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# A STUDY OF THE DIFFUSION AND MIXING OF TRITIUM GAS IN AIR

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# A STUDY OF THE DIFFUSION AND MIXING OF TRITIUM GAS IN AIR

by

Morris J. Engelke
Edwin A. Bemis, Jr.

LOS ALAMOS NATIONAL LABORATORY
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# ABSTRACT

The mechanism by which small amounts of tritium gas mix with air has been investigated. Interpretations have been applied to the diffusion process for a small-volume source in a homogeneous and stationary atmosphere. Results from strict diffusion considerations indicate that the concentration velocity of tritium in air has a practical maximum value of less than 0.4 cm/sec. However, effective concentration velocities depend almost entirely on the air motion in the room and are as much as three orders of magnitude greater than those indicated by diffusion theory. Therefore, to obtain a representative breathing-zone sample, the detector of a monitoring instrument should be placed as close to a worker's face as possible and in a position to take advantage of the air flow in the room.

# ACKNOWLEDGEMENTS

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Richard L. Henkel assisted with the experiment and reviewed this paper, and Richard D. Hiebert assisted with setting up the instrumentation.

# INTRODUCTION

In the past, location of tritium detectors in relation to the worker was based on the assumption that tritium mixed rapidly with air. The detector position was not considered critical, and distances as great as 20 ft or more between worker and detector were used. The occurrence of personnel exposures when detectors did not alarm caused doubt about the validity of this method of placing detectors. Also, some persons showed no tritium in urine assays, even though a detector indicated that there had been an exposure.

This investigation was made to provide information on the mixing of tritium gas with air and to determine at what distance from the worker a monitoring device should be located to be adequate.

# DIFFUSION THEORY

Much work has been done to try to describe the motions of gases in the atmosphere. Instantaneous and continuous point, plane, and line sources have been considered under varying conditions of wind and of adjacent ground.[1] Of interest here is the case of an instantaneous, small-volume source diffusing outward in a homogeneous and stationary atmosphere. These conditions are assumed to be sufficiently ideal so that classical diffusion theory can be used to describe the motion of the diffusing gas.

Consider a spherical source of infinitesimal dimensions located at $r = 0$ and containing a fixed amount S of gas 1. At $t = 0$, the source is allowed to diffuse into an infinite space filled with stationary gas 2. The diffusion equation which applies, in polar coordinates, is[2,3]

$$\frac{\partial C}{\partial t} = D \frac{\partial^2 C}{\partial r^2} + \frac{2}{r} \frac{\partial C}{\partial r} \quad ,$$

where C = the concentration of gas 1 per unit volume, and D = the diffusion coefficient for gas 1 in gas 2, and is assumed to be constant under fixed conditions of temperature and pressure. The solution of the equation is [2,3]

$$C = \frac{S}{(4\pi Dt)^{3/2}} e^{-(r^2/4Dt)} ,$$

where S = source strength in curies, C is in curies/cm$^3$, D is in cm$^2$/sec, r is in cm, and t is in sec.

The value of the diffusion coefficient for tritium in air is uncertain. Figure 1 shows a plot of the molecular weight of diffusing gas vs the diffusion coefficient of gas in air and in argon at standard temperature and pressure. The values of interest are in the range of 0.1 to 1.0 cm$^2$/sec. Provided the variations of temperature, pressure, and concentration are small, the coefficient can be considered as constant. For a normal range of temperature and pressure, the value of D can be corrected with the relation[4]

$$D = D_o(T/T_o)^n(p_o/p) ,$$

where o refers to STP conditions, and n varies between 1.75 and 2. The variation of D with relative concentration of the diffusing gas[5] is less than 10%.

For convenience, let $\alpha$ = Dt and be in units of cm$^2$. The expression for the concentration then becomes

$$C = \frac{S}{(4\pi\alpha)^{3/2}} e^{-(r^2/4\alpha)} .$$

The values of $\alpha$ which are of practical interest lie in the range of 1 to 1,000. Figure 2 shows concentration vs distance for a family of $\alpha$'s, assuming source strength S = 1 curie. The same data are plotted

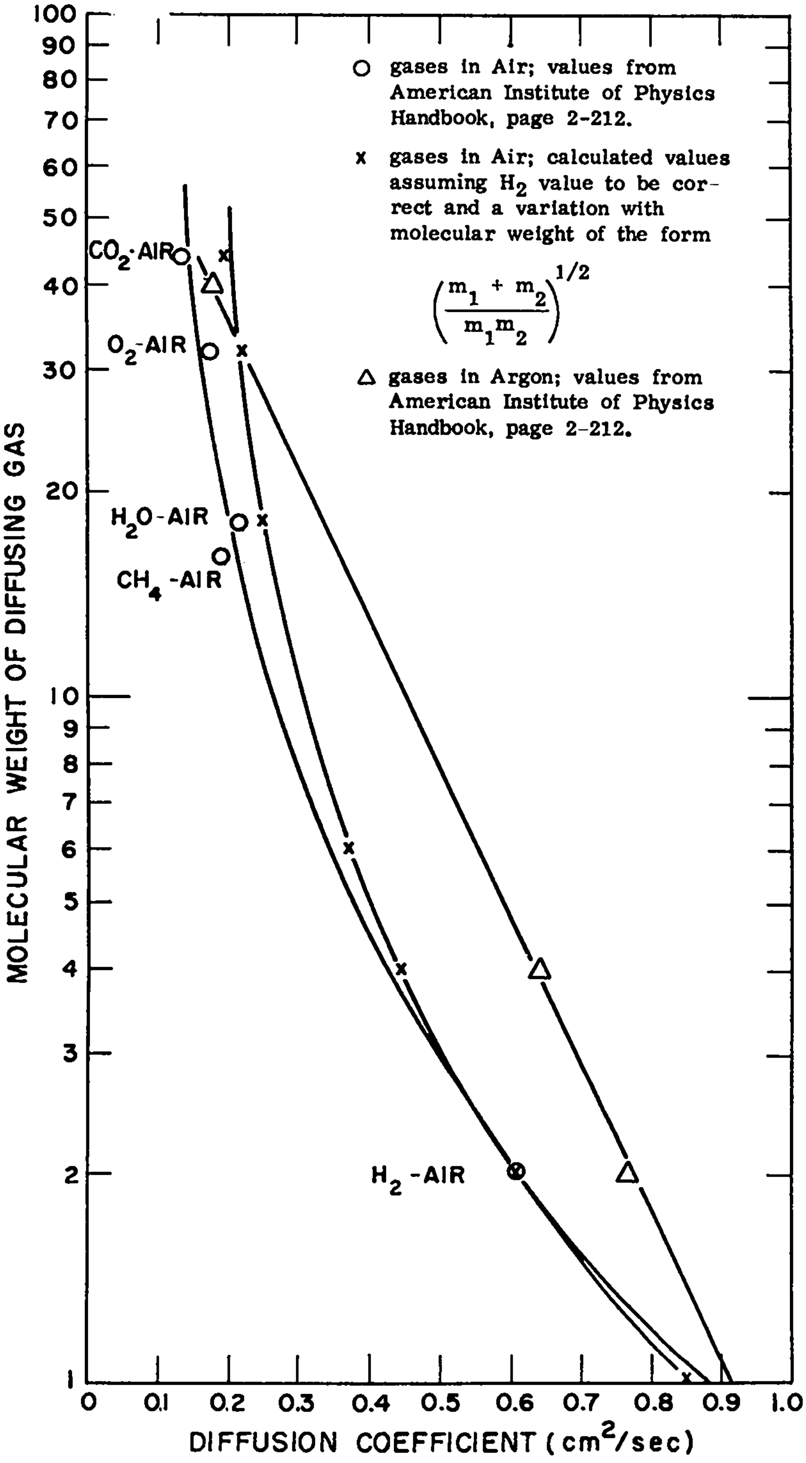


Figure 1. Molecular weight of diffusing gas vs diffusion coefficient in air and in argon.

