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ENVIRONMENTAL ASPECTS OF NATURAL GAS STIMULATION  
EXPERIMENTS WITH NUCLEAR DEVICES

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and W. L. Robison

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ENVIRONMENTAL ASPECTS OF NATURAL GAS STIMULATION  
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Abstract

It is likely that future activities administered by the USAEC Division of Peaceful Nuclear Explosives will be in the area of underground engineering, and particularly in the stimulation of deeply buried natural gas reservoirs of low permeability. The successful development of this technology could double the proved recoverable resources of natural gas in the United States. During the developmental stage of the technology, large quantities of THO are released to the environment as the gas is withdrawn.

Efforts to predict potential doses resulting from such releases have been severely handicapped by the lack of adequate data to predict the ecological movement of THO. A radioecological study was conducted in conjunction with Project Rulison, the second nuclear-stimulation experiment, to evaluate these parameters; controlled laboratory studies were also initiated. The results indicate that the environmental residence time of THO is at least an order of magnitude shorter than has been frequently assumed, and that this invalidates the commonly used deposition velocity approach for the calculation of doses due to food chain contamination by THO vapor.

A simple but realistic model is developed based on these findings to predict the dose to man via the forage-cow-milk, forage-cattle-beef, vegetation, and inhalation pathways. The dose predicted by this model for the Rulison area residents is  $10^3$  to  $10^4$  lower than the preflaring estimates made by a variety of organizations. Within the limits of the environmental data, the dose of  $3 \times 10^{-6}$  rem predicted from the model for the Rulison high-rate production flaring is verified as an upper limit.

These results are extrapolated to predict the dose to man if a field of nuclear-stimulated natural gas wells were to supply a 1000 MWe power station. The dose via all pathways would be an order of magnitude lower than the dose via inhalation alone if the same natural gas were to replace the natural gas supply delivered to the Los Angeles Basin or San Francisco Bay Area.

## INTRODUCTION

It appears likely that the future activities of the programs administered by the Division of Peaceful Nuclear Explosives of the U.S. Atomic Energy Commission will be in the areas of "underground engineering," and more specifically in the stimulation of natural gas reservoirs of low permeability. Substantial evidence that this will be true is provided by the fact that only two experiments involving nuclear explosives have been carried out with joint funding by private industrial concerns; both have been experiments in the stimulation of natural gas reservoirs of low permeability. Both were successful in that stimulation was achieved (1-4), although it is clear that costs must be substantially reduced to make the technology economically feasible (5, 6).

The potential benefits of the successful development of the nuclear-explosive gas-stimulation technology are highly significant in terms of the natural gas believed to be recoverable. The Bureau of Mines engineers have estimated that more than 300 trillion cubic feet of natural gas are recoverable by nuclear stimulation in Rocky Mountain basins alone (6). This is more than the present proved recoverable reservoirs of natural gas in the United States which are now being used at the rate of 20 trillion cubic feet per year (6, 7).

The basis for the technology is not radically different from what has been practiced in the past for low permeability formations. The production of natural gas can be increased by any mechanism that will increase the effective bore size of the well or increase the permeability of the medium around the well bore. Chemical explosives and hydraulic fracturing have been successfully used for this purpose, but are not economically feasible

for deeply buried formations of very low permeability (5). The very large amount of energy available from a physically small nuclear device, however, would be expected to provide enough stimulation to make recovery economically feasible, particularly if several devices can be emplaced within a single drill hole to stimulate a greater vertical thickness of the gas bearing zone (6, 8).

Many environmental aspects must be considered and carefully evaluated during the technology development phase. These include containment during and immediately after the detonation, seismic effects, and potential problems of ground water contamination and natural gas contamination by the radionuclides produced by the nuclear explosion. Most of these environmental aspects are not unique to gas stimulation, and considerable experience and expertise have been developed that can be applied to the potential problems of containment, seismic effects, and ground water contamination (9-16). Questions concerning the contamination of natural gas by explosion-produced radioactivities and the production of contaminated water and condensable hydrocarbon compounds concomitant with the withdrawal of natural gas, however, are unique.

During the development stage, such as the experiments Gasbuggy and Rulison, the produced natural gas has been flared (or burned) at the production site and any associated radioactivity has thereby been dispersed and released to the atmosphere. (Most of the separable liquids also produced have been reinjected into the flare stack stream and similarly dispersed.) Authority has not yet been granted to use the natural gas produced in these and future applications for any commercial purpose.

but among the several possibilities are the generation of electrical power at remote locations, the production of other hydrocarbon compounds, and the introduction of the natural gas into regular or special pipelines for regular or restricted commercial usage.

Jacobs et al. (17) have considered the latter two uses in some detail. The purpose of the present paper is to examine the unique environmental aspects of the experiments themselves where the natural gas is burned on-site, and to extrapolate these results to the possible use of the produced natural gas to generate electricity at remote locations.

#### Radioactivity from Nuclear Explosions to Stimulate Natural Gas Production

The explosion of any nuclear device produces radioactivity from fission products, fusion products (if a thermonuclear device is used), and activation products. For a contained underground explosion, the majority of the radioactive species will be contained in the melt or scavenged by the chimney rubble. The radionuclides of primary concern are those that may be present in a volatile form that will be withdrawn with produced natural gas. These include radionuclides of the noble gases and chemically reactive nuclides that will form gaseous compounds such as  $^3\text{H}$  and  $^{14}\text{C}$ . In addition, other possibilities must be considered such as the possible formation of volatile organic compounds of such elements as antimony, tellurium, tin, ruthenium and iodine and the more improbable possibility that some particulate radioactivity (most likely radionuclides produced by the decay of short-lived fission product gases) could be carried to the surface by the flowing gas stream (18).

For the two experiments, Gasbuggy and Rulison, considerable time

has elapsed between the detonation and re-entry of the well which has greatly reduced by radioactive decay the amounts of short-lived radionuclides. For Project Rulison, an inventory of the radionuclides present in the cavity 180 days after the detonation was calculated (19). Those radionuclides expected to be withdrawn with the natural gas are listed in Table 1. Several different organizations used these data to prepare estimates of possible population radiation dose via inhalation and food-chain contamination (19-22). Conclusions were that only the radionuclides  $^3\text{H}$  and  $^{85}\text{Kr}$  would be of possible concern, with  $^3\text{H}$  of substantially greater importance due to potential food-chain contamination.

Greater details of the source term data for these two radionuclides are given in Table 2. Data for both Gasbuggy and Rulison are based upon actual measurements as opposed to the calculated predictions given for Rulison in Table 1. All of the produced  $^{85}\text{Kr}$  is presumed to be in the gaseous phase. The tritium produced, however, is not all distributed in the gaseous phase, but most of it is in the form of water. Tritium will be present in the gaseous phase as  $\text{HT}$ ,  $\text{CH}_3\text{T}$ ,  $\text{C}_2\text{H}_5\text{T}$ , and  $\text{C}_3\text{H}_7\text{T}$  (23, 24). The relative amounts of tritium in these compounds and water depends upon chemical reactions at high temperatures and pressures which are influenced by the composition of the rock at the detonation point. Much of the knowledge needed to accurately predict the amount of tritium that would be in the gaseous phase is not yet available, but considerable progress has been made in constructing a predictive model (18, 25).

The Gasbuggy experiment employed a thermonuclear device whereas Rulison employed a larger-yield fission device. The tritium concentration in natural gas was thereby significantly reduced with only a minor increase

in  $^{85}\text{Kr}$  concentration. It should be noted that neither of these devices was specially designed for this application, but were spillovers from the weapons program (26). A new device is now under development at the Lawrence Livermore Laboratory (LLL) that is specially tailored for this application. Groseclose (26) has indicated that a reasonable lower limit for tritium production could be 2 Ci/kt if an all fission device is used, and shielding is effective in keeping prompt neutrons from reaching the soil or producing tritium in the shielding materials. The last column of Table 2 indicates what may be expected in the near future with the LLL device which is currently under test and development. These figures are estimated on the basis of the average concentration over a one-year period during which time about 5 billion cubic feet are withdrawn from a single well stimulated with four 100 kt devices (6). They are not therefore directly comparable with the Gasbuggy and Rulison data which are for early concentrations, but a reduction in the tritium concentration in gas of about a factor of 10 would be achieved even though it is conservatively assumed that 25% of the tritium would be in the gas phase. A minor reduction in  $^{85}\text{K}$  concentration would also be achieved.

