THE SIGNIFICANCE OF TRITIUM RELEASES TO THE ENVIRONMENT*

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Summary

Tritium is produced naturally and was present in low concentrations in precipitation and natural bodies of water before atmospheric testing of nuclear weapons. Other sources of tritium are now present from which tritium is released to the environment. Nuclear reactor tritium production, according to recent estimates, will equal natural tritium production before the year 2000. Predicted increases of tritium in the environment will take place first on a local ecological level and then appear on a biospheric level.

Tritium introduced into the environment as THO will move through ecological systems in the same manner as stable water. Tritium will enter the hydrologic cycle either via evapo-transpiration or the surface bodies of water.

Ecological experiments have been conducted to determine the movement of tritium in the environment. Field-grown plants were exposed to liquid and vapor THO for periods of one-half and one hours. Tritium concentrations were determined in leaf samples collected after exposure for periods of time up to 45 days. Tritium decays rapidly in the plant species studied and exhibited a three component half-life when plants were exposed to THO vapor. The length of exposure, and sources of THO in the soil affect the half-time of tritium in the plant tissues.

Data produced in ecological experiments on tritium movement are used in a theoretical consideration of acute and chronic vapor releases of tritium in an agricultural environment. An acute release of THO for one hour at a concentration of 2×10^5 pCi/meter^3 will persist in the environment long enough for transfer of tritium to man via cow's milk to occur. Plants will contain $6.66 \times MPC_w$ under these conditions but ecological dilution will reduce this to $.36$ MPC_w in milk and $.01$ MPC_w in the body water of man. Chronic release of THO vapor at a concentration of 2×10^5 pCi/meter^3 produces THO concentrations of $6.66 \times MPC_w$ in plants, $2.02 \times MPC_w$ in milk, and $.17$ MPC_w in the body water of man from the ingestion of milk.

Short-lived THO vapor releases as short as one hour will have some ecological and biological consequences in terms of human food chain effects. On one hand, rapid uptake and incorporation of tritium are offset by rapid losses and dilution in the ecological water compartments of the environment. To determine the significance of any tritium release to the environment, it will be necessary to know the rate at which tritium moves from the source through the ecological system to ultimate dilution in the oceans.

Tritium Production

Tritium is the only radioactive isotope of hydrogen and it is produced naturally by cosmic ray interactions with nitrogen and oxygen in the upper atmosphere. Various estimates of natural tritium production were reviewed by Nir et al.[1] and it was concluded that the natural production rate resulted in a total equilibrium inventory in the biosphere of approximately 69 megacuries of tritium. Most of this naturally-produced tritium (90%) is in the hydrosphere consisting of the oceans and other bodies of water. Previous to the detonation of thermonuclear explosives in the atmosphere, we therefore had a natural tritium background which was very low, on the order of 10 tritium atoms per 10^{18} hydrogen atoms** (10 T.U.) in continental rainwater. In conventional radioactivity units, this is equivalent to 32.3 pCi of tritium per liter of water which is below the detection limits of liquid scintillation spectrometers. Recent precipitation in the northern hemisphere today is still about 10x the pre-thermonuclear level of 10 tritium units.

Tritium was released to the biosphere during the period of atmospheric testing of nuclear weapons and Eriksson[2] estimated that approximately 1700 megacuries of tritium were produced in tests up to 1962. This is almost 25 times the steady-state inventory from natural tritium production. A three year half-time of tritium in the stratosphere was suggested for this weapons-produced tritium[2], however, recent study of tritium in worldwide precipation indicates a much shorter half-residence time of 10 days[3]. Rainfall during the high fallout period had an increased tritium content and values in the range of 10^3 T. U. were measured in northern hemisphere rainfall, with a high of 12,950 T.U. in precipitation in Denver, Colorado during May 5-18, 1963[4]. Our environmental tritium background therefore increased during the period of maximum fallout deposition and would gradually return to the low level of the pre-thermonuclear period if no further additions of tritium to the biosphere were made.

Other sources from which tritium is released to the environment are now present, and as electrical power production by nuclear reactors increases, we may expect some increases in the concentration of tritium in the environment to occur. This will occur initially on a local ecological basis, and later on a biospheric level. The Environmental Protection Agency (EPA) has recently established a Tritium Surveillance System[5] which began to collect and analyze surface water samples in 1970 from the 50 states, Panama Canal Zone, Guam, and Puerto Rico. The data obtained from this network will provide the EPA with information to evaluate any future changes in environmental

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** 1 tritium unit (T. U.) = 1 tritium atom/10^{18} hydrogen atoms also called a tritium ratio.

tritium levels. Cowser et al.[6] estimated that total reactor tritium production will not equal the natural tritium production rate until about 1995. The rate of tritium production and the levels in the environment will increase at a faster rate if (1) an increased use of heavy-water reactor occurs throughout the world, and (2) the development of thermonuclear fusion reactors reaches an operationally useful level by this time.

Tritium produced in nuclear reactors is released to the environment by several pathways. The amount and the specific pathway by which tritium is released depends upon the type and design of the reactor. Tritium is produced by several reactions in the primary coolant of the reactor. Ternary fission, reactions with deuterium, lithium, and boron, and other nuclear reactions produce tritium during the course of reactor operation. Heavy water moderated reactors have the highest concentrations of tritium in the primary coolant while boiling water reactors have the lowest, according to Weaver and Stigall[7].

Tritium as tritium gas (T_2) may be released in stack gases in boiling water reactors as primary coolant is cooled in the condenser and air and non-condensable gases are discharged to the plant stack. Tritium as THO vapor will be removed by the condensation process. In a pressurized water reactor, leakage must occur between the primary and secondary coolant before the secondary coolant becomes contaminated by tritium. A smaller amount of gaseous tritium (HT and T_2) is thus released by the pressurized-water reactors. However, only 1% of the total tritium released by both types of reactors is as gaseous tritium. Most of the tritium is released as THO in liquid radioactive wastes.

Because tritium as THO is not removed by filtration or ion-exchange, it remains in the coolant which is either reused in the reactor, stored for future use, or placed in the liquid waste disposal system. Liquid wastes may be discharged to the environment in the coolent effluent if the concentrations of radioactivity are low, and large dilutions provided by the high flow of condenser cooling water produces low effluent concentrations of tritium. Reactors release from 5 to 1,300 curies of tritium annually and these amounts are usually less than 1% of the discharge limit[8].

Most of the tritium released to the environment will be in the form of tritiated water or THO, however some facilities will release tritium gas (T_2). Tritium gas is absorbed by mammals through their lungs[9] but to a less extent than THO, approximately 4 orders of magnitude less according to Osborne[10]. The oxidation of tritium gas occurs at a rate of 0.2 to 3% per day[11] by various catalytic reactions and therefore tritium gas releases have a reduced biological significance. Tritium gas releases may occur at very high levels, however, and should be evaluated just like a THO release, taking into account the oxidation rate. Tritium gas is also taken up by the foliage of vascular plant to a

limited extent[12]. Tritiated water may be released either as liquid water or as a vapor and because phase changes of water continually occur in ecological systems all phases of water in the system will contain tritium.

It appears therefore that we will live with a low but measurable level of tritium in our environment, as indeed we have for millenia, and that these levels of tritium in the biosphere will increase as nuclear power sources are developed. These predicted increases in tritium concentrations in the environment will not occur initially on the biospheric level, but will take place on the local, ecological level. It is often assumed that such rapid mixing takes place in the hydrologic cycle that any tritium introduced into it, will soon be distributed and diluted so that no significant radiation dose is received by any segment of the general population. As more information is obtained, from tracer studies and ecological research, the critical pathways of tritium movement in ecological systems, especially those leading to man, will be described. Our efforts which are described in this report, have been directed toward this goal.