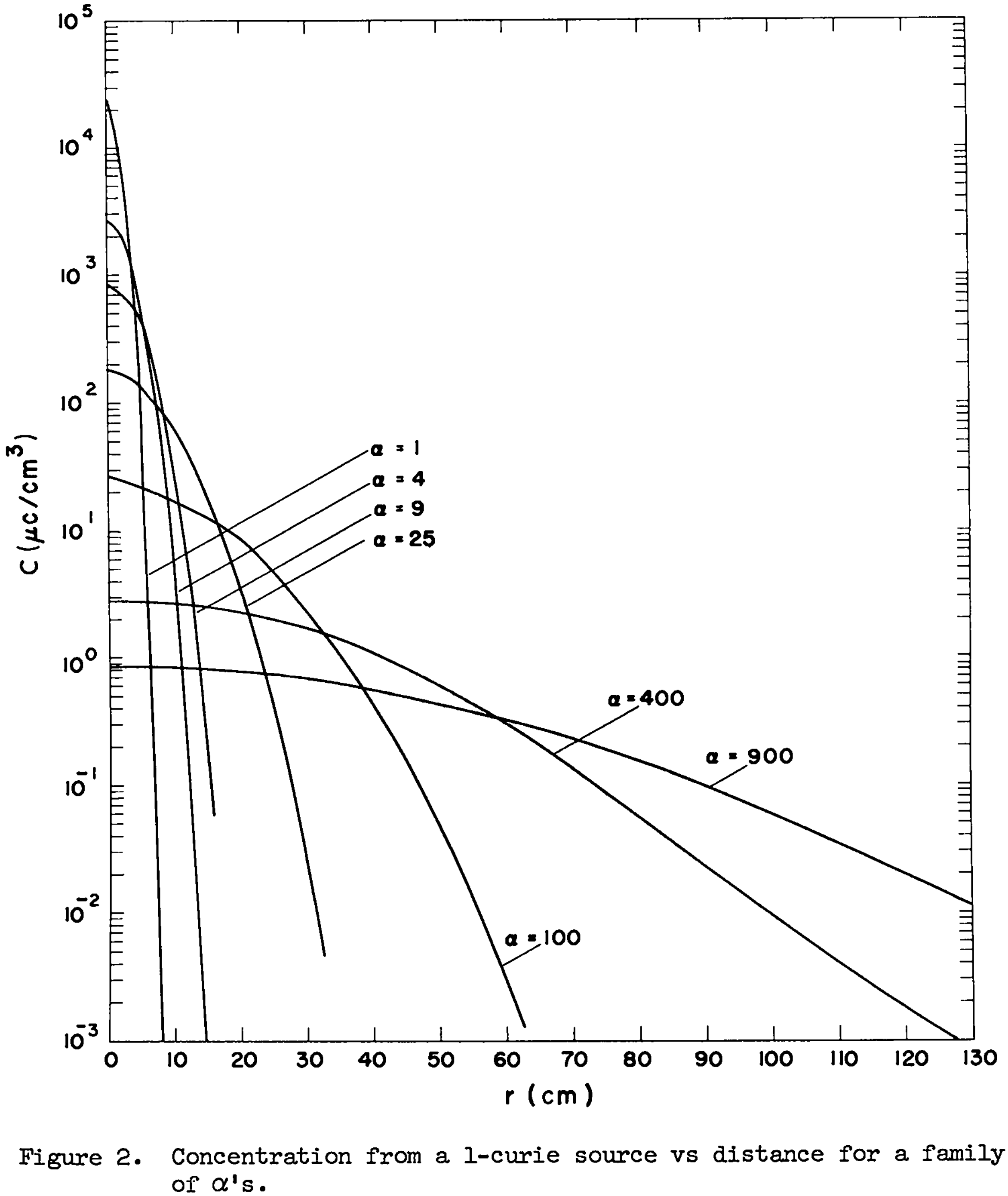


Figure 2. Concentration from a 1-curie source vs distance for a family of $\alpha$'s.

in Figure 3 as concentration vs $\alpha$ for a family of r, and in Figure 4 as $\alpha$ vs r for a family of concentrations.

The utility of these curves lies in the fact that one set suffices for all values of D. To get the curves in units of time, it is only necessary to use an appropriate value for the diffusion coefficient to adjust the numbers on the $\alpha$ coordinates by means of the relation $t = \alpha/D$. As an example, the coordinates marked in Figures 3 and 4 show the time, assuming $D = 0.4$ (a value approximately correct for the diffusion of tritium in air).

A concentration velocity can be defined as the velocity of a small-volume element moving in such a manner as to maintain a constant concentration within it at all times. This velocity $v_c$ can be found by determining the total derivative of the expression for the concentration and equating it to zero. Thus,

$$dC = \frac{\partial C}{\partial \alpha} d\alpha + \frac{\partial C}{\partial r} dr = 0 \quad ,$$

from which

$$\frac{dr}{d\alpha} = \frac{r}{2\alpha} - \frac{3}{r} \equiv \beta \quad .$$

Since $\alpha = Dt$, and D is constant for any two given gases,

$$\frac{dr}{d\alpha} = \frac{dr}{D dt} = \frac{v_c}{D} \quad ;$$

therefore,

$$v_c = D\beta \quad .$$

Curves of $\beta$ vs r and $\alpha$ are given in Figures 5 and 6, which show several values of the concentration with a source strength of 1 curie. The