It should be noted that the activity reported in the gaseous phase does not include water vapor or other condensable hydrocarbons. During the Rulison high-rate production testing about 30-35 barrels of water per  $10^6 \text{ ft}^3$  were produced, or about  $6 \times 10^7 \text{ ml/day}$ . Thus large volumes of contaminated water are produced which must be disposed of in some manner. For Gasbuggy and Rulison most of the water (and other condensates) was injected into the gas stream and released to the atmosphere. During the Rulison experiment, the water did in fact contribute a larger total release of tritium to the environment than did the natural gas. When the long-term production flaring operation was completed on 23 April 1971, a total of 2800 Ci of tritium had been released

(of which approximately 1500 Ci originated in the water) with the flaring of  $4.6 \times 10^8 \text{ ft}^3$  of gas; on the last day 9 Ci were released with only 0.08 Ci originating in the natural gas (29).

Extensive tests during the production of the Gasbuggy well failed to detect any radionuclide of elements that potentially could form volatile organic compounds, or radionuclides that would be expected to be associated with particulates such as  $^{137}\text{Cs}$  or  $^{90}\text{Sr}$  (18, 23, 30). During flaring of the Rulison gas, however,  $^{203}\text{Hg}$  was detected at levels of 10-13 pCi/m<sup>3</sup> of natural gas (2,3). This very low level of  $^{203}\text{Hg}$  evidently originated from neutron activation of stable  $^{202}\text{Hg}$  contained in rock at the detonation point (3). A reduction in the number of neutrons reaching the emplacement medium will substantially reduce the production of such products in future gas-stimulation events.

#### Radioecological Aspects of the Release of Tritiated Water

The results of our analysis of the potential biological hazards due to the flaring of the Rulison gas indicated that by far the greatest potential hazard would be due to food chain contamination by tritiated water (20); other independent analyses came to the same conclusion (19, 21, 22). All of these analyses, however, were based on several parameters for which no solid data existed, and used models more appropriate for the deposition of particulate fallout rather than exposure to tritiated water.

We therefore undertook a radioecological study at the Rulison site to evaluate some of these parameters, and also initiated controlled laboratory studies. The results of these studies, while not definitive at this time, clearly indicate that the assumptions and models used to evaluate potential

radiological hazards from Project Rulison grossly overestimated the actual exposures.

The radioecological study was carried out in conjunction with the calibration flaring (short releases over several days) and the eight day high-rate production testing with gas flows of approximately 12 to 16 million cubic feet per day (2). The study was divided into two operational phases. The first phase involved extensive, broad-area sampling of atmospheric moisture to rapidly locate areas where studies on ecological transport of tritiated water could be conducted, and the second phase was the actual conduction of such studies. The first phase was accomplished by fielding over an area of about 60 square miles up to 58 very simple samplers consisting of a 3.8 liter tin can filled with crushed dry ice. The condensed moisture was analyzed rapidly in a field laboratory by liquid scintillation spectrometry to determine activity per ml of air moisture. Measurements of air temperature and relative humidity were taken at the beginning and end of each sampling period so that absolute humidity could be calculated to convert activity per ml of air moisture to activity per m<sup>3</sup> of air.

Figure 1 is a map of the Project Rulison area showing the locations of the sampling sites. These sites were selected on the basis of accessibility, human habitation and meteorological predictions which indicated that the greatest ground-level, off-site air concentrations of tritium should occur under nighttime conditions and at a distance of about 5 km from the flare stack (19). The 90 feet high flare stack is located in Battlement Canyon about 2200 feet above Morrisania Mesa, the location of a small community of farmhouses at about 5 km. During the nighttime under conditions permitting radiative cooling, the plume was expected to be caught in the drainage winds down Battlement Canyon to Morrisania Mesa where it

would join the drainage winds associated with the Colorado River flowing to the southwest. It was not, however, a certainty that such would be the case. The thermal boost to the plume causes the effective stack height to be substantially higher than the actual stack height so that the plume could penetrate the inversion layer and move with the upper level winds which generally blow from the southwest. Several sampling sites were therefore located northeast of the release point.

The highest level of tritium measured off-site was at location B15. During this time period at least, the plume rise was apparently high enough so that the plume moved with the upper level winds over Doghead Mountain. Vertical mixing on the downwind side of the mountain then resulted in trapping the activity in the drainage winds of the adjacent canyon.

Representative results of the extensive, broad-area tritium measurements are shown in Table 3. The highest activity,  $170 \text{ pCi/m}^3$ , is 400 times less than the USAEC Concentration Guidance (CG) for off-site exposures of a suitable sample of the general population (31). Low levels of activity were observed over widespread areas on 28 and 29 October, but efforts to locate off-site areas for more detailed studies were largely unsuccessful. Efforts to study food-web transfer were initiated at sites B3 and B10 (single-family milk cow locations), but the levels of air activity were not sufficiently high to permit transfer studies.

Several experiments were carried out at locations within a few hundred feet of the flare stack. Here, as shown in Table 4, there was always significant activity above background levels. The unusually high values observed on 5 October 1970 were due to a malfunctioning in the water separator, and local rainout was occurring.

At several locations shown in Table 4 attempts were made to measure three parameters of the movement of tritium through the environment—the deposition velocity of tritium on a forage-type surface, the fraction of tritium deposition that is retained on vegetation, and the mechanical residence time of tritium in or on vegetation. Experiments were accomplished by bringing circular grass plugs of 3 7/8 inches diameter X 5 inches deep in from a nonexposed location, and placing them at various locations within several hundred feet of the flare stack. When removed, the samples were frozen, shipped to Livermore, and the water extracted by lyophilization and measured for tritium activity. No results are available for the tritium that may be organically bound. The experiments were carried out in October and November, and while the grass was still green, it probably was not metabolizing as actively as it would earlier in the growing season.

Results for two of the parameters are shown in Table 5. The measured values are segregated according to two exposure time periods. It is important to point out that the parameter referred to as a deposition velocity is really a rate constant and has been defined (32) as the deposition of activity per unit time per unit area divided by the air activity per unit volume, or

$$v = \frac{\mu\text{Ci}}{\text{sec-cm}^2} \cdot \frac{\text{cm}^3}{\mu\text{Ci}} = \frac{\text{cm}}{\text{sec}}$$

This is not, however, the way deposition velocities are usually measured, but rather the integrated air activity is measured ( $\mu\text{Ci-sec/cm}^3$ ) and the total deposition ( $\mu\text{Ci/cm}^2$ ):

$$v = \frac{\mu\text{Ci}}{\text{cm}^2} \cdot \frac{\text{cm}^3}{\mu\text{Ci-sec}} = \frac{\text{cm}}{\text{sec}}$$

Usually, the two expressions are equivalent particularly when dealing with particulate fallout because the residence time of particulate fallout (rate of loss constant) is long compared to the time of measurement. The results shown in Table 5, however, indicate that this is not the case for the "dry deposition" of tritium. The deposition velocity for tritium has been estimated by Jacobs (33) to be between 0.4 and 1 cm/sec. Measurements based on a 1 day exposure period yielded a value of 0.32 cm/sec, approaching the lower limit of Jacobs estimate. When exposures continued over a second day, however, the average result was halved -0.15 cm/sec. This indicates that the residence time of tritium on vegetation is short compared to the times of exposure used, and the residence times frequently assumed (19-22).

Measurements of the fraction of activity retained on grass at the end of the exposure periods were much more variable (Table 5), and perhaps increased with time. Much of the variability was removed by dividing by the density (wet weight) of the grass in  $\text{g/cm}^2$ . The average of all 15 measurements is  $2.9 \text{ cm}^2/\text{g}$ . It's interesting, but perhaps fortuitous, that approximately the same results were observed by Martin (34) for the deposition of radioiodine and radiostrontium following the Sedan Cratering event in Nevada. A value of 0.1 for the fraction of tritium intercepted by vegetation was used in one analysis (19), whereas most assumed 1.0.