Tritium and Water in the Environment

Eventually any tritium released into the environment will reach the ocean by the typical routes of the hydrological cycle. Hydrologic and ecological studies indicate that between 40 and 65% of the water received as rainfall is recycled back into the atmosphere by evapo-transpiration which involves the vegetation[13,14]. During the process, tritium in THO may be incorporated into organic matter where its fate may be similar to that of carbon. In plants that grow quickly, such as most crop plants, the level of incorporation of tritium from water will be greater than in slow-growing plants such as trees[15].

In our studies of tritium movement in the environment, we have conducted experiments on both types of tritium exposures, as liquid and as vapor. Some difference in the behavior of these two types of releases may be seen as the long-term aspects of the release is studied. It is obvious that when one studies the movement of tritium as THO in the environment, you are in reality studying the movement of water and hence this research has considerable basic scientific content. Many of the contaminants prominent in the biosphere today are transported via water and therefore information on the kinetics of water in ecological systems is an important aspect of environmental quality.

Water in ecological systems exists in a continuum between the soil, or soil-water, the plant, and the atmosphere. Water is taken up passively by plants from the interstitial water of the soil and moves through the plant in specific tissues by essentially hydrodynamic forces. Water is then released from the foliar surfaces of the plant as vapor in a process called transpiration. If tritium as THO water or vapor enters the soil-plant-atmosphere continuum, it then follows the pathway of water either being recycled as transpiration, or it enters the surface or

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ground water hydrological cycle. On the ecological level, tritium as THO is an excellent water tracer, and tritium moves through the ecological water compartments in the same manner as water does. The major effect seen will be one of dilution as the tritium pulse enters the various water compartments of the soil-plant-atmosphere continuum. It is possible that fractionation effects occur, on theoretical grounds at least, especially where water changes phase in the ecological system, namely at the leaf surfaces where water is evaporated.

When THO is sprayed on vegetation in an experiment, or is deposited on the vegetation as a vapor, it is absorbed directly into the plant foliage. In fact, many elements are absorbed by plant foliar surfaces and modern horticultural and agricultural practice make use of this knowledge derived from radio-tracer studies. A certain fraction of the THO applied enters the leaves of the plant, and there it is either utilized in photosynthesis and biochemical processes within the plant, or it enters the transpirational stream and is released as vapor to the surrounding atmosphere. It is possible that secondary deposition of THO released in transpiration occurs but the significance of this phenomenon is not known. In liquid THO applications, a fraction of the THO applied will fall off the plant foliage and enter the soil, constituting another input of tritium into the soil-plant-atmosphere continuum.

Experiments on Tritium Behavior in the Environment

One of a recent series of experiments conducted at the Lawrence Livermore Laboratories was concerned with the fate of liquid THO applied as a spray to natural, uncultivated vegetation. Approximately 12.6 mCi of THO in four liters of water were applied to a plot of weedy vegetation 1x2 meters in size. The major species of plants in this plot were the burclover (Medicago hispida), fiddleneck (Amsinckia grandiflora), wild ryegrass (Hordeum californicum) and the slender oat, (Avena barbata). After the application of THO, samples of leaves were collected several times a day from each plant species. The experiment extended to 872 hours post-application when only the burclover was present in the plot. The leaf samples were frozen in the field on dry ice immediately after collection. The tissue-water of the leaf samples was extracted in the manner described by Koranda et al.[19] which is essentially freeze-drying. The sample is extracted for 20-24 hours at which time the loosely-held water in the leaf tissues has been distilled from the sample. The dry residues of the sample may then be assayed for bound tritium by a modified flask combustion method. All tritium analyses are performed by liquid scintillation counting methods.

The data obtained in this acute THO (liquid) exposure of natural vegetation are shown in Figure 1. Since the plot of vegetation was affected by typical climatic factors, such as wind, rain, temperature variations, and photoperiod, the behavior of tritium in these plants is comparable to that which occurs under real environmental conditions. No irrigation or watering was used on the experimental plot during the study. The half-life of THO in the plant species in the plot occurs in two phases or components, the second of which decays about 10 times slower than the

first component. Because a significant input of tritium into the soil occurs in a THO-water exposure, the plants have access to THO for a period of time after the THO application and it is possible that the long component of the half-life is related to the turnover of water in the soil. Soil cores were taken in the late stages of this experiment to determine residual tritium present in the soil beneath the plot and the data are shown in Table I. THO activity was still present in the soil beneath the vegetation plot in concentrations of 10^3 to 10^4 dpm/ml. of soil water more than 30 days after application (applied THO-6.90×10^6 dpm/ml.).

Jordan et al.[15] demonstrated that the half-time of THO in the soil-plant system was significantly longer than the plant THO half-time alone. The tree half-time for THO was 6.6 days in the Puerto Rico rainforest, while the soil-tree half-time was 43 + 4 days. The reason for this extended half-time of tritium in the plant after soil application is that tritium movement in the soil occurs in a lens-like layer and at a rate proportional to the rainfall rate. As the relatively discrete stratum of THO in the soil-water zone moves through the soil profile, and the root zone of the plant, the plant will take up the THO pulse for a longer time than if only the aerial parts of the plant are exposed.

On the basis of this experiment and others we have conducted[16], three factors appear to affect the persistence of tritium in vegetation. These are (1) the specific water metabolism or physiology of the plant, (2) the presence of tritium in the soil, and (3) climatic factors. Zimmermann et al.[17] have studied the effects of different soil types upon the movement of THO or DHO in soil systems and have demonstrated the lens-like movement of a labelled water pulse through the soil profile.

While liquid THO exposures may occur only under rather specific conditions, such as the rain-out of stack gases[18], a common mode of release and deposition of THO in ecological systems will be as tritiated water vapor (THO). Several experiments have been carried out by Biomedical Division scientists at the Lawrence Livermore Laboratory in which THO vapor exposures to various kinds of vegetation and soil were made under natural field conditions. A THO-vapor exposure of uncultivated, native vegetation containing the same species involved in the THO liquid exposure was conducted on a .5x.5 meter plot of ground. In these vapor exposure studies, exposures have been made for periods of one-half and one hour to determine if uptake levels are dependent upon length of exposure. These experiments were designed also to measure the uptake of THO vapor by field-grown plants and to determine the persistence of the THO that was absorbed by the plant species. The persistence of any contaminant in a biological system may be realistically expressed as the half-time of the substance in the system, and therefore THO half-times in plants will be compared under various experimental conditions.

A lucite chamber 57x57 cm. was used to expose approximately $\frac{1}{4}$ meter^2 of field-grown vegetation. An air pump circulated the chamber air at a rate of 30 liters per minute through a large THO bubbler. The chamber volume was 185 liters and the chamber air was therefore circulated through the THO bubbler every 6.1 minutes. Chamber air concentrations of THO vapor

were determined by sampling the chamber air through a rubber serum bottle closure with a 100 cc. glass syringe. The 100 cc. air sample was circulated twice through 20 ml. of scintillator solution in a standard counting vial. No significant increase in the count-rate from a sample was recorded with further cycling of the air sample through the scintillator solution.

After exposing the vegetation for either one-half or one hour, the chamber was removed and samples of the foliage were collected at various time intervals after exposure. The samples were frozen immediately on dry ice (CO_2) in the field. The tissue-water of the leaf samples was extracted as described previously and assayed by liquid scintillation counting methods[19].