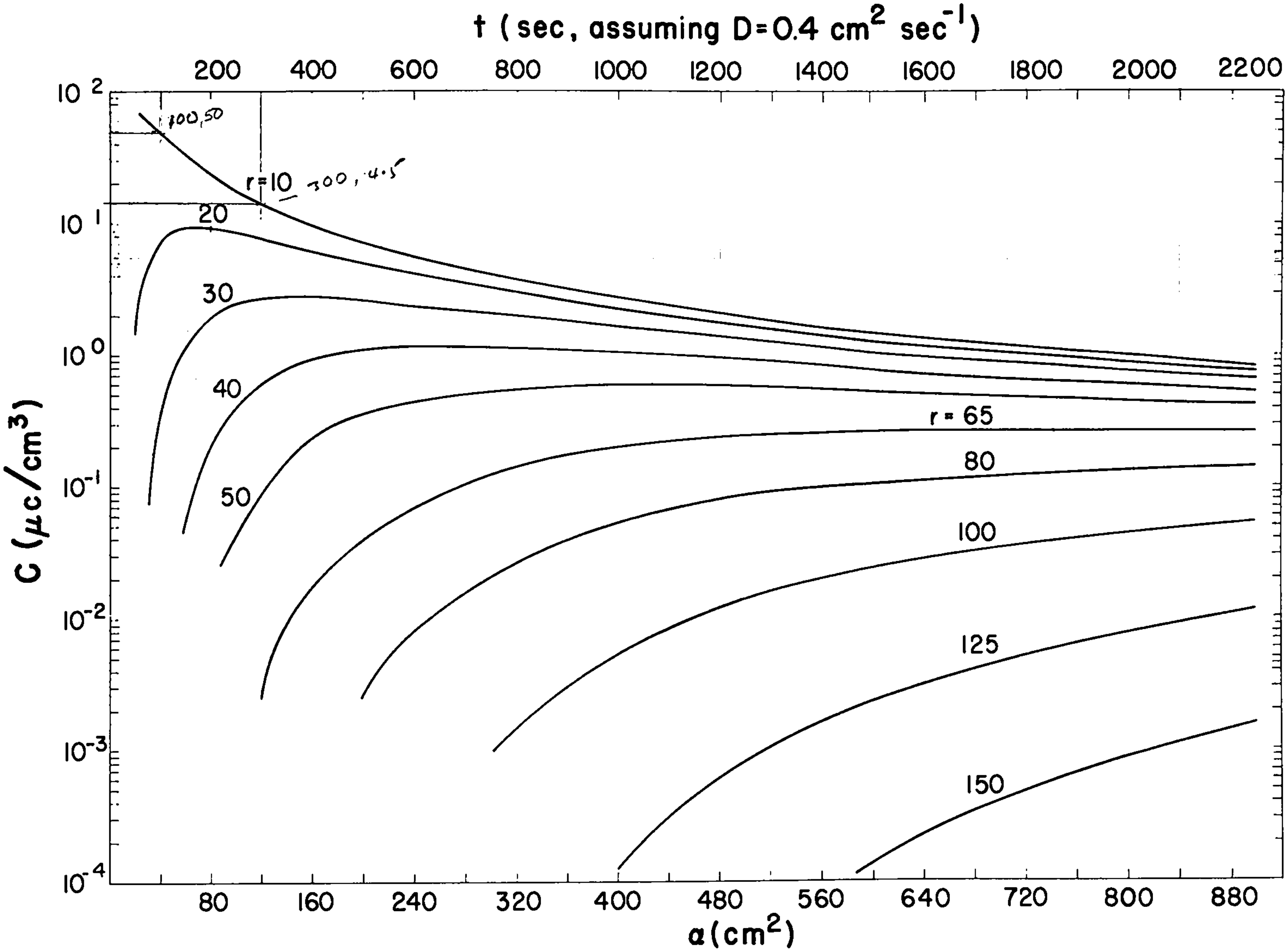


Figure 3. Concentration from a 1-curie source vs α or time for a family of distances from point of release.

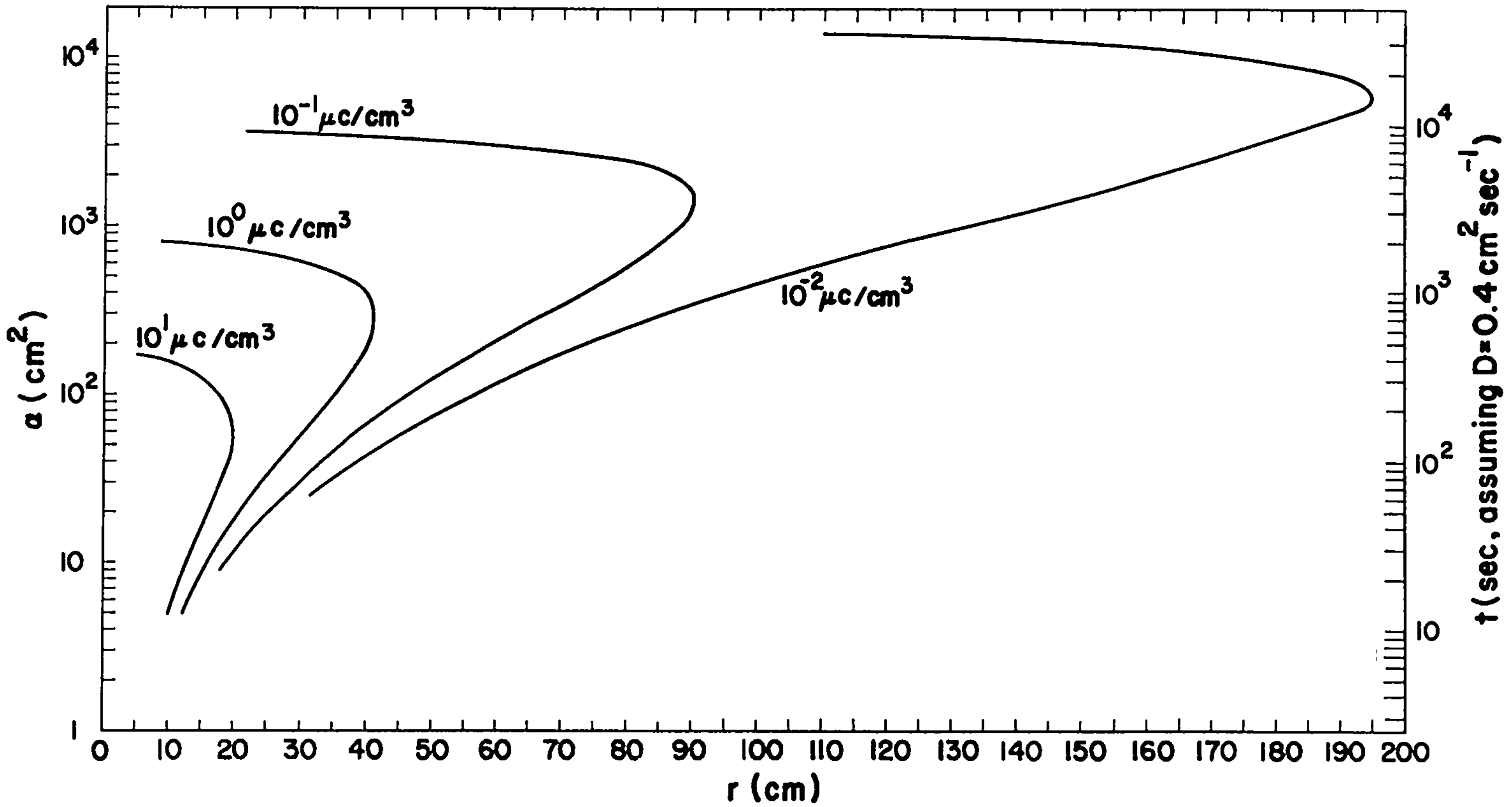


Figure 4. Distance from point of release of a 1-curie source vs $\alpha$ or time for a family of concentrations.