Some evidence that the residence time of tritium in vegetation may be short (less than 1-2 days) has already been discussed in connection with the experiments designed to measure deposition velocity. Preflaring estimates assumed values for the half-residence time of 14 days (20), 20 days (22), 28 days (19), and 85 days (21). The latter estimate was derived

from data taken during Project Gasbuggy (35), but is based on only two measurements separated by 260 days, during which time flaring and the release of additional tritiated water was continuing.

An experiment was designed to directly measure the half-residence time at Rulison. Twenty-one grass plugs were emplaced 44.7 hours before the high-rate production test ended, and were retrieved at various times following the well shut-in. The results (Fig. 2) are best fit by a two component curve with the equation

$$\frac{A}{A_0} = \frac{1}{4} \exp \left[ \frac{-1.8}{\text{day}} t \right] + \frac{3}{4} \exp \left[ \frac{-0.01}{\text{day}} t \right]$$

which corresponds to half-times of 0.4 and 70 hour. Two factors may make these results atypical. First is the probable lack of metabolic activity, and second the plugs were placed in an environment that contained tritiated water with activity levels 4 times higher than that obtained in the plugs. After exposure ceased there no doubt was some redeposition of tritium from the environment due to the loss from native vegetation.

Laboratory experiments on the half-residence time of tritium in vegetation water have also been conducted at Livermore. Here exposure was achieved through the use of a chamber containing THO. Results for 0.5 hour exposures are shown in Fig. 3 for two different plants. Two components are clearly evident. Results of a 1.0 hour exposure are shown in Fig. 4 where an additional longer component is evident. Data on the organically bound portion of the tritium in these samples is not yet available, but data on other short-term experiments indicate that the second component is associated with the organically bound tritium (36). Results following a liquid application equivalent to a 2mm rain are shown in Fig. 5. Sampling did not begin soon enough to detect a possible rapid component, but again

turnover was relatively rapid, and a diurnal pattern is evident with peaks occurring in the afternoon indicating the effects of transpiration.

Best fit, multiple-exponential curves were derived for all controlled exposures using a CDC 6600 computer and an iterative, least-squares program (37). These curves are superimposed upon the data in Figs. 3-5, and the derived parameters are summarized in Table 6. The data indicate that the loss of activity following the exposure of vegetation to THO is a complex process, but that nearly all of the activity is lost very rapidly. The second component may represent organified tritium, and the relative magnitude of this component appears to increase as the time of exposure increases, which is consistent with the field experiments conducted at Project Rulison.

Two important conclusions can be drawn from these controlled exposure field experiments. One is that the residence time of tritium deposited upon vegetation is short, approximately an order of magnitude shorter than values frequently assumed. The second is that the concept of a deposition velocity cannot be applied to the exposure of THO, unless the deposition velocity is considered as an instantaneous rate constant in competition with high loss rates. The indiscriminate application of the deposition velocity concept does, in fact, lead to vegetation water concentrations orders of magnitude higher than the activity of the atmospheric moisture—an obvious impossibility unless isotopic fractionation mechanisms higher than any yet reported were to be discovered.

### Estimates of Dose Potentials Following the Rulison Flaring

The results presented in the previous section are not definitive enough to construct a highly accurate model of the movement of tritium through human food chains, but they do indicate that food chain contamination by THO is much less of a problem than has been predicted. Under conditions of considerable plume meandering, it may perhaps be insignificant. The purpose of this section is to develop a realistic, but simple model to predict the dose to man from the release of THO vapor. Doses arising from exposure to  $^{85}\text{Kr}$  will also be calculated. Emphasis is placed on situations similar to the flaring at Project Rulison.

The first consideration in predicting doses due to the release of radioactivity from an elevated stack is the prediction of the maximum or average downwind concentration of the effluent. The usual approach is to make many meteorological measurements at each location and use this as input to diffusion equations. A simpler approach to the problem has been suggested by Barry (38) and by Bryant (39). This approach uses a parameter known as the weighted mean dispersion constant (or dilution factor) to predict the average air concentration downwind using the equation

$$\bar{C} = \bar{K}Q$$

where  $\bar{C}$  is the average concentration in activity/ $\text{m}^3$ ,  $\bar{K}$  the weighted mean dispersion constant in  $\text{sec}/\text{m}^3$ , and  $Q$  the release rate in activity/sec. If we limit ourselves to the downwind distance where  $\bar{C}$  is maximum, it has been shown that the value of  $\bar{K}$  depends to a significant extent only upon the effective stack height of release. The effect of prevailing winds and differences in geographical location are relatively unimportant in the determination of  $\bar{K}$ , and introduce variations no greater than that associated with the accuracy of the meteorological models themselves (38, 39).

Values of  $K$  actually measured at the Rulison site in locations where maximum concentrations were predicted are shown in Table 7. 337 determinations were made. These data may be used to calculate a parameter essentially similar to the weighted mean dispersion constant,  $\bar{K}$ . Results give a value of  $\bar{K}$  varying between 1 to  $2 \times 10^{-8}$  sec/m<sup>3</sup> depending upon how the values of  $K$  less than the detection limit are treated. This is in good agreement with the theoretical calculations of Bryant (39) if the effective stack height is considered. The effective stack height at Rulison was calculated to be 2700 ft for a flaring rate of  $2 \times 10^7$  ft<sup>3</sup>/day (21).

The source term at Rulison over the eight-day, high-rate production test was  $1 \times 10^9$  pCi of <sup>3</sup>H/sec (2, 24). By using  $\bar{K} = 1.5 \times 10^{-8}$  sec/m<sup>3</sup>, an average air activity of 15 pCi/m<sup>3</sup> may be calculated and used to predict inhalation and submersion doses and as a starting point for food chain dose calculations. The <sup>85</sup>Kr source term over the same period was  $6 \times 10^8$  pCi/sec, which gives an average air activity of 9 pCi/m<sup>3</sup>.

#### Doses from <sup>85</sup>Kr Exposure

The dose resulting from immersion in <sup>85</sup>Kr has recently been considered in detail by Hendrickson (40), who has pointed out that previous estimates using methods (41) recommended by the International Commission on Radiological Protection (ICRP) have overestimated the <sup>85</sup>Kr whole body dose by a factor of 70.

Hendrickson calculates that immersion in air with an <sup>85</sup>Kr concentration of  $3 \times 10^{-7}$  μCi/cm<sup>3</sup> will deliver the following doses: 7 mrem/year to the whole body, lens of the eye, and gonads; 12 mrem/year to the internal surface of the lung; 300 mrem/year to the shallowest layer of live skin; and 500 mrem/year to the skin surface. These values may be used to calculate the dose from any <sup>85</sup>Kr exposure if the time integrated

concentration of  $^{85}\text{Kr}$  air activity is known.

Food chain contamination by  $^{85}\text{Kr}$  usually has not been considered. Quantitative justification was recently provided by Voilleque, et al (42) who measured the transfer of  $^{85}\text{Kr}$  from air to grass. They reported a deposition velocity of  $2.3 \times 10^{-11}$  cm/sec under their experimental conditions and stated that a value of  $10^{-10}$  cm/sec can be used as a conservative upper limit under other meteorological conditions.

#### Doses from HTO Vapor Exposure

Inhalation and immersion doses for exposure to HTO vapor are also easily calculated. The internal dose is two orders of magnitude more important than the immersion dose to the skin (41), so the latter is neglected. Body burdens during exposure are calculated by

$$q(t) = \frac{I}{\lambda_e} (1 - e^{-\lambda_e t})$$

and after exposure ceases at time T by

$$q(t') = q(T)e^{-\lambda_e t'}$$

where  $q(t)$  is the time-dependent body burden,  $I$  is the intake rate,  $\lambda_e$  is the effective elimination rate, and  $t'$  is  $(t - T)$ . The intake rate is the product of the air activity, the breathing rate, and a factor to account for additional absorption through the skin.

Doses arising through food chain contamination are much more difficult to evaluate; the failure of the usual approaches has already been discussed. As a reasonable alternative based upon the data presented above, the following model was developed. It is not completely accurate, but represents a substantial improvement.

The first assumption is that the vegetation water instantaneously reaches the same THO concentration as the atmospheric water. The instantaneous uptake assumption is reasonable considering the rapid turnover rates demonstrated in Figs. 3 and 4. The assumption of equal concentration at equilibration, however, is probably quite conservative. Once exposure ceases, THO in vegetation water is assumed to be lost with a half-time of one day. This is probably also conservative, but only data on short exposures are now available. The relative magnitude of THO contained in the second and third components (Figs. 3 and 4) is very likely a function of exposure time, although this has yet to be adequately demonstrated.