Figure 2 shows the results of a one hour exposure to THO vapor for a single species of plant, the burclover (Medicago hispida) which is a relative of alfalfa. The decay curve was fitted with a least squares fit method and exhibits a three component decay of THO in the tissue-water of the plant. Tissue-bound tritium was also determined in the samples after the tissue-water had been extracted, and these data are expressed as disintegrations per gram of dry sample. The first component had a half-life of .9 hours and accounts for the loss of approximately 91% of the absorbed tritium. This rapid component of the half-life is most likely associated with the release and evaporation of THO from the stomatal chambers within the leaves and the surfaces of the exposed leaves. The second component of the decay curve with a half-time of 17 hours is tentatively assigned to the transpirational water loss from the plant. The third component with a long half-life of 270 hours may be associated either with the turnover of water in biochemical reactions in the leaves, or may represent the half-time of THO in the soil-plant system. The half-time of the tissue-bound tritium is also on the order of 270 hours which provides some evidence for the former hypothesis. The soil-water also acquires a low level of THO during the vapor exposure even though the entire surface of the ground was covered by vegetation. In Figure 3, a comparison of soil-water THO concentrations is made at 36 days after exposure for both the one-half and one hour THO vapor exposures. The amount of THO in the soil-water apparently increases with the length of exposure.

Tritium concentrations in the tissue-water of the burclover after the .5 and 1.0 hour THO vapor exposures are compared in Figure 4. The .5 hour exposure data exhibit the same type of decay as the one hour exposure data but with the first component accounting for a greater fraction of the total loss curve in the .5 hour exposure. Apparently with a shorter exposure time, less tritium enters the compartments responsible for the longer components of the decay curve.

The plot exposed to THO vapor for .5 hour was sampled to 1075 hours or almost 45 days. The data obtained in this long-term study of a single THO vapor exposure are shown in Figure 5. The initial

concentrations of THO in the soil-water are shown near the left edge of the figure. Rain occurred at 187 hours and may have influenced the THO concentrations in the plants because at that point the data exhibits more variability than at other times in the study. The soil-water THO concentrations at 865 hours are shown in the inset on Figure 5 and very likely are responsible for the long half-life component of tritium in these plants. It is apparent that from a single short exposure of THO vapor (.5 hour) that long-term effects extending to beyond 40 days are seen in both the vegetation and in the soil.

Vapor and liquid THO exposures are compared in Figure 6. In this figure we compare THO vapor and the liquid THO data obtained by analyzing leaf samples of the same species, burclover. The decay of THO activity in the tissue-water of the leaves is similar for both types of exposure, indicating that the phenomena responsible for the retention of tritium in the plant is the same, regardless of the mode of exposure. A slightly higher level of THO is present in the liquid-exposed plants at late times beyond 140 hours which relates to the higher concentrations of THO in the soil-water resulting from the liquid exposure.

Acute and Chronic Release of THO Vapor to the

Environment

Anspaugh et al.[20] developed models to estimate the radiation dose to man from tritium releases to the environment. Using a similar approach, which is based on known biological constants and data derived from ecological experiments on tritium behavior under natural conditions, the following consideration of an acute and chronic tritium release will be made.

Two conditions will be considered in this discussion: (1) an acute exposure of THO vapor at 2×10^5 pCi/meter^3 air for one hour in an agricultural environment, and (2) continuous release of THO vapor at the same concentration also in an agricultural environment. The actual pattern of chronic releases under operational conditions is probably an irregular series of closely spaces acute releases. The ecological system partially recovers or "clears" the contaminant when another pulse is received.

The relationships between THO concentrations in the air, vegetation, milk, and man's body water under chronic release conditions and after equilibrium has been attained (after 30 days) are shown in Table 2. Most of the constants used in these equations were determined independently in other research and are discussed in Anspaugh et al.[20]. The rate of transfer of THO vapor from the air to the plant has been the unknown factor in previous analyzes. The data presented here and confirmed by others[21] indicates that this transfer is essentially instantaneous.

Under acute conditions of THO vapor release for one hour at 2×10^5 pCi/meter^3, the relationships are more complex because of the simultaneous decay of tritium in the various compartments, each compartment decaying with its own rate constant. The acute release relationships are shown in Table 3.

When the input value of 2 x 10^5 pCi/meter^3 THO concentration in the air is used, the concentration of THO in each of the ecological compartments may be determined by using the equations in Tables 2 and 3. When this is done, the values in terms of MPC^{22} levels are obtained and are shown in Table 4. Under both types of release higher than MPC_w levels of tritium occur in plants but after tritium has been transferred to the cow, and into its milk, the levels are seen to be below MPC levels.

The acute situation merits more attention because occasionally a release may be made in which MPC levels will be reached or exceeded for a short period of time, and the scientist is asked to determine the significance of the release of THO vapor. Based on air concentrations, the release apparently dissipates by diffusion and dispersion of the air mass into which the release was made. The approach used in the following discussion may be used as a guideline for scientists attempting to evaluate the effects of acute releases of THO to the environment.

The results of an acute release of THO vapor for one hour into an agricultural environment are shown in Figure 7. The THO vapor in the air is rapidly absorbed by the vegetation and after the exposure, tritium concentrations in the tissue-water of plants rapidly decays with a 1 day half-life. Data presented earlier demonstrated a complex, three component half-life of THO in plants (Figure 2) but the more conservative one-day half-life has been used to compensate for the expected wide variation in the THO behavior between plant species and at different times of the year. The integrated activity (pCi-days/ml.) under the 1 day half-life curve is 8 times greater than the total activity under the three component curve. The vegetation partially equilibrates with the specific activity of THO in the air with peak concentrations occurring in the vegetation just after exposure[16]. The air is assumed to have 5 ml. of HHO per meter^3 in these calculations, and the concentrations in plants (pCi/ml.) just after exposure are one order of magnitude less than the THO in the air (pCi/m^3). The resulting concentration of THO in the plants will be 2 x 10^4 pCi/ml. of tissue-water at the end of exposure.

When a grazing cow ingests the tritium-contaminated vegetation, the initial daily rate of intake is on the order of 6 x 10^9 pCi per day, assuming 30 kg. of vegetation tissue-water ingested per day (14 kg. dry matter). THO in the cow is excreted at the rate of .002 per liter of the body burden[23,24], and with a half-life in the cow of 3.5 days. Peak milk concentrations of THO will occur 2.5 days after the exposure and will be approximately 36% of the MPC in water. The binding of THO in milk solids will occur in an acute exposure as described here because milk is formed each day de novo from precursors but not solely from body water. The excretion of THO in cows and the incorporation of tritium into milk constituents has been studied recently by Kirchmann et al.[23], Potter[24], and Kistner[25].

Results presented by Kirchmann et al.[23] indicate that greater incorporation of tritium into milk solids occurs when cows ingest tritiated forage than when tritium was administered to them as tritiated water (THO). Kistner[25] found that the rate of tritium binding in milk solids with acute or chronic exposure to THO was essentially the same. The explanation given was that because milk is newly formed each day, the maximum levels attained are a function of the

body water THO concentration at the time of milk formation and the exchangeable hydrogen content of the precursor compounds from which milk is formed.

When an adult man consumes this milk at the rate of 260 grams per day,[26] a peak of tritium activity will appear in the body water at 9-10 days after the exposure to the vegetation, and decay with a 9-10 day half-life. The concentrations of THO in the body water will be .01 MPC or about 1/1000 of the concentration appearing in the vegetation after exposure. The reduction of 1.5 x 10^{-3} between the vegetation and man occurs because of the large daily flux of water in the ecological system from evapotranspiration, uncontaminated soil-water uptake, and other biological losses. If rain occurred during early times after exposure, post-exposure levels could be lowered.