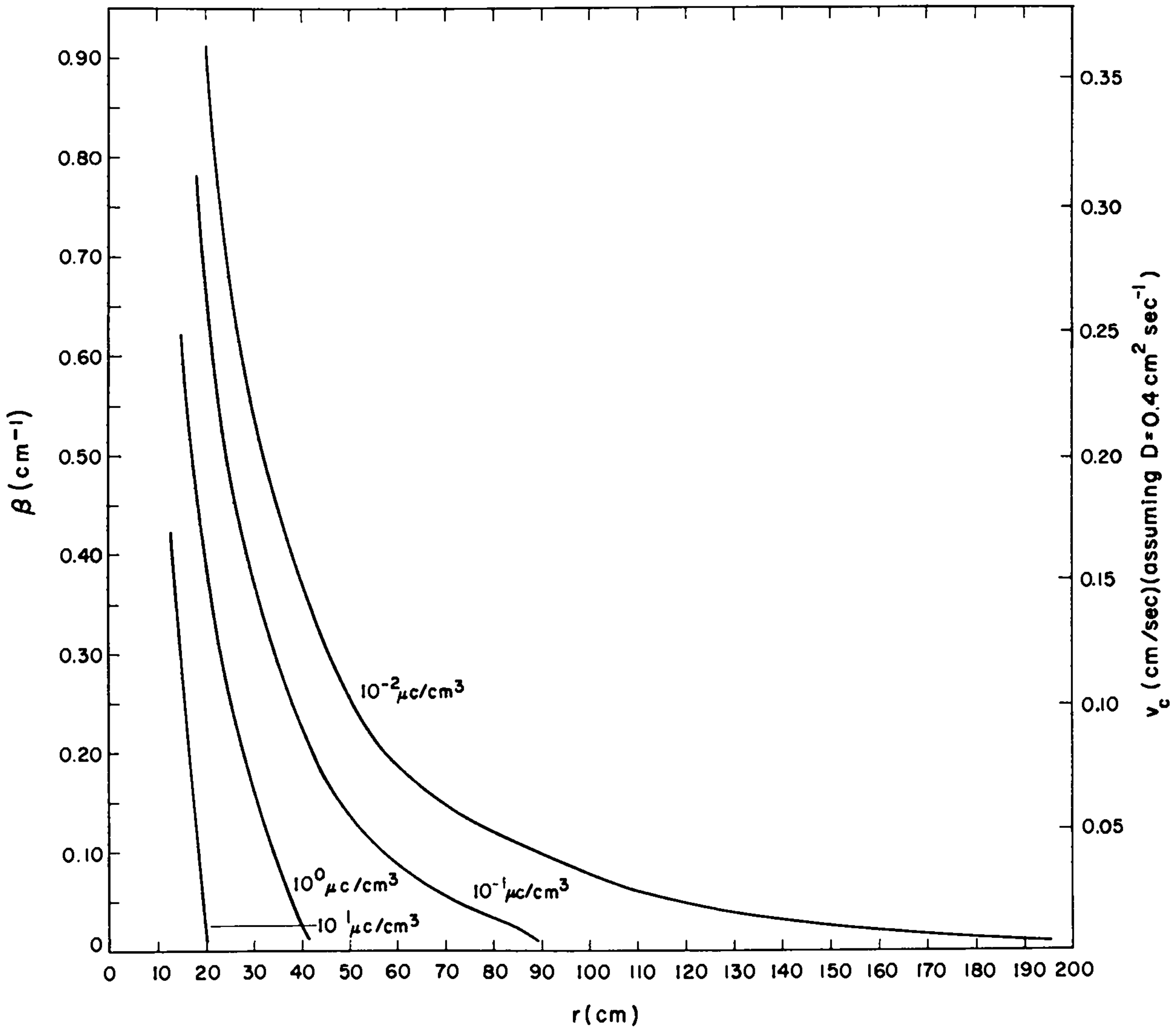


Figure 5. Distance from point of release of a 1-curie source vs β or velocity for a family of concentrations.

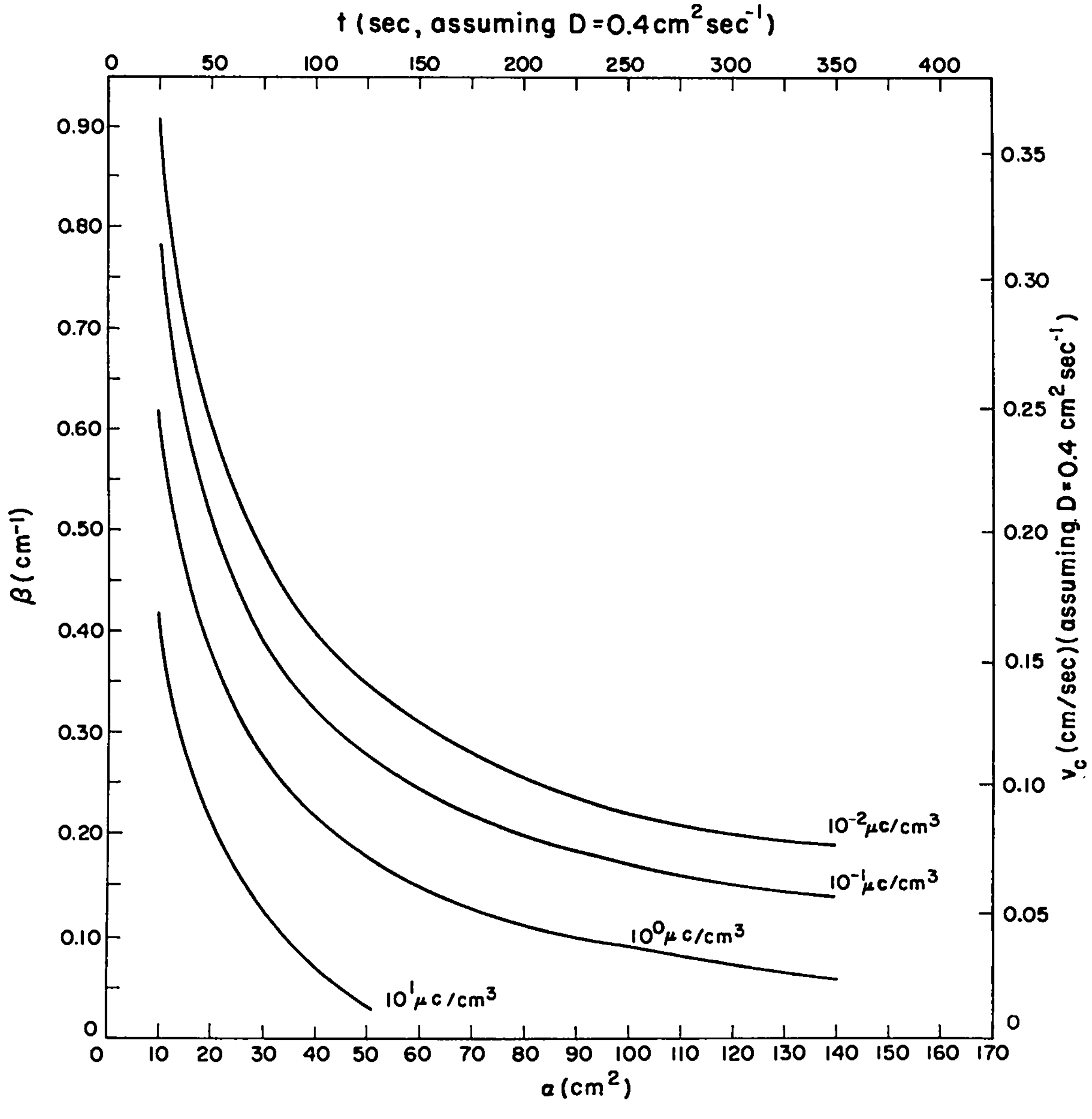


Figure 6. β or velocity vs α or time for a family of concentrations for a 1-curie source.

concentration velocity can be found once a value of D is assumed. The appropriate coordinates for $v_c$ are shown for the case of D = 0.4.

The maximum concentration attained at any given distance r from the point of source release occurs when the concentration velocity at that point is zero. Setting $v_c$, and hence $\beta$, equal to zero in the above relations,

$$\alpha = \frac{r^2}{6} \quad ,$$

and

$$t = \frac{r^2}{6D} \quad .$$

This is the time for a maximum concentration of the diffusing gas when the diffusion process is the only one taking place. The approximate values of $\alpha$ and t, assuming D = 0.4, are shown for representative distances in Table I. From Figures 2 and 3, the value for the maximum concentration can be determined for any pair of values of r and $\alpha$. The peaks of the concentration curves in Figure 4 show clearly the maximum distance attained by a given concentration.

TABLE I

EXAMPLES OF $\alpha$ AND TIME VS DISTANCE FOR MAXIMUM CONCENTRATIONS OF A DIFFUSING GAS

| r (cm) | $\alpha$ ($cm^2$) | t (sec)* |
|---|---|---|
| 10 | 17 | 42 |
| 20 | 67 | 170 |
| 50 | 420 | 1050 |
| 100 | 1700 | 4200 |
| 200 | 6700 | 17000 |

*D = 0.4

## DESCRIPTION OF EXPERIMENTS

Tritium was released in the accelerator area of the LASL 12-Mev Van de Graaff building to serve as a check on the response of its new, Model 128, multi-station tritium monitoring system, as well as to study the mixing of tritium with air. Two independent experiments were performed, involving the Model 128 system and detection instruments of 4 different designs which were distributed in the Van de Graaff accelerator area. In the first experiment 1 curie of tritium was released, and in the second, 10 curies.