The above provides sufficient input to calculate doses from ingestion of vegetables. For the forage-cow-milk pathway, the concentration of  $^3\text{H}$  in milk for continuous exposure is given by

$$C_m(t) = \frac{f_m(\text{MPD})A}{\lambda_m} (1 - e^{-\lambda_m t})$$

and after exposure ceases at time  $T$ , assuming that the additional intake of THO is negligible compared to the THO reservoir in the cow, by

$$C_m(t') = C_m(T)e^{-\lambda_m t'}$$

where

$C_m(t)$  = Activity in milk, pCi/l

$f_m$  = Fraction of THO body burden secreted in milk,  $l^{-1}$

(MPD) = Cow's daily intake of forage water, ml/day

$A$  = Forage (and atmospheric)  $^3\text{H}$  concentration during exposure, pCi/ml

$\lambda_m$  = Rate constant of THO removal from milk,  $\text{day}^{-1}$

These two equations provide the necessary input to calculate the dose to man via the forage-cow-milk pathway. Because the THO concentration in milk is the same in the cow's body water (43), these equations may also be used to calculate the dose via the forage-cattle-meat pathway. Black, et al (44) have demonstrated that the body water elimination rate is essentially the same in lactating dairy cows, non-lactating dairy cows, and in young steers and heifers. The size of the body water pool may, however, be quite different (44). For THO, the parameter  $f_m$  (or fraction secreted per liter of milk) is obviously the reciprocal of the cow's body water pool. As beef cattle weigh less than dairy cows and also contain less body water as a fraction of body weight (44, 45) it will be assumed that the value of " $f_m$ " for beef cattle is twice that of dairy cows.

The numerical values of the parameters necessary to make the THO dose calculations are given in Table 8. The relative biological effectiveness (RBE) for  $^3\text{H}$  beta particles is taken to be 1.0 in accordance with the latest ICRP recommendations (52). The conservative assumptions are made that cattle are always on fresh forage; that all milk, meat, vegetables, and fruit are produced in the exposed area and consumed immediately following harvest; that all such foods are 100% water; and that the dose is delivered only to the body water compartment. This conservatism is balanced by the neglect of the organically bound tritium in forage, milk, vegetables, fruit, meat and human tissue; and by neglecting that such organically bound tritium would have a much slower elimination rate (20).

#### Results of Dose Calculations

Dose potentials were calculated for the 8-day, high-rate production flaring for Project Rulison using the models discussed above. During this period the absolute humidity in the Rulison area averaged  $3 \text{ ml/m}^3$  (53).

and this was used to convert the average THO activity of  $15 \text{ pCi/m}^3$  to the THO activity of atmospheric moisture. The results of these calculations are shown in Table 9. The total whole body dose to the infant,  $3 \times 10^{-6} \text{ rem}$ , is lower by a factor of  $10^4$  than our preflaring prediction of the dose via the forage-cow-milk pathway alone (20).

The actual doses received by residents in the Rulison area cannot, of course, be verified. It is useful, however, to calculate the levels of THO activity that our model would predict and compare these to the available observations. The model predictions for  $^3\text{H}$  activity in the food chains and in man are given in Table 10. The activity in atmospheric moisture would be expected to vary widely due to variable meteorological conditions. The level of  $5 \text{ pCi/ml}$  is actually the average of many measurements as previously discussed. The highest atmospheric moisture level observed was  $38 \text{ pCi/ml}$ , which was recorded during the calibration flaring preceding the 8-day, high-volume production flaring. During and immediately after the production flaring, the Environmental Protection Agency (EPA) collected 20 urine samples from local residents, 7 animal tissue samples, 8 milk samples, and 14 vegetation samples (2, 54). The only samples above background levels ( $\sim 1 \text{ pCi/ml}$ ) were 4 vegetation samples with the highest value  $1.9 \text{ pCi/ml}$  above background. We collected 6 vegetation samples and 27 milk samples (55); none contained tritium levels above background at the 5% confidence level. Within the limits of the available data, the conclusion can be made that the dose estimates in Table 9, although very small, are realistic upper limits of the doses actually received by residents in the vicinity of the Rulison wellhead. Doses much larger than those predicted here would have been signified by readily detectable  $^3\text{H}$  concentrations in environmental samples.

### Washout of THO Vapor by Rain

The potential doses that might arise from the deposition of THO in a liquid form due to the washout of THO vapor by rain were not included in Table 9. Chamberlain and Eggleton (56) have considered this problem from a theoretical viewpoint, and concluded that at downwind distances equal to a few effective stack heights the specific activity of rain reaching the ground is less than the specific activity of water vapor in ground-level air. The data shown in Fig. 5 demonstrate that THO deposited as rainout is very rapidly lost as is the THO deposited by "dry deposition". Similar results have been reported by Kirchmann, et al (57).

Several hundred precipitation samples were collected during the total Rulison flaring operation (2, 3, 29, 54, 55). The results are consistent with the predictions of Chamberlain and Eggleton. High levels (up to 5100 pCi/ml) were observed on-site; at 0.2 miles the highest reported value was 16 pCi/ml and at 1 mile 5.0 pCi/ml. Nearly all samples collected beyond 0.5 miles did not contain THO above background levels. The additional possible dose potential due to the washout of THO vapor, therefore, appears to be of minor significance except at distances very close to the wellhead.

### Extrapolation of Results to a Commercial Utilization

Also shown in Table 9 are the calculated doses if it is assumed that the same rate of radioactivity release observed during the Rulison 8-day, high-rate production flare had continued for one year. This did not occur at Rulison, but for tritium it is a close approximation to the release rate to be expected during the first year if a field of nuclearly-stimulated natural gas wells were used to provide fuel for a 1000 MWe electrical generating plant (58).

The release rate for  $^{85}\text{Kr}$  would be about 7 times higher than occurred during the 8-day Rulison flare. The quantity of natural gas required per day would be about  $2 \times 10^8 \text{ ft}^3$  — an order of magnitude greater flow rate than achieved at Rulison. Under these conditions, the effective stack height would be somewhat higher than at Rulison, and the whole body doses shown in Table 9 should be upper limits of the dose expected from the operation of such a power plant. The expected average concentration of tritium ( $10 \text{ pCi/cm}^3$ ) during the first year would decline over the first few years and stabilize in the sixth year at levels about an order of magnitude lower (58).

If natural gas with the same tritium concentration were to replace the normal natural gas supply for the Los Angeles Basin or the San Francisco Bay Area, the weighted average dose via inhalation and absorption alone would be  $5 \times 10^{-3} \text{ rem/year}$  (17), compared to the value of  $2.1 \times 10^{-4} \text{ rem/year}$  for the infant via all pathways (Table 9) if the natural gas is used to produce electricity.

FOOTNOTE

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Table 1. Predicted inventory of radionuclides of noble gases or chemically reactive species that will form gaseous species for Project Rulison 180 days post detonation. (From Ref. 19.)

Nuclide	Half-Life	Inventory (curies)
$^3\text{H}^a$	12.3 y	$10^3$ to $10^4$
$^{14}\text{C}^a$	5770 y	$10^{-2}$ to $10^{-3}$
$^{37}\text{Ar}^a$	34.3 d	$10^1$ to $10^2$
$^{39}\text{Ar}^a$	260 y	2 to $2 \times 10^1$
$^{85}\text{Kr}$	10.8 y	$9.6 \times 10^2$
$^{133}\text{Xe}$	5.3 d	$8.6 \times 10^{-4}$

<sup>a</sup> Produced wholly or partly by neutron activation, and subject to greater errors in prediction.

Table 2. Tritium and  $^{85}\text{Kr}$  activity in the natural gas and water produced from the Gasbuggy and Rulison wells, and predicted activity in natural gas based upon the parameters of a new nuclear device specifically developed for gas stimulation.

