release of tritiated water into the atmosphere, most of the exposure will be to the environment close to the release site. The majority of the tritium will remain in the atmosphere as the tritium is carried away from the site by the wind. Therefore, the most important component in determining exposure is the atmosphere. In this case, the magnitude of the exposure is determined by the atmospheric conditions at the time of the release. The conditions include the amount of air humidity that will dilute the release, the wind speed, and the mixing processes in the atmosphere that will dilute the release. Only the leaf water of terrestrial plants will approach the concentration of tritium in the air. Animals will absorb tritiated water from the air through inhalation and skin absorption but the ratio of intake to volume of water in an animal is smaller than in a plant leaf and the concentration of tritium will be lower.

In a release of tritiated hydrogen to the atmosphere, an even greater fraction of the tritium will remain in the atmosphere. The solubility of hydrogen is lower than that of tritiated water and the only part of the environment that interacts readily with tritiated hydrogen is the soil. Therefore the residence time is determined by the residence time of tritiated water in the soil. In terms of the total tritium entering the soil, the amount of tritium oxidized in the soil to tritiated water is very small. The only reason that this cannot be totally ignored is that it has been shown that the tritium dose to man following atmospheric releases of tritiated hydrogen is almost totally from oxidation in the soil and the readmission of tritiated water into the atmosphere. However, since this process is much less efficient than direct absorption of atmospheric tritiated water vapor by man, the dose is always considerably smaller than from a similar release of tritiated water.

The exposure from a release of tritiated water to the soil is determined by the transport of tritium from the soil to the other parts of the environment. Vegetation will transport tritiated water by mass flow and the concentration in the vegetation will be the same as that in the soil. Animals will take up tritium in the vegetable or animal food they eat and from the water vapor that enters the atmosphere from the soil. The residence time of tritium in this case will be determined by the residence time in the soil. The residence time generally is larger in arid systems than in humid systems.

Table 2. The the range of residence times and importance of environmental components for short durantion release of tritium.

Environmental Component	Residence Time (min)	Release Type					
		Atmospheric					
		HTO	HT	Soil Surface	Stream	Lake	Ocean
Atmosphere							
Troposphere	$10 - 10^2$	*					
Terrestrial							
Soil-Water	$10^3 - 10^5$		*	*			
Leaf-Water	$10 - 10^2$	x					
Plant							
Water	$10^2 - 10^3$	x					
Organic	$10^4 - 10^5$						
Animal							
Water	$10^3 - 10^4$	x					
Organic	$10^4 - 10^4$						
Aquatic							
Physical							
Streams	$10^2 - 10^3$				*		
Lakes	$10^4 - 10^6$					*	
Biological							
Water	$10 - 10^2$				x		
Organic	$10^2 - 10^4$					x	
Groundwater	$10^3 - 10^8$						
Ocean							
Estuary	$10^3 - 10^4$						x
Surface	10^7						
Deep	10^8						

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* The most important component in determining the residence time in the environment.

x Components that participate in tritium cycling and have residence times that are large enough to be considered under some circumstances.

The exposure of aquatic systems following short duration releases are most affected by the physical residence time and dilution through mixing in the system. The physical residence time in stream systems is determined by the water flow in the system. In lakes the physical residence time is determined by flow and evaporation of tritiated water, the balance depending on the relative importance of each factor in a particular lake. The water in aquatic organisms has a short residence time and the concentration tends to follow the concentration in the water. Only a small fraction of the tritium enters into the organic matter in these systems; however, the residence time in organics, particularly sediments, can by quite long, in the order of tens of years.

Groundwater and ocean systems are not affected greatly by short duration releases. In the case of groundwater, little of these releases will reach the groundwater unless the release is directly to the groundwater. Estuaries are to some extent extentions of the stream systems which feed them and act like the streams with the additional dilution due to mixing with coastal waters. The quantity of water in the open ocean is too large to be affected by any reasonable magnitude, short duration release.