The accelerator area of the Van de Graaff building consists of two adjoining sections, a 10-story tower and a 4-story, barn-like structure (Figure 7). The construction is of thin sheet metal, with windows opening at the top of the tower and with a roll-up door on the first floor. Entrance to the building from the control room is through a long hallway which enters the building on the second floor. The volume of air in the building is about $8 \times 10^3$ $m^3$. Air in the building was made as static as possible by closing all doors and windows.

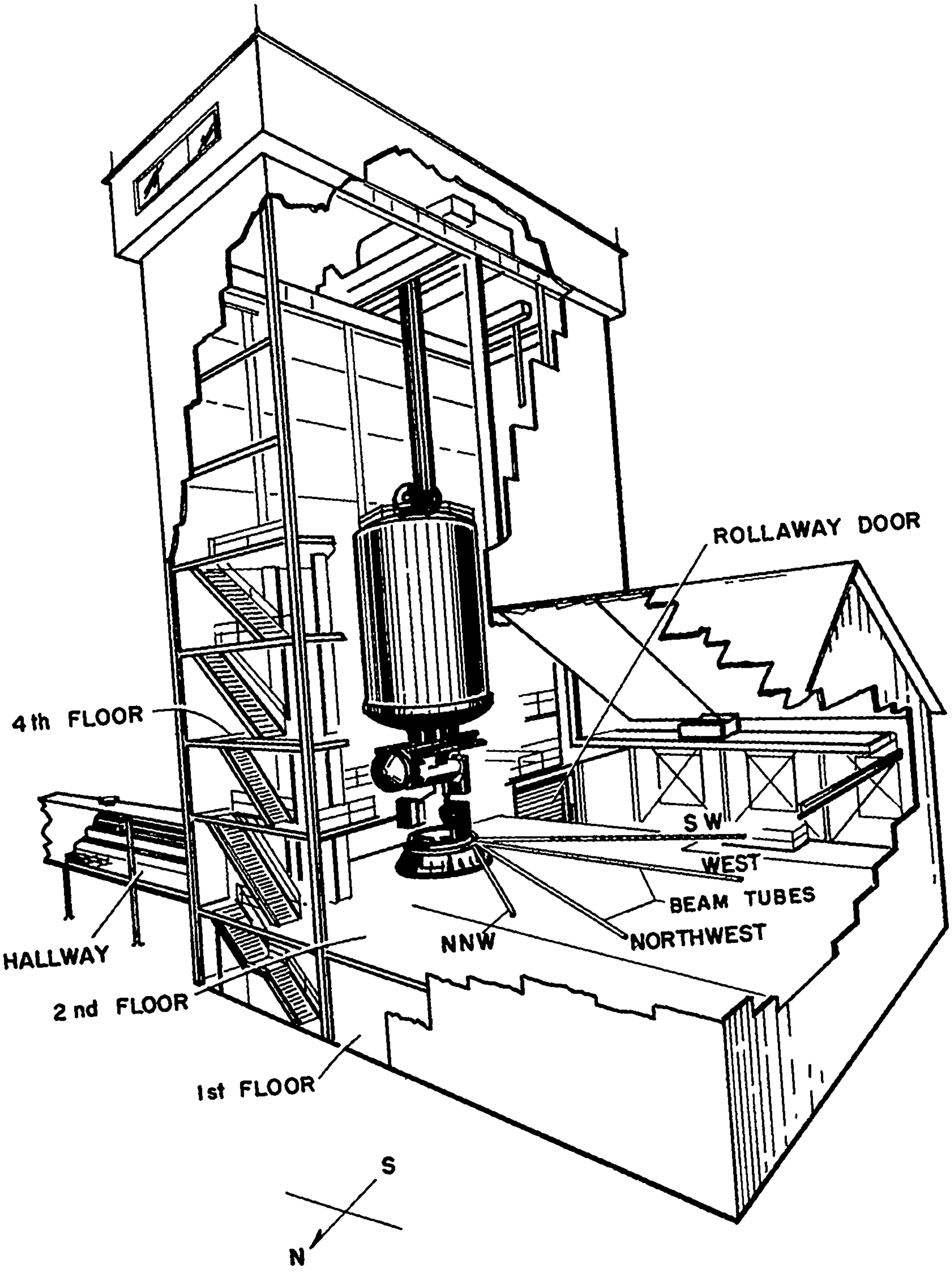


Figure 7. LASL 12-Mev Van de Graaff accelerator building where tritium experiments were performed.

The following instruments were used in these experiments:

1. Model 128. The detectors of this permanently installed multi-station system are all located 6 feet 9 inches above the floor, one in the hallway and one near the target area at the end of each beam tube (Figure 8). Each detector has a 660-cc ion chamber that is sensitive to ions produced inside the chamber or produced outside and drawn into the chamber. The air is circulated through the chamber at the rate of 15 $ft^3$/min. For each detector there are 3 indicating meters, located as follows: (1) at the control desk in the control room, (2) at an instrument panel in the hallway, and (3) near the detector at the target end of each beam tube. Each meter covers 4 decades and reads about $10^5 \mu c/m^3$ at full scale.

During these experiments, each Model 128 was equipped with a 100-mv Varian recorder (G-11A). These recorders were positioned in the hallway and synchronized to start with the release of tritium.

One Model 128 detector ion chamber was calibrated with tritium water vapor (Figure 9), and curves were extrapolated to conform with instrument sensitivity data. All other chambers were calibrated by taping a 10-mc $Co^{60}$ source to the outside of each chamber and adjusting the chambers to read the same as the one calibrated with tritium.

The instruments were checked for proper response with pieces of zirconium-tritium target. Two well-ventilated cans (Figure 10) were loaded with a selection of target pieces so that the instruments read 1/4 and 3/4 full scale. The loss of tritium gas to the room atmosphere from these target pieces was small and did not present a health problem.[6]