These calculations in effect take into account tritium entering the soil because the use of the 1-day half-life more than compensates for soil-water tritium uptake during the vapor exposure. Koranda and Martin[16] described soil uptake of THO vapor by bare soil and vegetated soil plots. In Figure 5, initial soil-water THO concentrations are almost two orders of magnitude less than the initial vegetation THO concentrations. After 865 hours (36 days) soil-water THO concentrations were reduced by a factor of 100 in the 0-6 inch stratum indicating a rapid decay of soil-water tritium absorbed during the vapor exposure. It is apparent that although rapid uptake and incorporation of tritium occurs in the ecological system, even during a short exposure, the large flux of water through the system rapidly dissipates the pulse of labelled water. As more information becomes available, the maximum rate at which an ecological system could dissipate THO either as water or vapor may be estimated from these and similar data.

Under chronic release conditions of 2 x 10^5 pCi/meter^3 and after equilibrium between the ecological compartments has been established, the relationships shown in Figure 8 will pertain in the environment. Vegetation will contain approximately 1/10 of the air THO concentration on a pCi per ml. basis, and this will be transferred to the cow's milk at about 30% of the concentration in the vegetation. When an adult man consumes this milk at the rate of 260 grams per day[26] the resulting body water concentration is approximately 2.6% of that found in the vegetation. Inhalation under these conditions will contribute .87 of the MPC level in man while the equilibrium value from milk is .17 MPC. The ingestion of vegetation would also contribute .87 MPC.

Conclusions

These studies have shown that tritium releases to the environment, even those of an short-lived nature, have some ecological and biological consequence when evaluated in terms of human food chain effects. Rapid uptake and incorporation of tritium occurs in vegetation, either from vapor or liquid exposures, but because of the high rate of water utilization by plants, the absorbed tritium is rapidly lost by transpiration. In a closely-coupled agricultural environment, tritium is transferred to grazing animals and to milk within the short time the tritium pulse is in the ecological system. On one hand, rapid uptake and incorporation of tritium are offset by rapid losses and dilution in the ecological water compartments of the environment. The initial contact of the tritium in the environment, from the standpoint of human food chains, is the vegetation.

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When the uptake of THO by plants can be characterized, perhaps for a realistic range of environmental conditions, the transfer of tritium to man may be accurately estimated with the other known parameters at our disposal.

It is apparent that the ecological approach in determining the significance of future releases of tritium to the environment will provide the necessary information to describe the radiobiological effects of the release in terms of radiation dose to man. This approach involves the analysis of samples from the appropriate ecological compartments in the environment, and an understanding of the dynamic nature of those compartments.

References

1. Nir, A., S. T. Kruger, and R. E. Lingenfelter, E. J. Flamm, Natural Tritium, Rev. Geophys. 4, 441-456 (1966).

2. Eriksson, Erik, An Account of the Major Pulses of Tritium and Their Effects in the Atmosphere, Tellus 17(1), 118-130 (1965).

3. Krey, P. W. and B. Krajewski, Tropospheric Scavenging of ^{90}Sr and ^{3}H, Proc. Symposium on Precipitation Scavenging, Pac. N. W. Lab. Battelle Mem. Instit. and Div. Biol. and Medicine, AEC, Richland, Washington, 2-4 June, 1970 (1970).

4. Stewart, G. L. and R. K. Farnsworth, United States Tritium Rainout and its Hydrologic Implications, Water Resources Research 4 (2), 273-289 (1968).

5. H.S.M.H.A. Health Reports, June, 1971, 86(6), 521 (1971).

6. Cowser, K. E., W. J. Boegly, Jr, and D. G. Jacobs, ^{85}Kr and Tritium in an Expanding World Nuclear Power Industry, U.S. AEC Report ORNL-4007, pp. 34-39, Oak Ridge, Tennessee (1966).

7. Weaver, C. L. and G. E. Stigall, Public Health Evaluation of Nuclear Power Plants, Health Physics 13, 189-196 (1967).

8. Blomeke, J. O. and F. E. Harrington, Waste Management at Nuclear Power Stations, Nuclear Safety, 9, 239-248 (1968).

9. Pinson, E. A., and W. H. Langham, Physiology and Toxicology of Tritium in Man, Journal Applied Physiology, 10, 108-126 (1957).

10. Osborne, R. V., Permissible Levels of Tritium in Man and the Environment, Symposium on Tritium - Its Biology and Behavior in Man and other Systems, Radiation Research Society Meetings, May 9-13, 1971, Boston, Massachusetts (1971).

11. Doury, A., Risks Presented by the Diffusion of Large Quantities of Tritium in the Atmosphere, in Seminaire sur la Protection Contre les Dangers du Tritium, Service Central de Protection Contre des Rayonments Ionissants, Les Vesinet, France, April 16-18, 1964 (1964).

12. Cline, J. F., Absorption and Metabolism of Tritium Oxide and Tritium Gas by Bean Plants, Plant Physiol., 28, 717-723 (1953).

13. Ovington, J. D., Quantitative Ecology and the Woodland Ecosystem Concept, Advances in Ecological Research, 1, 103-192, Academic Press, J. B. Cragg, editor (1962).

14. Carlston, C. W., Use of Tritium in Hydrologic Research - Problems and Limitations, Bull. Int. Assoc. Sci. Hydrol., 3, 38-42 (1964).

15. Jordan, C. F., J. J. Koranda, J. R. Kline, and J. R. Martin, Tritium Movement in a Tropical Ecosystem, Bioscience, 20, 807-812 (1970).

16. Koranda, J. J. and J. R. Martin, The Movement of Tritium in Ecological Systems, Proc. of Tritium Symposium, Environmental Protection Agency, Las Vegas, Nevada, Aug. 30-Sept. 2, 1971 (1971).

17. Zimmerman, V., O. Munnich and W. Roether, Downward Movement of Soil Moisture Traced by Means of Hydrogen Isotopes, Isotope Techniques in the Hydrologic Cycle, Geophys. Monograph Series 11, pp. 28-36, I.A.E.A., Editor, G. E. Stoot (1967).

18. Chamberlain, A. C. and A. E. J. Eggleton, Washout of Tritiated Water Vapor by Rain, International Journal of Air and Water Pollution, 8, 135-149 (1964).

19. Koranda, J. J., P. Phelps, L. Anspaugh, and G. Holladay, Sampling Systems for the Measurement of Environmental Radioactivity, Symposium on Rapid Methods for Measuring Radioactivity in the Environment, Munich, Germany, July 4-9, 1971 (1971).

20. Anspaugh, L. R., J. J. Koranda, W. L. Robison and J. R. Martin, The Dose to Man Via Food Chain Transfer Resulting from Exposure to Tritiated Water Vapor, Proc. of Tritium Symposium, Environmental Protection Agency, Las Vegas, Nevada, August 30-Sept. 2, 1971 (1971).

21. Mason, B. J., H. W. Hop and C. L. Miller, Deposition Velocity of Tritiated Methane and Tritiated Water Vapor on Forage Crops, Proc. of Tritium Symposium, Environmental Protection Agency, Las Vegas, Nevada, August 30-Sept.2, 1971 (1971).

22. Report of I.C.R.P. Committee II on Permissible Dose for Internal Radiation, Health Physics, 3 (1959).

23. Kirchmann, R., L. Lafontaine, J. Van Den Hoek and G. Hoch, Transfer et Repartition du Tritum dans les Constituents Principaux du lait de vaches alimentees avec de l'eau contaminee, Seanc. Soc. Biol., 163, 1459 (1969).