When the environment approaches a steady state following long duration "chronic" releases of tritium, the concentration in different components of the environment is determined by the balance of concentration and flow from other components of the environment. Concentrations are characterized by the long term average and exposure is this average integrated over the exposure time of interest (usually the life of the component).

In the vicinity of atmospheric releases of tritiated water, the concentration in the soil will be similar to that in rainfall, which is usually lower than that in the air water vapor because the source of water in rainfall is from a location away from the site. The concentration in vegetation will be intermediate to the concentration in the air and the soil. Vegetation organic tritium concentrations will be slightly lower than those of the water in the vegetation. The concentration in the water of animals will be most strongly influenced by drinking water, which may or may not be rainwater. The concentration in animal water is also influenced by the air concentration, and the concentration in the water of food. The tritium concentration in the organic matter of animals is influenced by the concentration in the food the animal eats and the water in the animal. The net effect of all these transfers is that the concentration of tritium in organisms chronically exposed to a tritiated environment is somewhat less but very near the average concentration in the environment. Because of the complexity of the inputs, it is rare to be able to measure all of these factors and it should not be surprising that there are some, though normally no great, differences in the concentration in one component, say animal water, and some other single indicator component, say the air water concentration.

The same type of balance determines the steady state tritium concentration in aquatic systems. However, the transport of water between the components of these systems is so rapid that almost all components have identical concentrations of water. Organic tritium in aquatic systems is somewhat different because of the dominant role detritus, dead organic matter, originating from outside the system plays in some aquatic systems. In this case, the concentration of tritium in the organic matter of the aquatic organisms may be supplemented or diluted, depending on the ratio of tritium to the other isotopes of hydrogen in the detritus.

In all of the systems studied, there has been no indication of biomagnification of tritium through food chains. This is what one would expect because the availability of hydrogen in the environment is so high that there is no reason for organisms to evolve mechanisms to enrich hydrogen. Studies which show higher concentrations of tritium in a single component of the biota have always been traced to some other source of tritium which is itself at a high concentration or to differences in retention times after transient exposures. An example of this in a terrestrial system is the elevated levels of tritium in vegetation that was traced to high soil water concentration due to oxidation of tritiated hydrogen, which was at relatively high concentrations (Murphy and Pendergast 1979). An example in an aquatic system was the relatively high organic tritium concentrations in organisms exposed to the high relative tritium concentrations of reactor water purification resins (Kirchmann et al. 1986b).

The preceding situations describe what can be expected locally following releases of tritium to the environment. The global consequences have been intensively studied as part of evaluating the consequences of the tritium released into the atmosphere following the nuclear weapons testing in the 1960's. This information has been summarized and was used in NCRP Report No. 62 to evaluate a number of scenarios of tritium releases to the environment.

This methodology was used to evaluate the fate of tritium released from the SRS. The model is based on seven compartments which are the main global reservoirs for water. The compartments are the atmospheric moisture, soil moisture, groundwater, surface fresh waters (rivers and lakes), inland saline surface waters, the surface (well-mixed) ocean, and the deep ocean. Tritium is assumed to move with the water. The water transport between compartments is based on the generalized global water budget (NCRP 1979, Killough and Kocher 1988).

The fate of SRS tritium was estimated by using the SRS atmospheric and stream releases as input for the model. The tritium released to the burial ground and the tritium that still resides under the seepage basins on the site was not included since it has not left the SRS. The results are illustrated in Figure 19 (Murphy et al. 1990). The results indicate that most of the tritium is rapidly moved to the surface ocean. After only ten years, 70% of the tritium remaining after radioactive decay is in the surface ocean. The movement into the deep ocean is somewhat slower, but by the end of 1988, 91% of the total tritium remaining in the environment from SRS releases is in either the surface (47%) or deep ocean (44%). The tritium in the ocean is greatly diluted by the large mass of water in the ocean. Surface ocean concentration due to SRS releases are calculated to be only 0.0002 pCi/mL. The concentration in the deep ocean is calculated to be even less, 0.000003 pCi/mL.