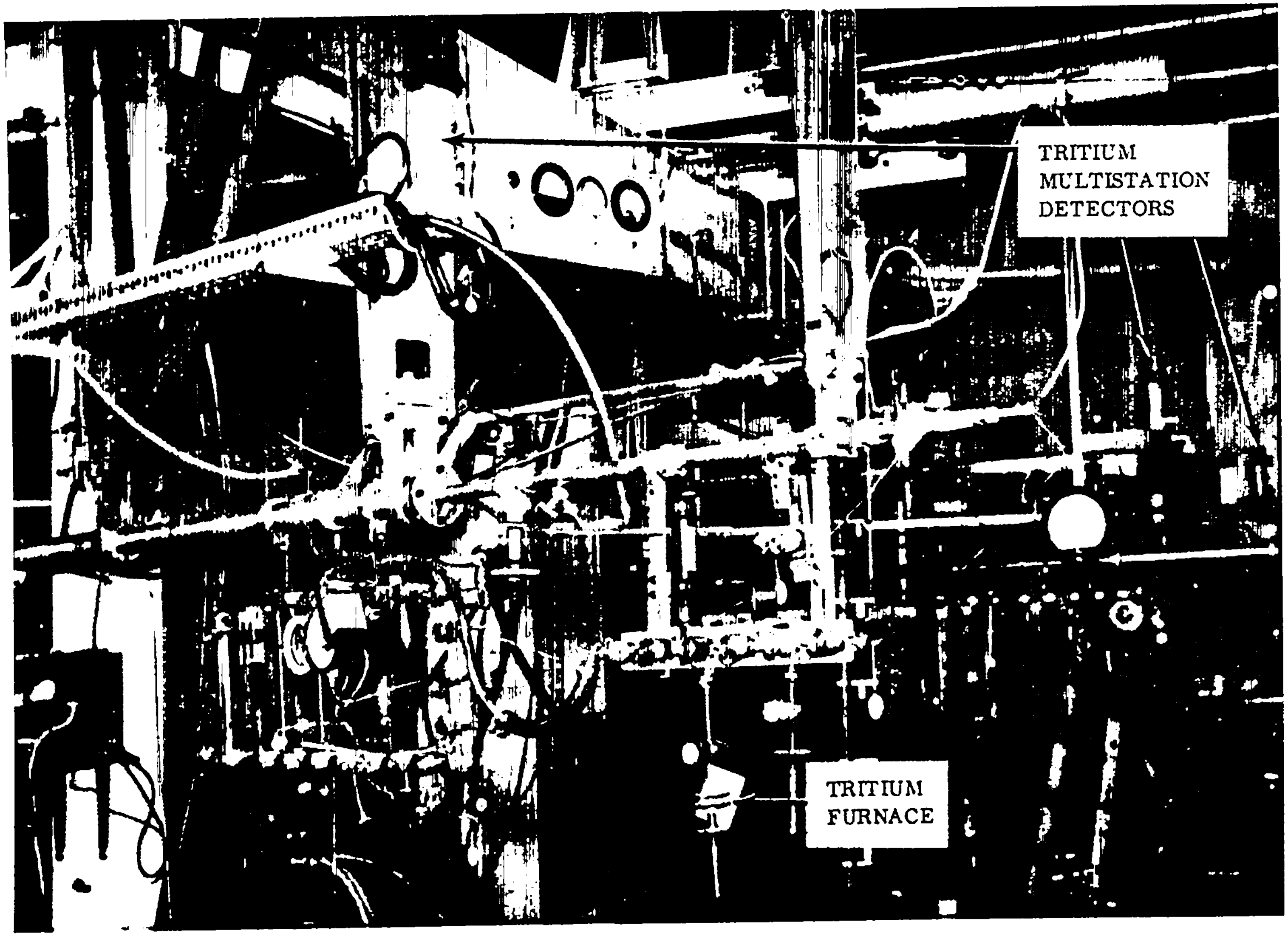


Figure 8. Part of reaction area of Van de Graaff accelerator.

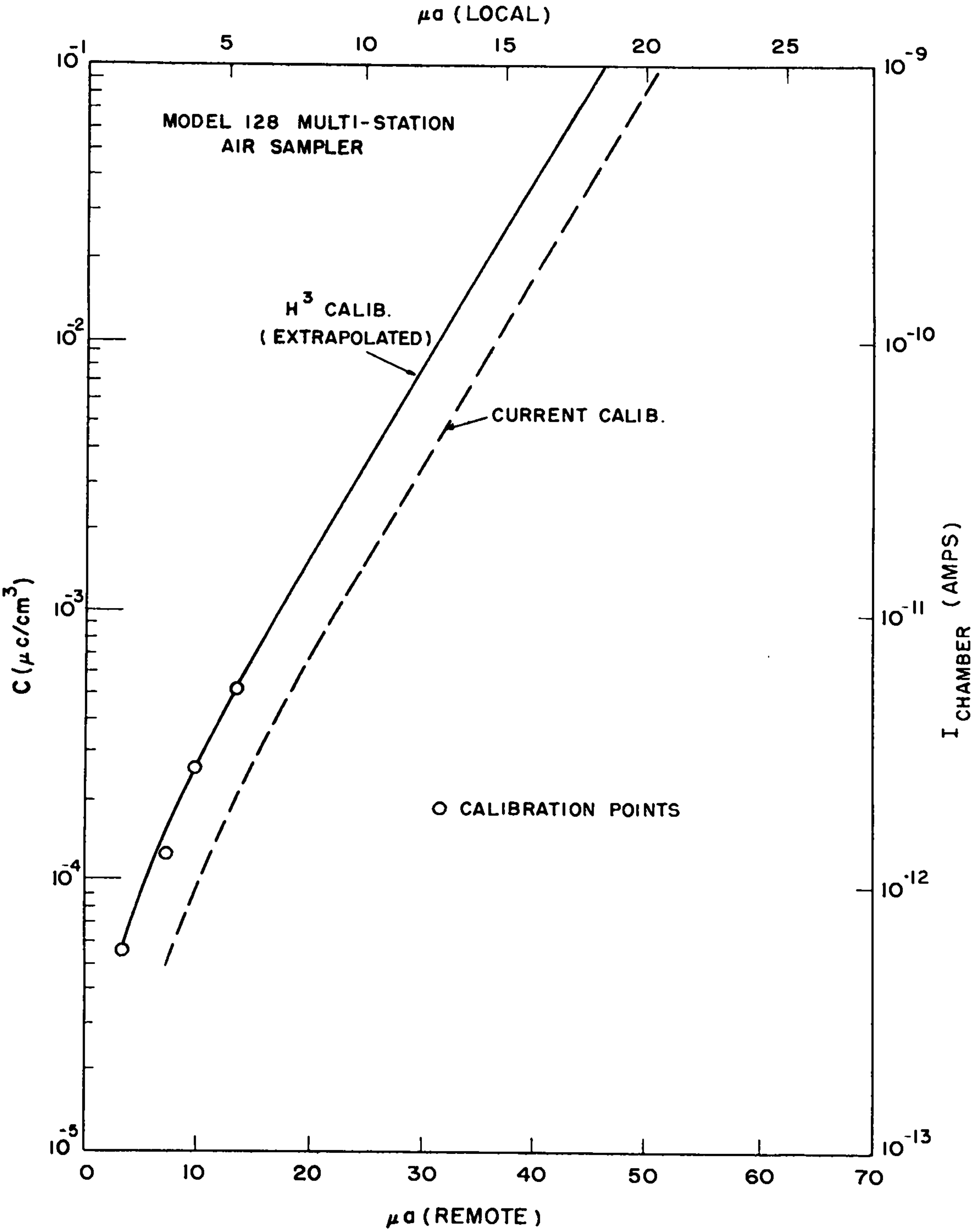


Figure 9. Calibration curve for the Model 128 ion chamber calibrated to tritium water vapor.