	Gasbuggy	Rulison	Future <sup>a</sup> (est.)
Tritium			
Total in gas (Ci)	2000 <sup>b</sup>	1300 <sup>f</sup>	
% in gas	5 <sup>b</sup>	13 <sup>g</sup>	25
Early concentration in gas (pCi/cc)	620 <sup>c</sup>	180 <sup>h</sup>	11
Early concentration in H <sub>2</sub> O (pCi/ml)	$> 1 \times 10^6$ <sup>d</sup>	$4 \times 10^5$ <sup>j</sup>	
$^{85}\text{Krypton}$ (100 % in gas)			
Total in gas (Ci)	350 <sup>b</sup>	1110 <sup>k</sup>	
Early concentration in gas (pCi/cc)	100 <sup>c</sup>	150 <sup>h</sup>	71
Yield (kt)	29 <sup>e</sup>	43 <sup>k</sup>	$4 \times 100$

<sup>a</sup> Ref. 6. These concentrations are averaged over a year, or  $5 \times 10^9 \text{ ft}^3$  of gas. They are not therefore directly comparable to the Gasbuggy and Rulison figures.

<sup>b</sup> Ref. 27.

<sup>c</sup> Ref. 23. Based upon five samples taken during the first flow test ( $5 \times 10^6 \text{ ft}^3/\text{day}$ ).

<sup>d</sup> Ref. 23.

<sup>e</sup> Ref. 1.

<sup>f</sup> Calculated from Ref. 24 using corrected  $^{85}\text{Kr}$  value from Ref. 28.

<sup>g</sup> Calculated assuming  $10^4 \text{ Ci}$  were produced.

<sup>h</sup> Ref. 24. Based on six samples taken during the calibration flaring period.

<sup>j</sup> Ref. 2.

<sup>k</sup> Ref. 28.

Table 3. Representative results of off-site tritium activities measured during the Project Rulison calibration and high-rate production flaring periods. All stations (1-15) are on the B route shown in Fig. 1. Results are expressed in pCi, m<sup>3</sup> of air.

Time	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15
8/22 0640-1034	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
0/04 0800-0935	-	-	-	-	-	50	-	-	-	-	12	-	46	-	170
0/05 0615-1028	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
0/07 0516-1206	-	-	-	-	-	-	-	-	-	-	-	-	-	19	-
0/28 0603-1106	3.5	-	6.6	9.1	-	9.1	-	7.1	-	16	27	12	18	11	8.8
0/28 1930-0708	-	-	-	-	-	-	-	-	-	-	16	-	7.8	-	-
0/29 0610-1050	6.5	-	-	5.1	5.1	5.1	4.4	5.8	4.6	5.8	5.6	6.5	10	-	5.1
0/30 0608-1114	-	-	-	-	5.8	-	-	-	5.8	9.0	6.9	-	-	4.5	-
0/31 1330-1747	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
0/31 1622-0949	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
1/01 0806-1341	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
1/02 0553-1024	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
1/03 0609-1030	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-

- Sampled, <sup>3</sup>H not detected.

Table 4. Tritium activity in pCi/m<sup>3</sup> of air measured on-site during the Project Rulison calibration and high-rate production flaring periods. All stations (1-9) were within a few hundred feet of the flare stack.

Time	1	2	3	4	5	6	7	8	9
8/18 0712-0850	-								
8/22 1029-1118	-								
10/04 0714-0916	79	170							
10/05 0602-0930	53	180	3,000	43,000	11,000	22,000			80
10/05 0845-1100	290	290	2,000						
10/27 0842-1200	86	89	49	25	52	20			35
10/27 1145-1503	84	220	37	27	47	17	12		21
10/28 0719-1036	190	180	240	210	170	190	140		
10/28 1019-1053	86	53	44	77	58	53	26		560
10/29 0735-1015	150	130	170	140	110	120	110		180
10/29 1315-1100				94					
10/30 1100-1358	21			81					18
10/30 1350-1528	16			120					
10/31 1310-2005	15			70	36			14	16
10/31 1945-0903	15			180	38			14	14
11/01 0830-1615	20			88	35			18	12
11/01 1700-0940	76			50	21			73	65
11/02 0930-1645	11			93	56			8	5
11/02 1640-0855	43			53	25				
11/03 0855-1405	220			210	170				

- Sampled, <sup>3</sup>H not detected.

Table 5. Results of measurements at Project Rulison of the fraction of tritium (THO) retained on vegetation and the "dry deposition" of tritium (THO). The term "deposition" is used rather than "deposition velocity" because a true rate constant was not derivable from the data (see text). Data do not include the tritium that may be organically bound.

EXPOSURE TIME (hr)	INTEGRATED AIR ACTIVITY ( $\mu\text{Ci-sec}/\text{m}^3$ )	NUMBER	FRACTION ON GRASS	FRACTION ON GRASS DENSITY OF GRASS ( $\text{cm}^2/\text{g}$ )	DEPOSITION* (cm/sec)
23	7.7	8	-----	-----	0.37
27	13.	2	0.29	3.0	0.20
27	3.6	1	0.38	4.1	0.21
27	1.6	2	0.14	1.7	0.35
27	1.5	2	0.14	1.9	0.47
AVERAGE		7 OR 15	0.22 (0.02-0.50)	2.5(0.5-2.9)	0.32(0.20-0.58)
44.7	12.	2	0.30	4.0	0.18
51	18.	2	0.32	2.2	0.15
51	6.3	2	0.54	5.2	0.16
51	6.4	2	0.19	2.4	0.12
51	6.0	2	0.28	2.6	0.14
AVERAGE		8	0.32(0.08-0.85)	3.3(1.3-6.8)	0.15(0.10-0.29)

\*DEPOSITION:  $\frac{\text{pCi}}{\text{cm}^2} \times \frac{\text{cm}^3}{\text{pCi-sec}}$

Table 6. Summary of the parameters derived from the experiments shown in Figs. 3-5.

Magnitudes of the components for the liquid application were calculated assuming a short component does not exist.

Condition	$\frac{A_1}{A_0}$	$T_{1/2}^1$	$\frac{A_2}{A_0}$	$T_{1/2}^2$	$\frac{A_3}{A_0}$	$T_{1/2}^3$
<b>Vapor Exposure</b>						
0.5 hr Burclover	0.98	0.8 hr	0.02	25 hr		
0.5 hr Fiddleneck	0.997	0.7 hr	0.003	24 hr		
1.0 hr Burclover	0.908	0.9 hr	0.091	17 hr	0.002	270 hr
<b>Liquid Application</b>						
Burclover (2 mm rain)	?	?	(0.85)	10 hr	(0.15)	100 hr

Table 7. Distribution of dispersion constants measured during Project Rulison calibration and high-rate production flares. All measurements were made at distances where maximum downwind concentrations were expected.

Date	Detection Limit for $K^*$ (sec/m <sup>3</sup> )	Number of observations falling within interval:						
		Less than Detection Limit	$1 \times 10^{-9}$ to $5 \times 10^{-9}$	$5 \times 10^{-9}$ to $1 \times 10^{-8}$	$1 \times 10^{-8}$ to $5 \times 10^{-8}$	$5 \times 10^{-8}$ to $1 \times 10^{-7}$	$1 \times 10^{-7}$ to $5 \times 10^{-7}$	$5 \times 10^{-7}$ to $1 \times 10^{-6}$
8/18/70	$\sim 1 \times 10^{-9}$	5	1					
8/22/70	$\sim 5 \times 10^{-9}$	17	xx		10			
10/04/70	$\sim 2 \times 10^{-8}$	24	xx	xx	6	1	3	1
10/05/70	$\sim 1 \times 10^{-8}$	51	xx	xx	4	1	1	
10/07/70	$\sim 1 \times 10^{-8}$	43	xx	xx	1			
10/27/70	$\sim 1 \times 10^{-9}$	13		2	2	1		
10/28/70	$\sim 1 \times 10^{-9}$	5	10	8	9			
10/29/70	$\sim 1 \times 10^{-9}$	14	15	1	2	1		
10/30/70	$\sim 2 \times 10^{-9}$	10	4	1				
10/31/70	$\sim 3 \times 10^{-9}$	28						
11/01/70	$\sim 3 \times 10^{-9}$	14						
11/02/70	$\sim 2 \times 10^{-9}$	14						
11/03/70	$\sim 1 \times 10^{-9}$	14						
Totals		252	30	12	34	4	4	1

$$* K = \frac{C}{Q} = \frac{\text{pCi/m}^3}{\text{pCi/sec}}$$

xx Interval below detection limit.