24. Potter, G. D., G. M. Vattuone and D. R. McIntyre, Metabolism of Tritiated Water in the Dairy Cow, Univ. of Calif. Report UCRL-73414, Livermore, California (1971).

25. Kistner, G. N., Tritium Excretion Via Cow's Milk after continuous Intake of Tritiated Water, Symposium on Tritium, Environmental Protection Agency, Las Vegas, Nevada, August 30-Sept. 2, 1971 (1971).

26. U. S. Dept. of Agriculture, Agricultural Statistics, U. S. Government Printing Office, Washington, D. C., pp. 582-583 (1969).

TABLE I

RESIDUAL TRITIUM IN SOIL AFTER SINGLE

LIQUID THO APPLICATION TO VEGETATION

(THO Applied - 6.90×10^6 dpm/ml.)

Depth (inches)	Concentrations in soil water 20 days post THO application Dpm/ml. soil water
0 - 3	1.80×10^4
3 - 6	1.54×10^4
6 - 9	2.71×10^3
9 - 12	1.54×10^3
12 - 15	8.70×10^2
15 - 18	6.71×10^2

TABLE II

TRITIUM CONCENTRATIONS IN THE ENVIRONMENT UNDER CHRONIC

RELEASE CONDITIONS AFTER EQUILIBRIUM (30 Days)

(Air concentration = 2×10^5 pCi/m^3)

Concentration of THO in vegetation (C_V) = $\dfrac{\text{Average air concentration THO (pCi/m}^3)}{(2)\ (\text{Absolute humidity} - \text{ml/m}^3)^*}$

Concentration of THO in milk (C_m) = $\dfrac{f_m\ (\text{MPD})\ C_V}{\lambda_m}$

where: f_m = fraction of body burden secreted per liter of milk (.002/liter)

MPD = daily water intake as tissue-water of forage consumed (30 kg/day)

λ_m = rate constant for tritium in milk $.693/T\frac{1}{2}$ THO in milk, $T\frac{1}{2} = 3.5$ days

Concentration of THO in adult man from milk (C) = $\dfrac{(\ell)\ (f_m)\ (\text{MPD})\ C_V}{\lambda_m\ \lambda_e\ M}$

where: ℓ = intake of milk per day (260 gm./day)

λ_e = rate constant for human body water (.693/10 d.)

M = body water volume of man (43 kg)

* Assume 5 ml./meter^3

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TABLE III

TRITIUM CONCENTRATIONS IN THE ENVIRONMENT AFTER AN

ACUTE RELEASE OF THO VAPOR

(One hour at 2×10^5 pCi/meter^3)

After exposure ceases

Concentration of THO in vegetation $(C_v) = \dfrac{\overline{C}}{2(H)} \, e^{-\lambda_p(t')}$ $\qquad \lambda_p = \dfrac{.693}{1 \text{ day}}$

Concentration of THO in milk $(C_m) = \dfrac{f_m(MPD)C_v^o e^{-\lambda_p(t')}}{(\lambda_m - \lambda_p)} + f_m(MPD)\,C_v^o \left[\dfrac{1}{\lambda_m} - \dfrac{e^{-\lambda_m(T)}}{\lambda_m} - \right.$

$\left. \dfrac{1}{\lambda_m - \lambda_p} \right] e^{-\lambda_m(t')}$

Concentration of THO in man from milk $(C) = \dfrac{\ell\, f_m(MPD)C_v^o}{\lambda_m M} \left[\dfrac{\lambda_m e^{-\lambda_p(t')}}{(\lambda_m - \lambda_p)(\lambda_e - \lambda_p)} + \dfrac{1}{\lambda_e - \lambda_m}\left(1 - e^{-\lambda_m(T)} - \right. \right.$

$\left. \dfrac{\lambda_m}{\lambda_m - \lambda_p} \right) e^{-\lambda_m(t')} + \left(\dfrac{1}{\lambda_e} - \dfrac{e^{-\lambda_e(T)}}{\lambda_e} - \dfrac{e^{-\lambda_e(T)}}{\lambda_m - \lambda_e} - \right.$

$\left. \left. \dfrac{\lambda_m}{(\lambda_m - \lambda_p)(\lambda_e - \lambda_p)} + \dfrac{\lambda_p}{(\lambda_e - \lambda_m)(\lambda_m - \lambda_p)} \right) e^{-\lambda_e(t')} \right]$

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where: t' = time since exposure ceased

T = duration of exposure $\qquad C_v^o = \dfrac{\overline{C}}{(2)\,\text{Absolute humidity}(H)}$