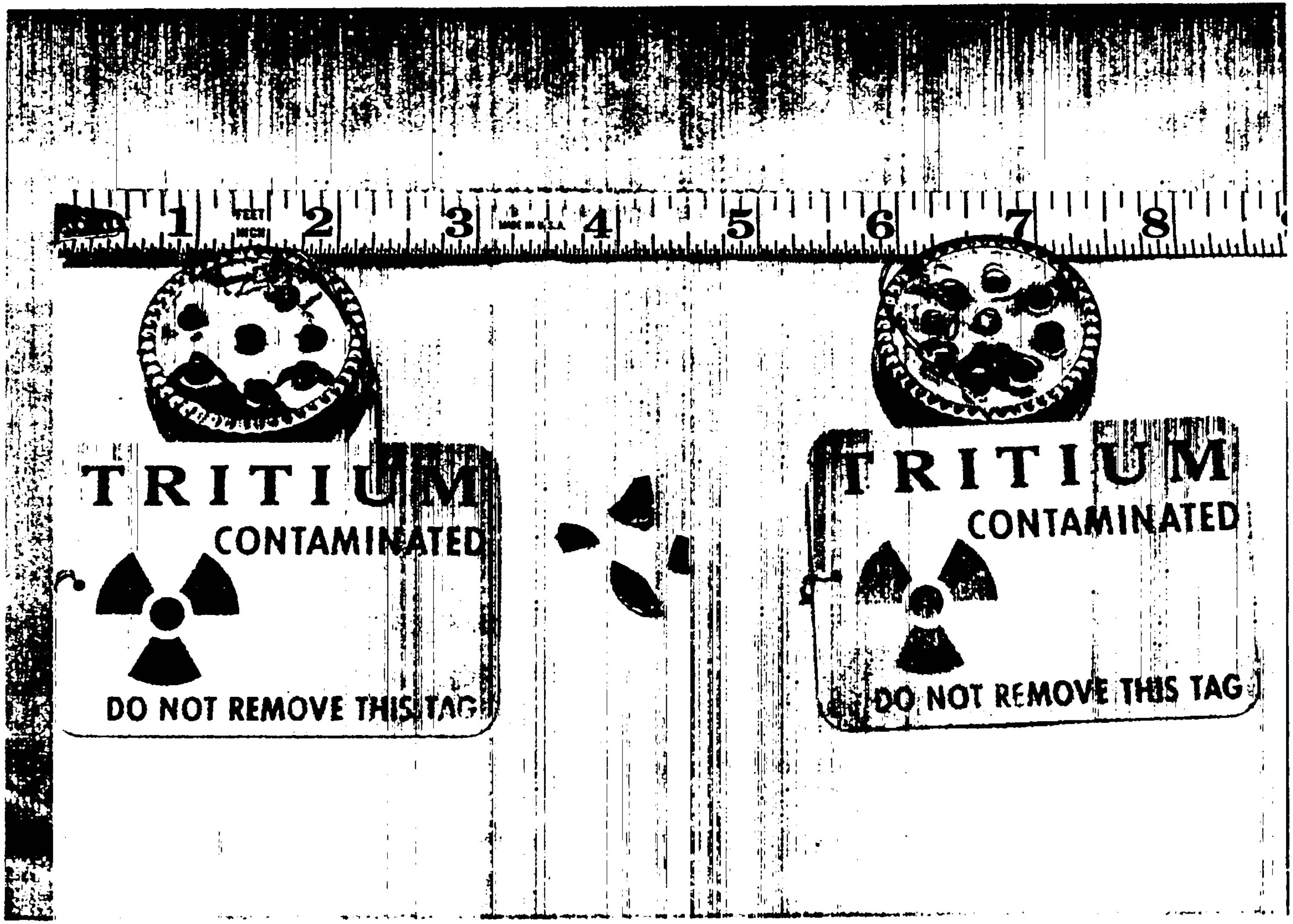


Figure 10. Cans containing pieces of zirconium-tritium target used to check response of Model 128 tritium detectors.

2. Model 101 Tritium Sniffer.[7] This is a portable instrument with the same characteristics and sensitivity as the Model 128 multistation air sampler.

3. T-289 Radioactive Gas Sampler. This instrument, a commercial version of the Model 101 Sniffer, has an ion chamber with a vibrating reed electrometer. The air passes through an ion precollector before entering the chamber, making the instrument insensitive to ions produced outside the chamber. There are 4 scales (X1, X10, X100, X1000), with the least sensitive reading $1.2 \times 10^4$ $\mu c/m^3$ at full scale. One T-289 was equipped with a 15-ft, 1/2-in. I.D. tygon hose on the air intake.

4. T-290 Radioactive Gas Sampler. This is a military portable survey meter, similar to the Model 101 but with the addition of an ion precollector. It has 3 scales (X1, X10, X100), the least sensitive reading $1.2 \times 10^5$ $\mu c/m^3$ at full scale.[8] One T-290 was equipped with a 6-ft, 1/2-in. I.D. tygon hose on the air intake.

5. NR-1. This instrument is a commercial version of the Model 101 Sniffer. There are 4 scales ranging from a full-scale reading of 75 $\mu c/m^3$ on the most sensitive to a full-scale reading of $1.5 \times 10^5$ $\mu c/m^3$ on the least sensitive.

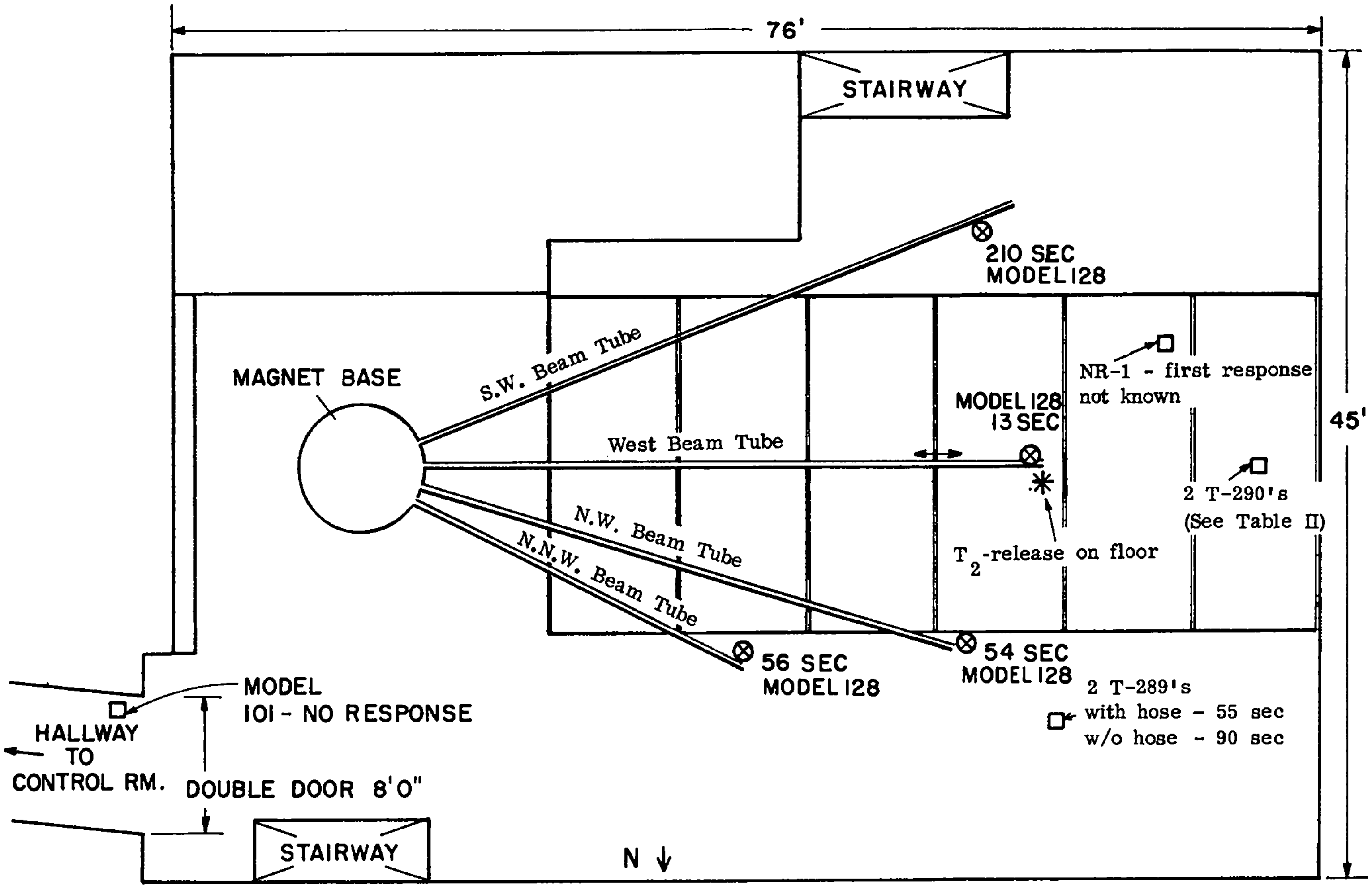
The instruments without recorders were observed by persons with stop watches. At a given signal, the tritium was remotely released and a series of readings vs time was recorded for each instrument.

The tritium reservoir was a stainless steel cylinder of approximately 10 $cm^3$ volume. Deuterium gas was mixed with tritium gas to provide pressure high enough to ensure that the tritium gas would eject rapidly into the air. The initial source, appearing nearly instantaneously, was estimated to be 1 ft in diameter.