Table 8. Numerical values of the parameters necessary to calculate doses from the exposure to THO.

<u>PARAMETERS</u>	<u>VALUE</u>	<u>REF.</u>
Breathing rate		
Adult	20 m <sup>3</sup> /day	41
Infant	5 m <sup>3</sup> /day	21
Skin absorption factor	2	21, 46
Mass of body water		
Adult	43 kg	41
Infant	6.1 kg	a
Half-times of elimination		
Vegetation	1 day	See text
Cattle, dairy and beef	3.5 days	43, 44, 47
Adult	10 days	46
Infant	3.2 days	21, 48
Fraction of THO body burden secreted in milk	0.002/l	43, 44, 47
Fraction of body water in beef	0.004/kg	See text
Water intake via fresh forage		
Dairy cow	30 kg/day	b
Beef cow	15 kg/day	b
Average <sup>3</sup> H decay energy	0.0063 MeV/dis	49
Food intake <sup>c</sup>		
Milk		
Adult (20-34 years)	260 g/day	50
Infant (<1 year)	700 g/day	50
Meat, poultry, fish		
Adult (20-34 years)	270 g/day	50
Infant (<1 year)	50 g/day	50
Vegetables, fruit, potatoes		
Adult (20-34 years)	400 g/day	50
Infant (<1 year)	220 g/day	50

- a Assumes a 10 kg body weight with the same ratio of body water to body weight as that of an adult.
- b Assumes that a dairy cow needs 14 kg of dry forage per day (51), and that fresh forage is 70% water; assumes beef cattle require half this amount.
- c It is assumed for the purpose of dose calculations that each food category is freshly harvested in the exposed area, and is 100% water.

Table 9. Calculated dose potentials for the 8-day Project Rulison high-rate production flaring and projected dose potentials over a one-year period.

	<u>INFANT</u>		<u>ADULT</u>	
	<u>High-rate flaring</u>	<u>Projected<sup>a</sup> One-year flaring</u>	<u>High-rate flaring</u>	<u>Projected<sup>a</sup> One-year flaring</u>
<sup>85</sup> Krypton				
Inhalation and submersion				
Whole body	$4.6 \times 10^{-9}$ rem	$2.1 \times 10^{-7}$ rem	$4.6 \times 10^{-9}$ rem	$2.1 \times 10^{-7}$ rem
Skin <sup>b</sup>	$2.0 \times 10^{-7}$	$9.0 \times 10^{-6}$	$2.0 \times 10^{-7}$	$9.0 \times 10^{-6}$
Tritium				
Inhalation and submersion	$2.9 \times 10^{-7}$	$1.3 \times 10^{-5}$	$5.2 \times 10^{-7}$	$2.3 \times 10^{-5}$
Milk	$5.5 \times 10^{-7}$	$9.2 \times 10^{-5}$	$1.1 \times 10^{-7}$	$1.5 \times 10^{-5}$
Vegetables, fruit, potatoes	$2.1 \times 10^{-6}$	$9.7 \times 10^{-5}$	$1.7 \times 10^{-6}$	$7.6 \times 10^{-5}$
Meat	$4.0 \times 10^{-8}$	$6.6 \times 10^{-6}$	$1.1 \times 10^{-7}$	$1.5 \times 10^{-5}$
Sum (whole body)	$3.0 \times 10^{-6}$	$2.1 \times 10^{-4}$	$2.4 \times 10^{-6}$	$1.3 \times 10^{-4}$

a Assumes flaring would continue with the same rate of release in Ci/sec as was observed during the 8-day, high-rate production testing.

b Dose calculated for the shallowest layer of live skin.

Table 10. Tritium activity levels predicted by the model through the food chains to man. Results are calculated for the end of the 8-day, high-rate production flaring of the Rulison well.

	<u>Tritium Activity</u>	
	<u>In fant</u>	<u>Adult</u>
Average atmospheric moisture	5	pCi/ml
Forage, vegetation, fruits, potatoes	5	pCi/ml
Milk	1.2	pCi/ml
Beef	1.2	pCi/ml
Man		
Via inhalation and submersion	0.093 pCi/ml	0.086 pCi/ml
Via milk	0.40	0.029
Via meat	0.028	0.031
Via vegetation, fruit, potatoes	0.69	0.29
	<hr/>	<hr/>
SUM	1.2	0.44

FIGURE LEGENDS

- Fig. 1 Off-site locations at Project Rulison where atmospheric moisture was collected and assayed for tritium.
- Fig. 2 Tritium (THO) loss curve from water of vegetation measured at the termination of the Project Rulison high-rate production flaring. Grass plugs (3 7/8" diameter X 4" deep) were brought to the site 44.7 hours before flaring stopped. The curve is a two component exponential fit with half-times of 0.4 and 70 hours, with 25% of the activity associated with the short half-time.
- Fig. 3 Tritium (THO) loss curve for two species following a controlled vapor exposure of 0.5 hours. Results are very similar except that the magnitude of the second component is nearly an order of magnitude greater for burclover
- Fig. 4 Tritium (THO) loss curve following an one hour controlled vapor exposure. The first two components have essentially the same rate constants as following the 0.5 hr. exposure, but the magnitude of the second component is 4.5 times larger and a third component is also evident.
- Fig. 5. Tritium (THO) loss curve following a liquid application equivalent to a 2mm rain. Sampling did not begin soon enough to detect an early component, but rapid turnover is again demonstrated. A diurnal pattern is evident with peaks occurring in the afternoon indicating the effects of transpiration.