NOTE: Other parameters the same as in Table 2

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TABLE IV

CHRONIC AND ACUTE RELEASES OF THO VAPOR TO THE ENVIRONMENT

CHRONIC RELEASE AT 2×10^5 pCi/meter^3

1 MPC$_{air}$ = 6.66 MPC$_W$ in plants = 2.02 MPC$_W$ in milk = .17 MPC$_W$ in body water of man from milk

= .87 MPC$_W$ in man from air

ACUTE RELEASE OF THO VAPOR - 2×10^5 pCi/meter^3 for one hour

1 MPC$_{air}$ = 6.66 MPC$_W$ in plants = .36 MPC$_W$ in milk = .01 MPC$_W$ in body water of man from milk

$T\frac{1}{2}$ = 1 day $T\frac{1}{2}$ = 3.5 days $T\frac{1}{2}$ = 10 days
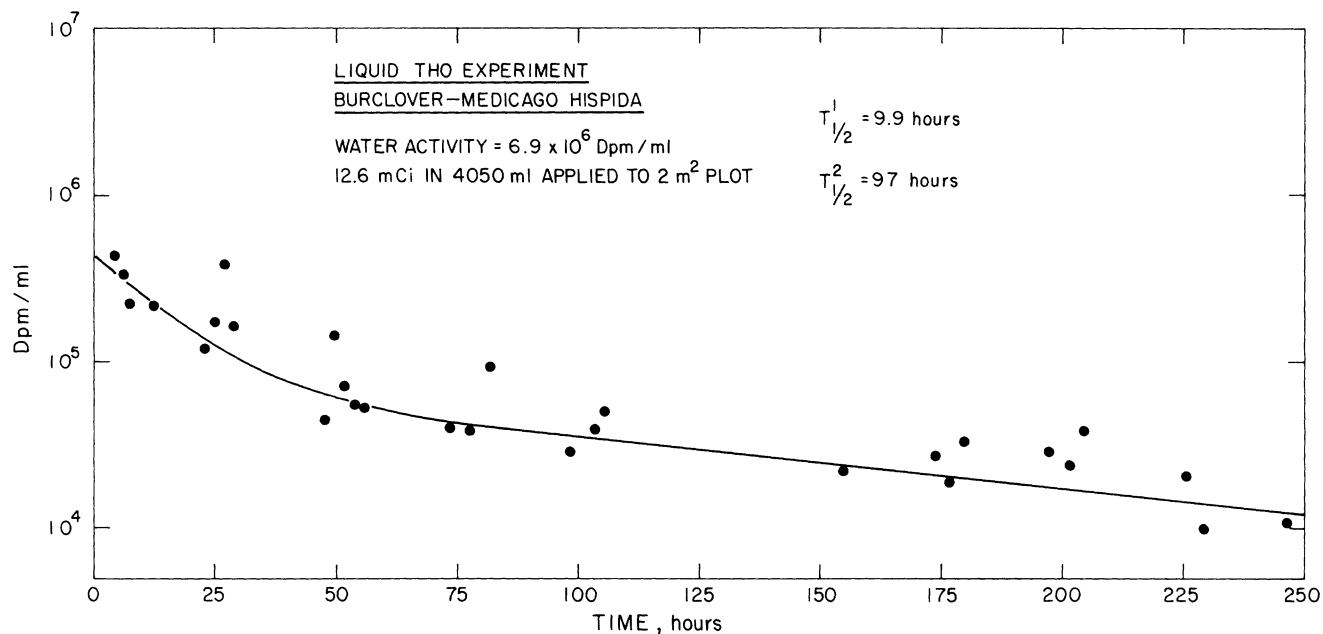
Figure 1. Liquid THO Exposure of Burclover (Medicago Hispida)
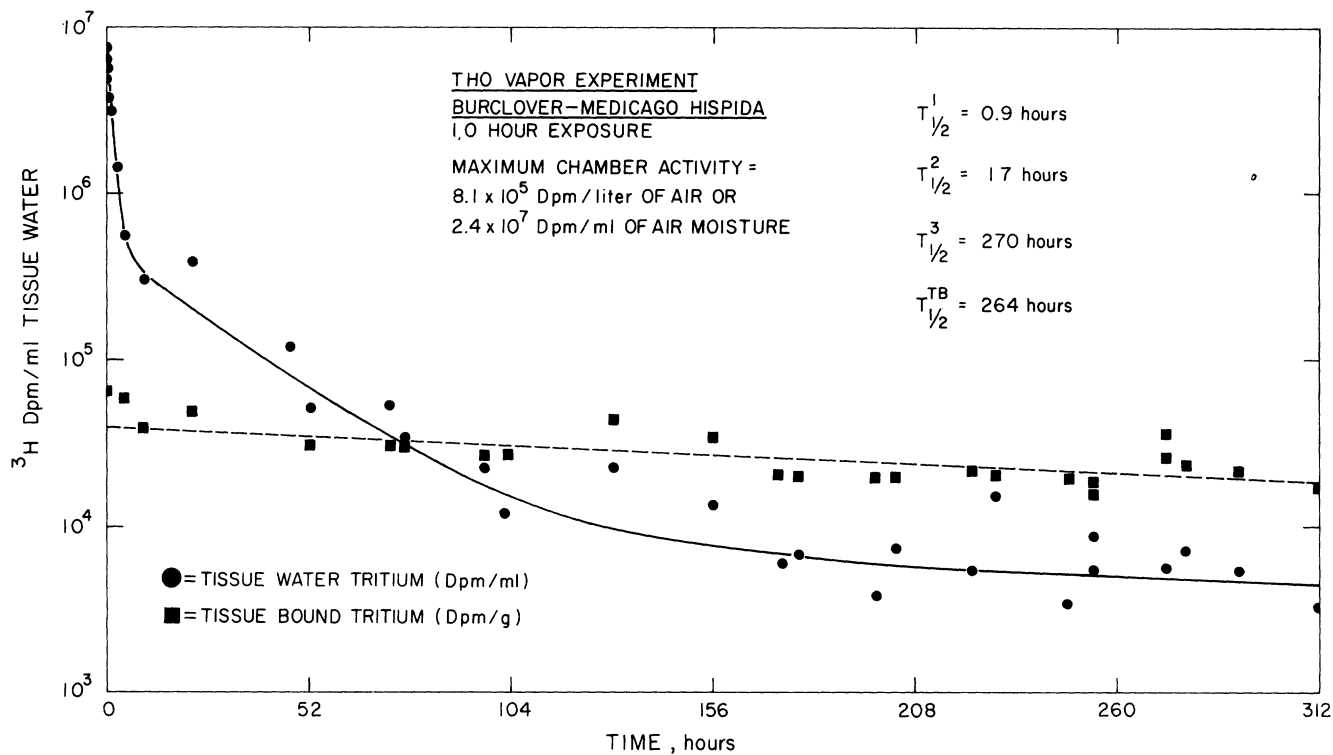
Figure 2. One Hour Vapor THO Exposure of Burclover (Medicago Hispida)
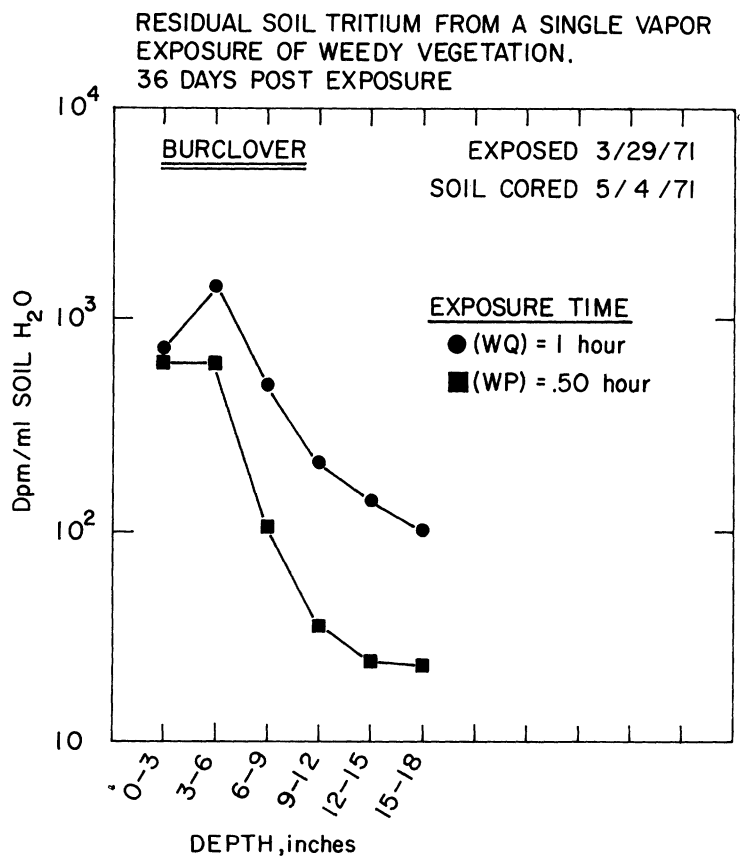
Figure 3. Residual Soil Tritium from a Single Vapor Exposure of Weedy
Vegetation - 36 days post exposure