The tritium in the atmosphere and soil have decreased slowly since a peak in 1957. This is a reflection of the trend of decreasing SRS releases following the peaks in the early 1950's. The decrease in the surface and groundwater tritium inventory is slower, with peaks in 1965 and 1973, respectively. This is a reflection of the slower turnover of groundwater.

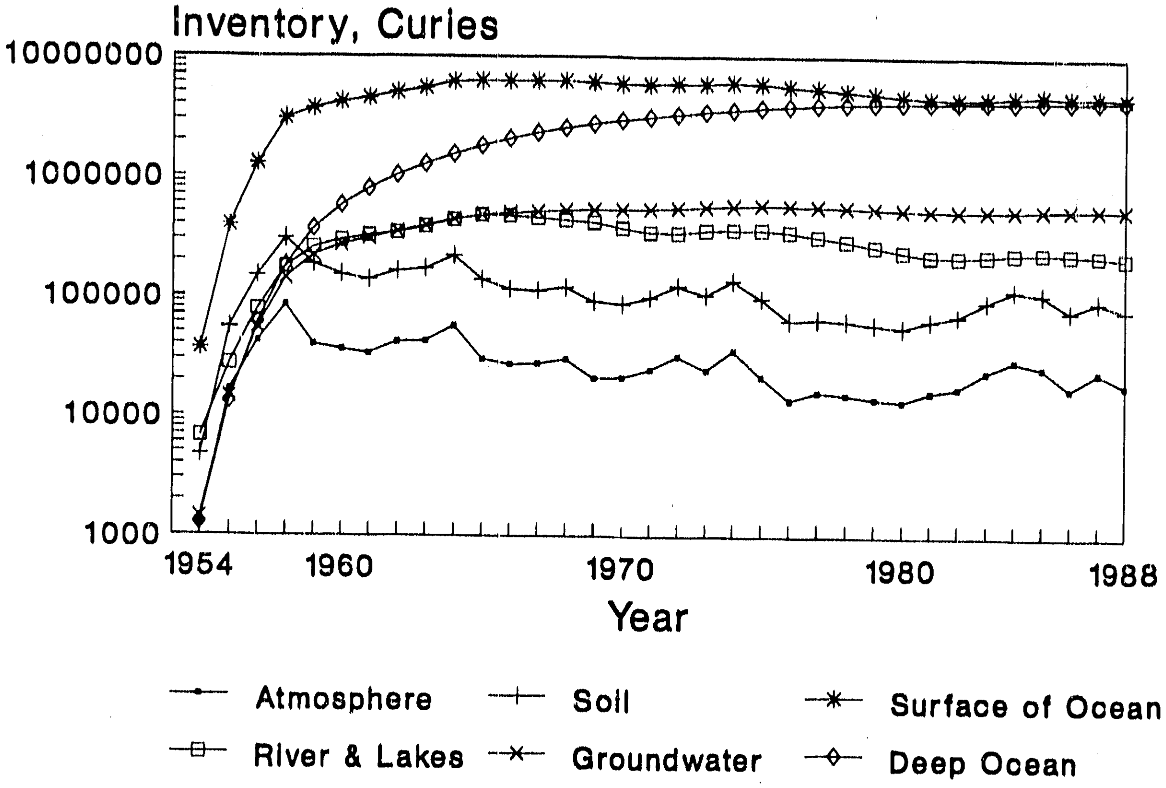

Figure 19. The global distribution of SRS tritium releases according to the NCRP seven-compartment model (Murphy et al. 1990).

Because of the rapid distribution and dilution of tritium into the global water reservoir, the consequences of tritium released from SRS nuclear facilities are quite small. Tritium releases from the SRS are large compared to nuclear power reactors and are on the same order of size as projected fusion facilities. Therefore the same, very small, effect would be expected for any other planned tritium using or producing facility.

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