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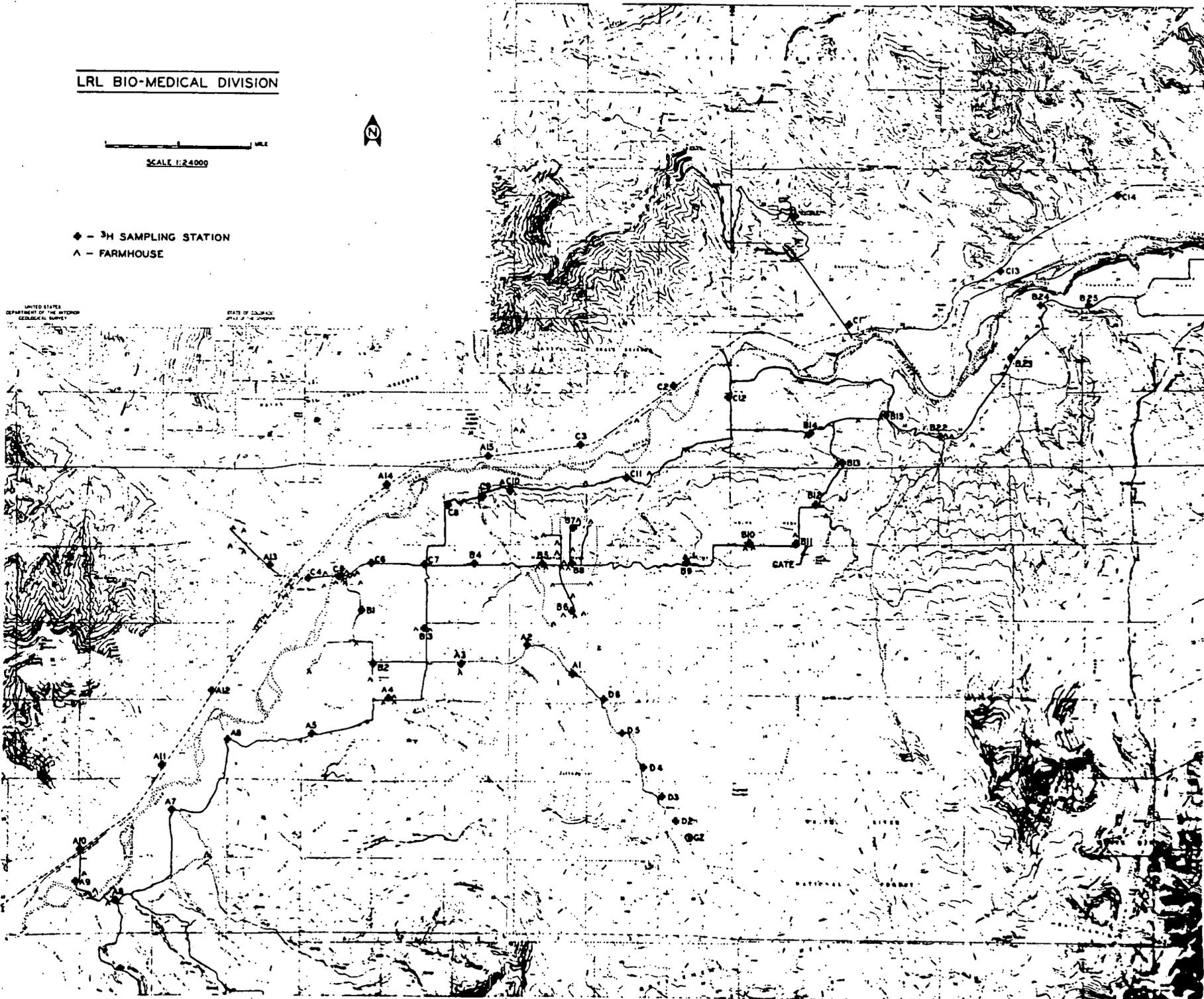
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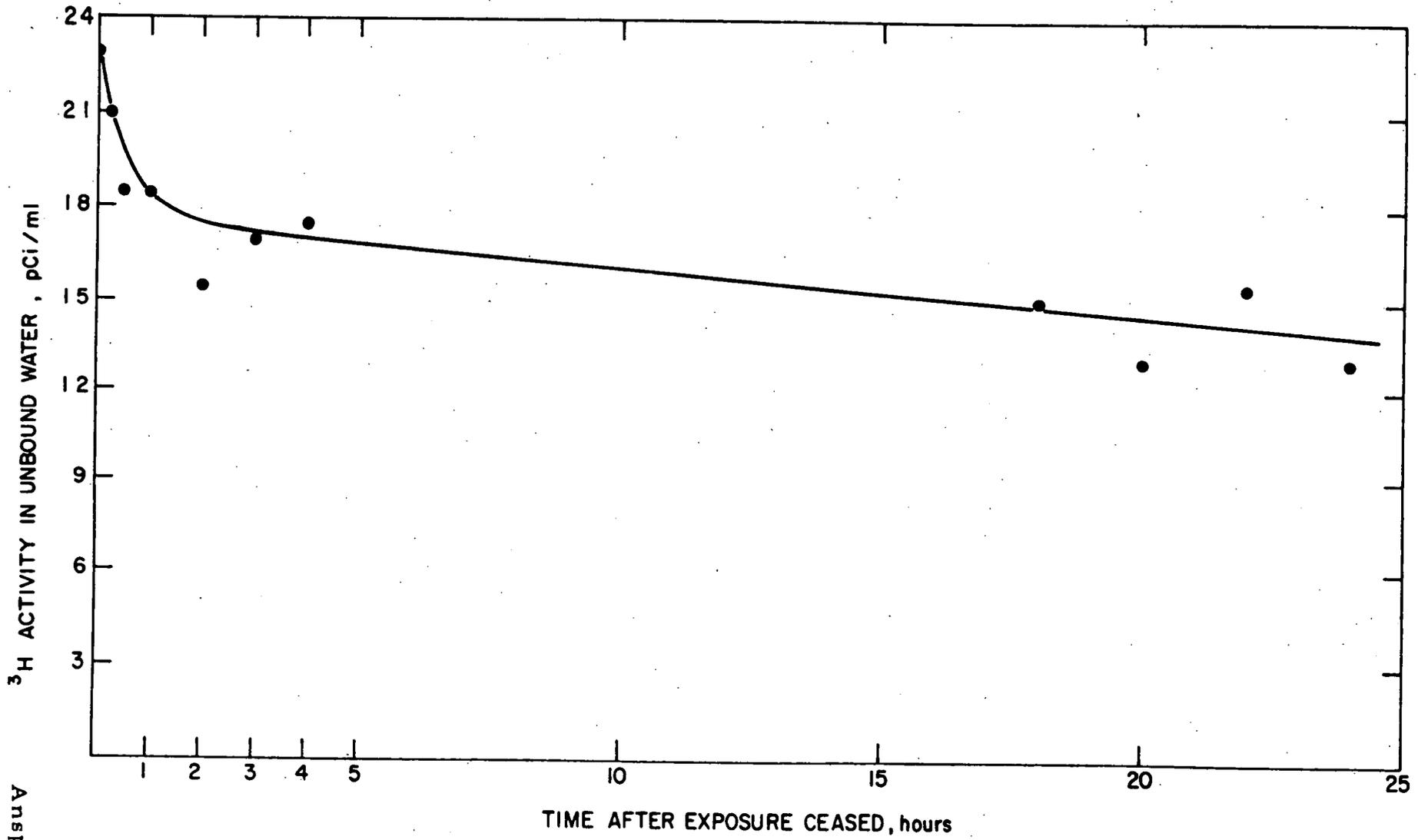
◆ - <sup>3</sup>H SAMPLING STATION  
▲ - FARMHOUSE