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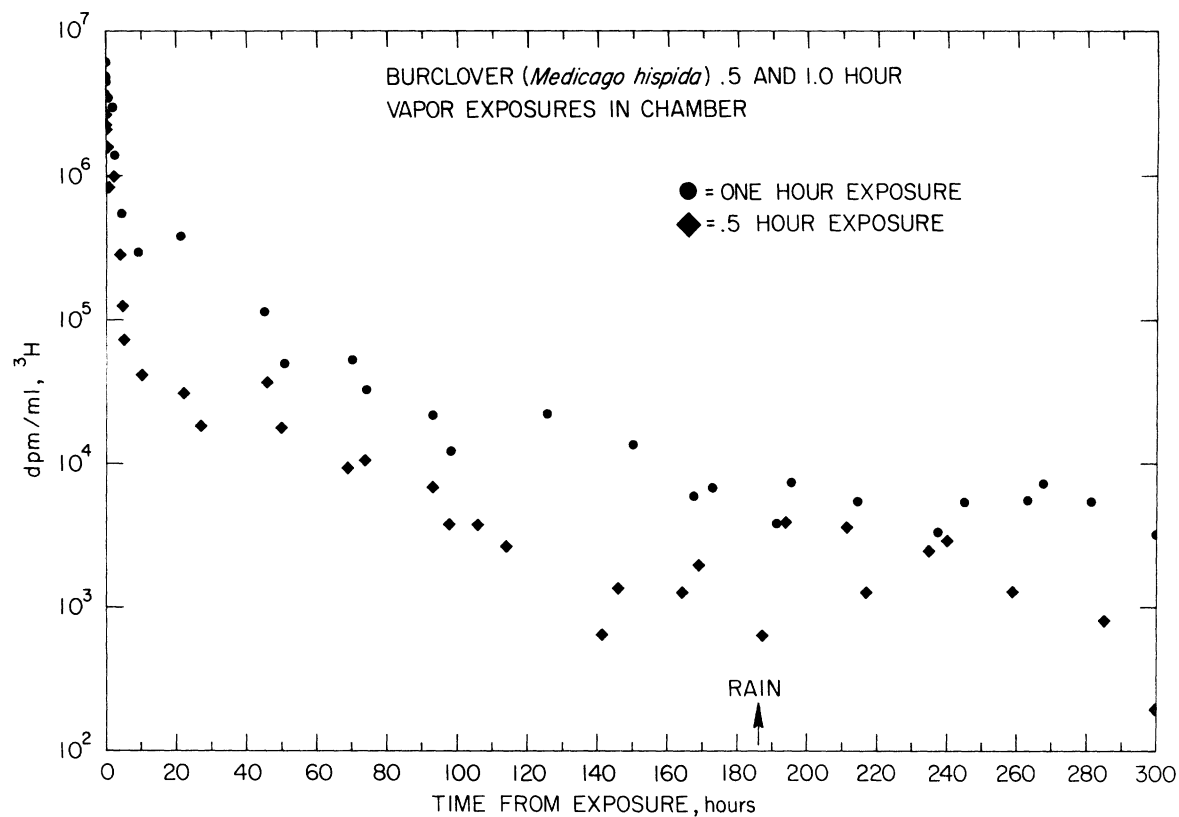
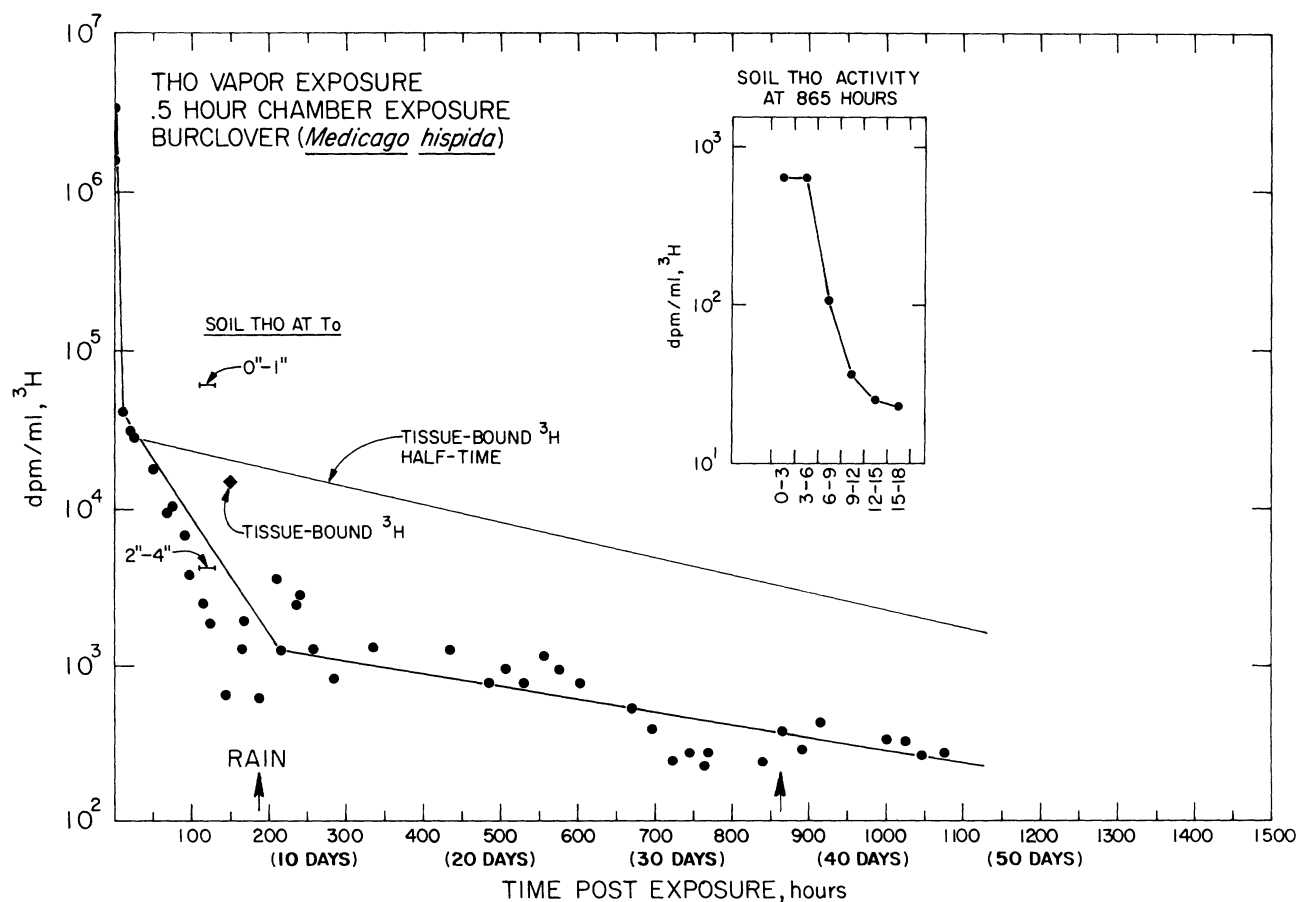Figure 5. Long-term Study of .5 Hour THO Vapor Exposure of Burclover