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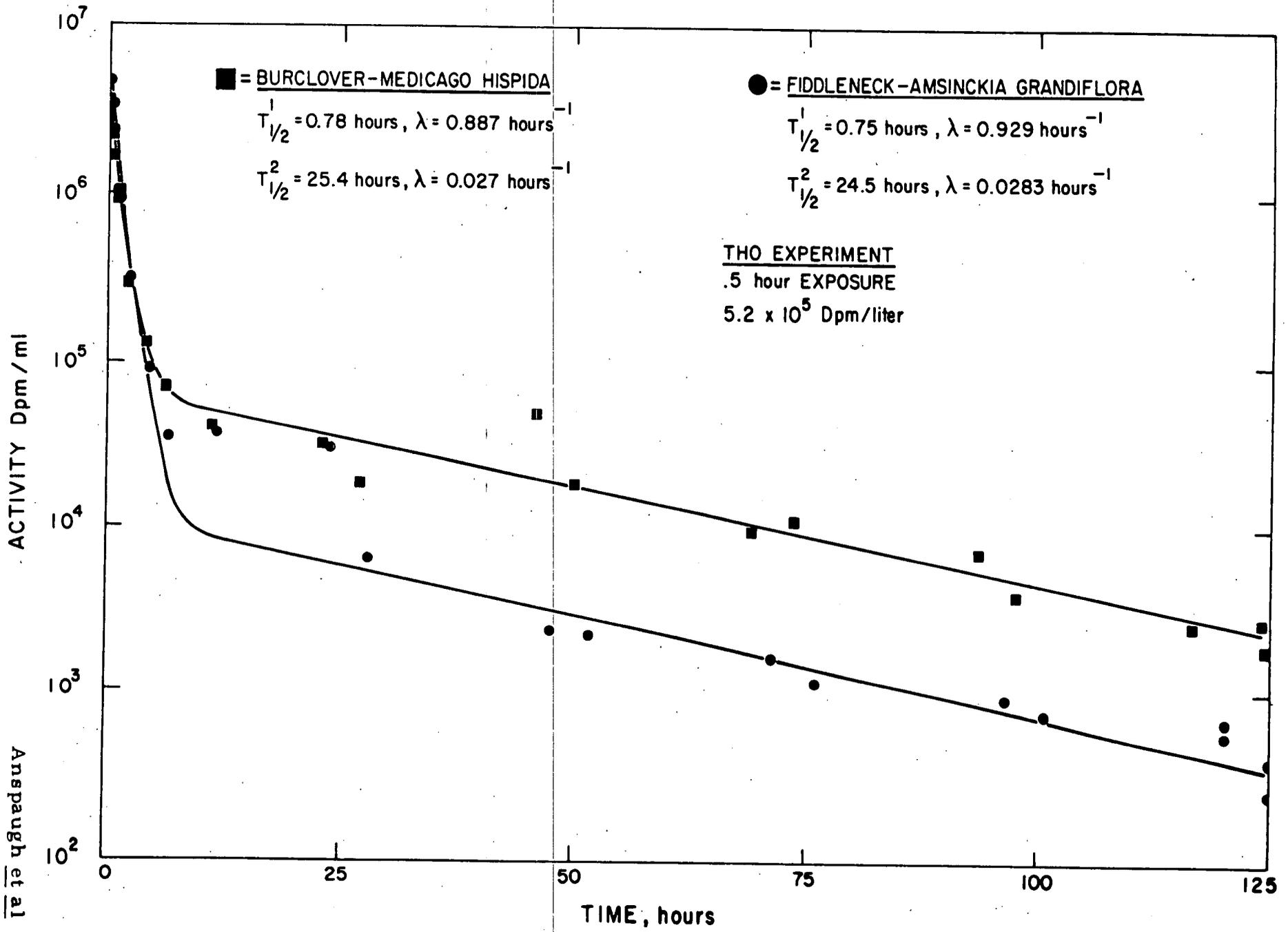
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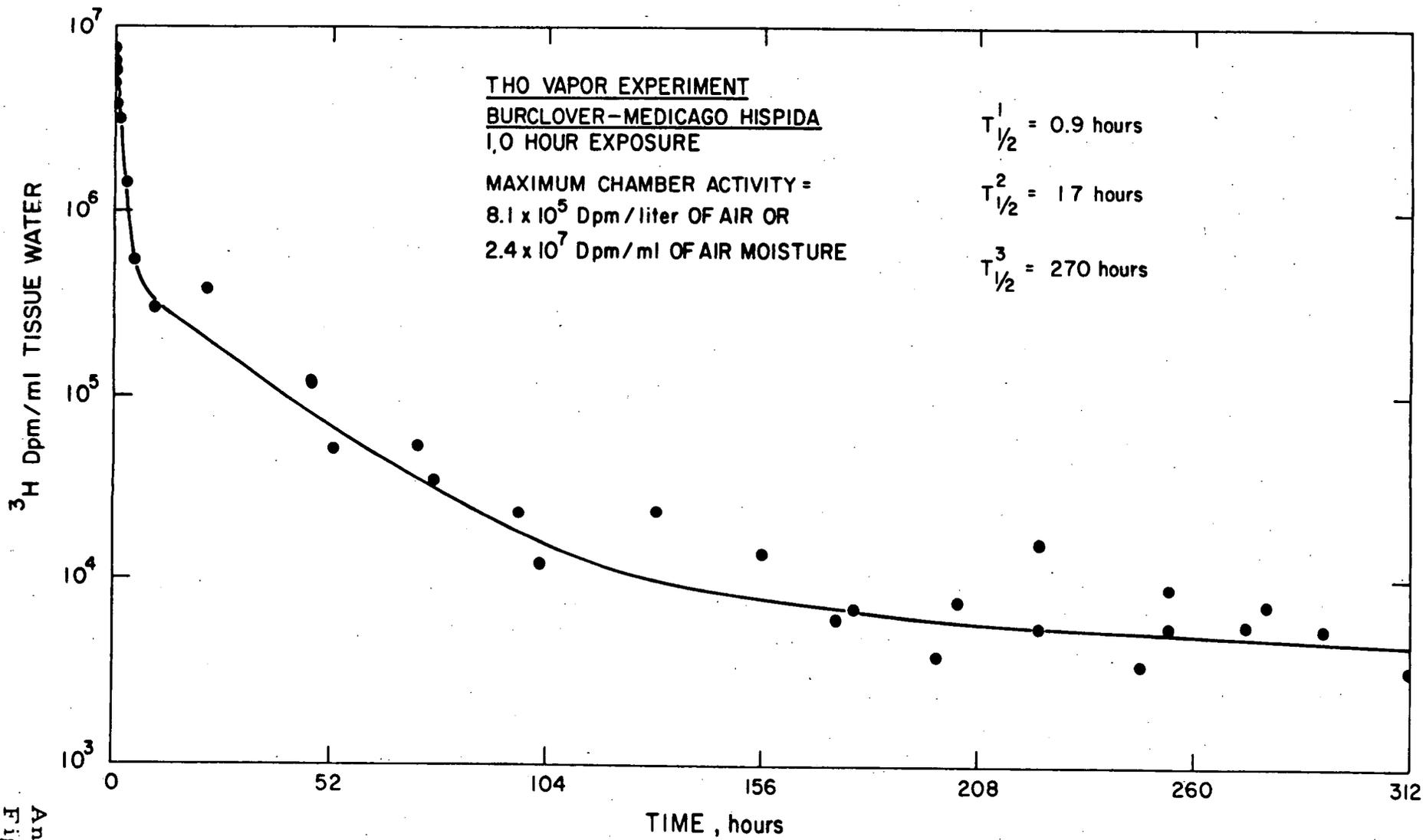




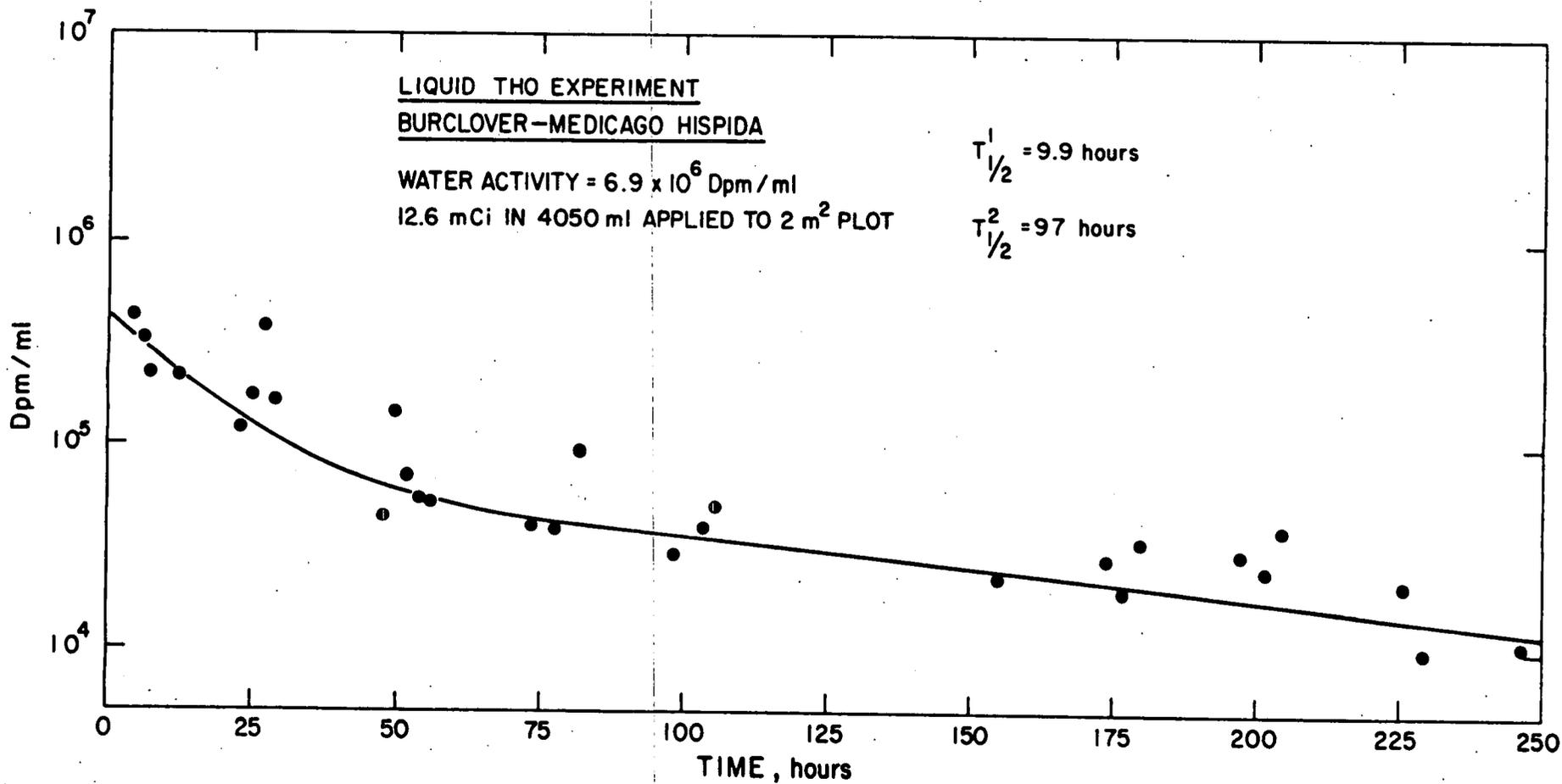
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Fig. 2



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Fig. 4



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 Fig. 5

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