Figure 4. .5 and 1.0 Hour Vapor Exposures of Burclover (Medicago Hispida)
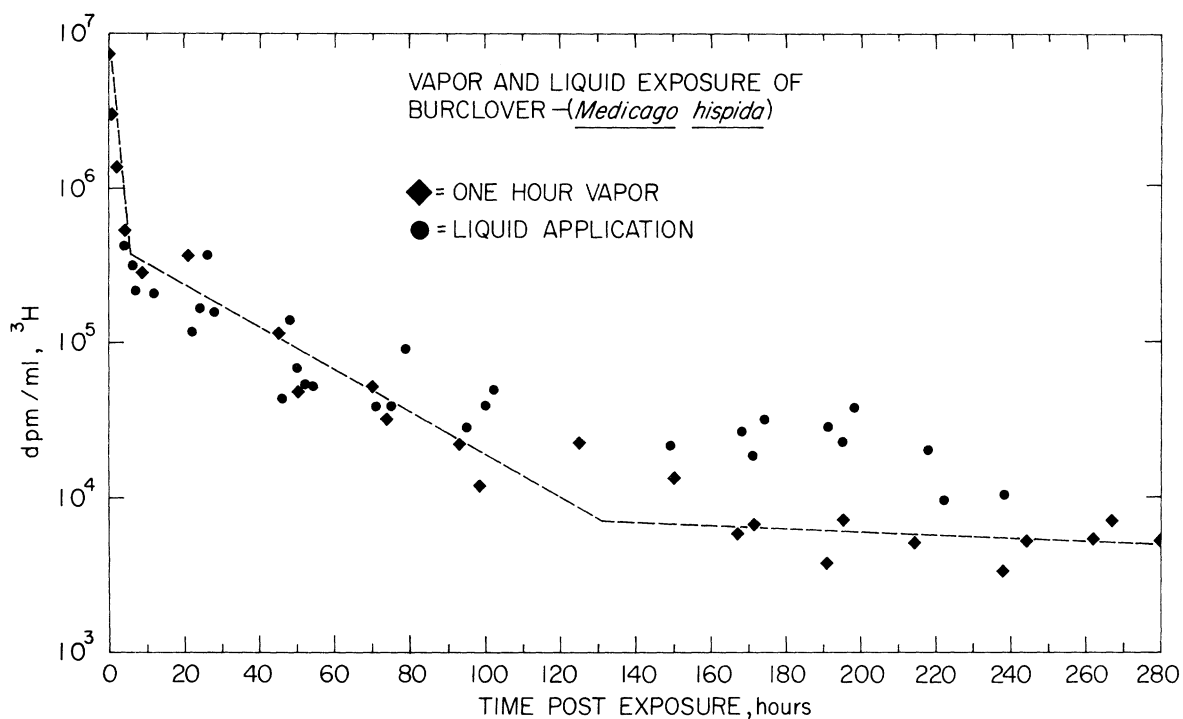
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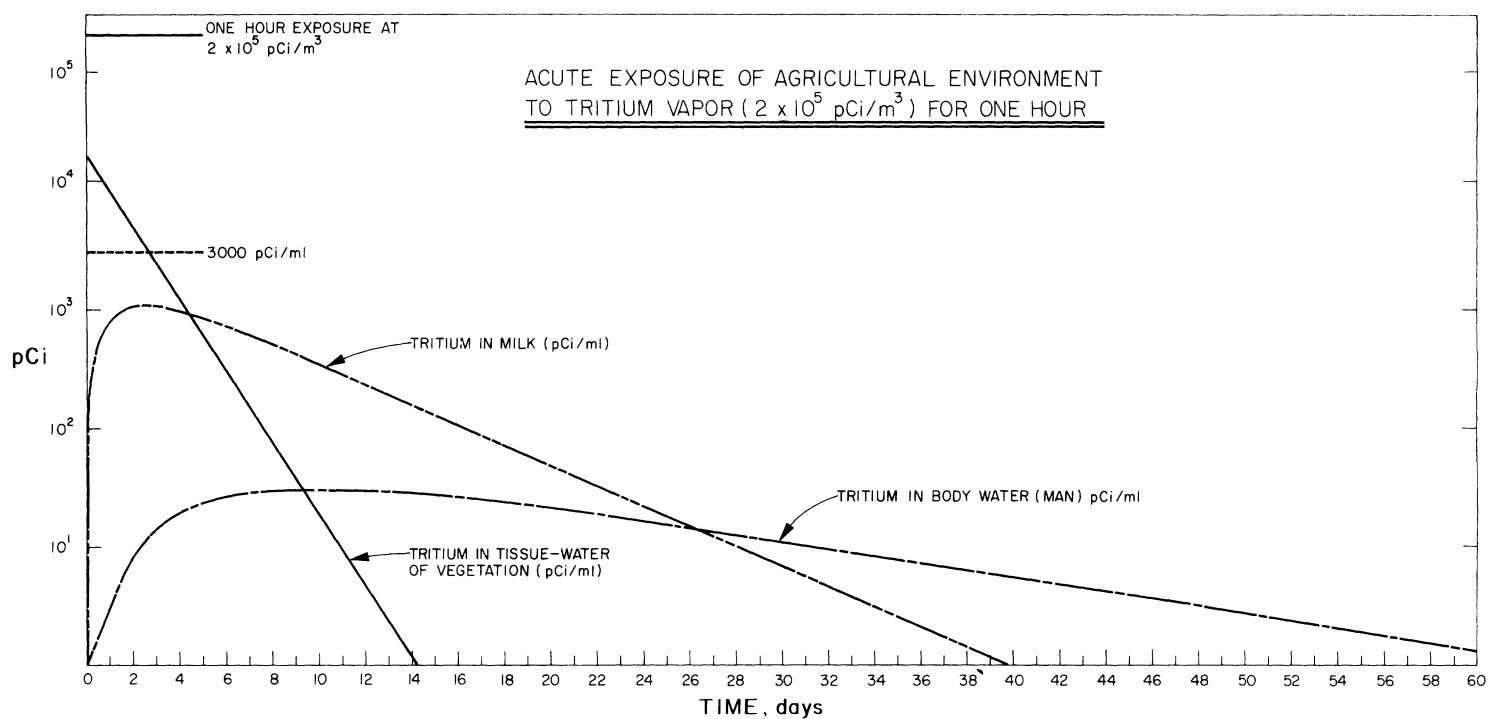Figure 6. Comparison of Liquid and Vapor THO Exposure to Burclover

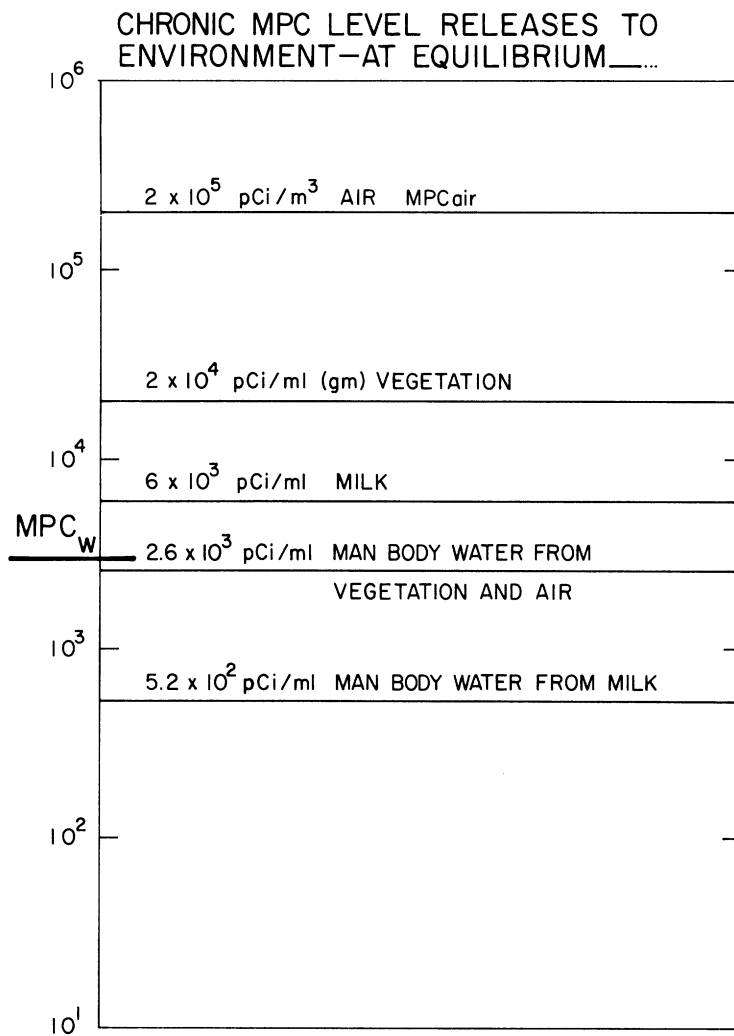Figure 7. Acute Exposure of Agricultural Environment to THO Vapor

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Figure 8. Chronic Releases of THO Vapor to the Environment