The reservoir containing 1 curie of tritium was placed on the second floor below the west beam tube, as shown in Figure 11. The approximate positions of the instruments and the times for the first indications are also shown in Figure 11. The experiment was repeated using 10 curies of tritium, with the reservoir placed adjacent to the northwest beam tube on a small platform 5 ft above the floor. The locations of the source and the instruments on the second floor during the 10-curie experiment are shown in Figure 12. To provide further data, these locations generally differed from those of the first experiment.

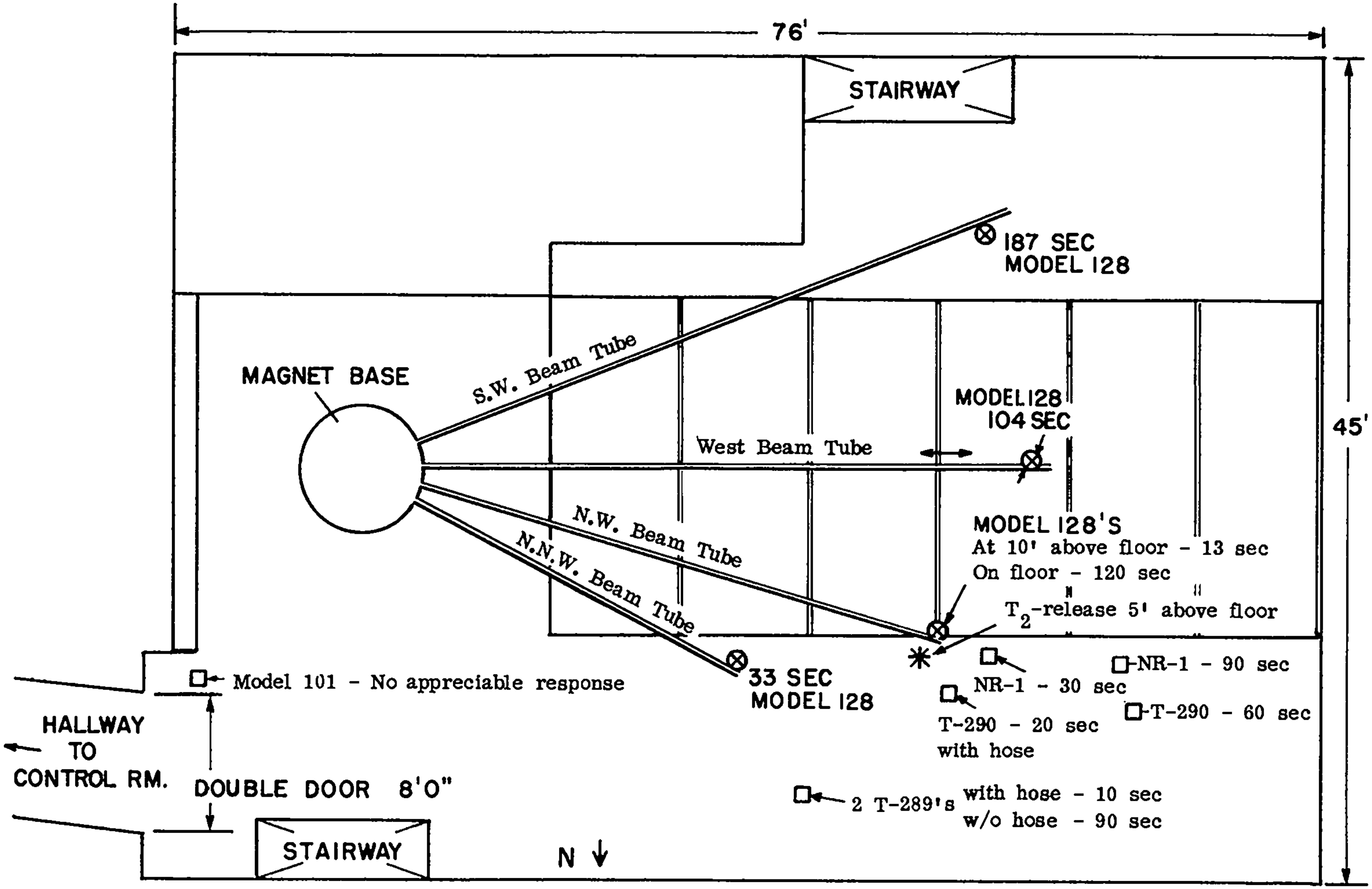
Prior to the tritium experiments, both smoke and bubbles, the latter made by blowing hydrogen gas through a soap solution, were released in an attempt to determine air movement in the room. As anticipated, the air moved toward the tower from the tritium gas release point. Outside weather conditions were noted, but their effect on air turbulence in the room was not evaluated.

Although urine samples were obtained from all persons involved in the experiments, no tritium could be detected in any of the samples, even though two men were stationed upwind 6 to 8 ft from the tritium reservoir. Since tritium gas is several orders of magnitude less toxic than tritium water vapor,[9] no health hazard was expected or experienced.



SECOND FLOOR, ACCELERATOR BUILDING

Figure 11. Locations of instruments and times of first response during 1-curie tritium experiment.



SECOND FLOOR, ACCELERATOR BUILDING

Figure 12. Locations of instruments and times of first response during 10-curie tritium experiment.

After each experiment, the atmosphere of the building was flushed by opening the tower windows and the roll-up door on the first floor. Twenty hours after each experiment, a 1-ft-square area on the floor under the point of release was swabbed with cotton and distilled water; the solution was run as a urinalysis with negative results.

# RESULTS OF EXPERIMENTS

The results obtained with the various instruments are given in Table II for the experiment with 1 curie of tritium, and in Table III for the experiment with 10 curies of tritium. The apparent velocity is determined by dividing the distance of a detector from the point of release by the time of first response. The value is obviously dependent upon the sensitivity of the instrument and the size of the source.

Figure 13 shows two examples of the records obtained from the Model 128 stations in the experiment with 10 curies of tritium.

The Appendix shows some observations made during the experiments with tritium sources of 1 and 10 curies.

## TABLE II

## ONE-CURIE TRITIUM EXPERIMENT

| Instrument | | Position | Distance from release point | Time of first response (sec) | Apparent velocity (cm/sec) | Time of maximum reading (sec) | Indicated maximum concentration (μc/cm³) |
|---|---|---|---|---|---|---|---|
| 1. Model 128 multi-station system | a. | West beam tube, 6'9" above floor | 6'9" | 13 | 15.8 | 16 | $7 \times 10^{-2}$ |
| | b. | Northwest beam tube, 6'9" above floor | 15'6" | 54 | 8.7 | 70 | $4.7 \times 10^{-3}$ |
| | c. | North-northwest beam tube, 6'9" above floor | 20' | 56 | 10.9 | 77 | $4.5 \times 10^{-3}$ |
| | d. | Southwest beam tube, 6'9" above floor | 14' | 210 | 2.0 | 550 | $1.6 \times 10^{-3}$ |
| 2. Model 101 Sniffer | | Floor of hallway | 40' | No response until windows and door were opened | | | $5 \times 10^{-5}$ |
| 3. T-289 | a. | North of source; equipped with 15' hose leading to 1' from release point | 16' (including hose) | 55 | 8.9 | 90 | $>10^{-2}$ |
| | b. | North of source and 30" above floor | 15' | 90 | 5.1 | 150 | $7.0 \times 10^{-3}$ |
| 4. T-290 | a. | On floor, west of source; equipped with 6' hose coiled about instrument | 21' (including hose) | 540 | 1.2 | 780 | Not recorded |
| | b. | On floor, west of source | 15' | 510 | 0.9 | 900 | Not recorded |
| 5. NR-1 | | On floor, southwest of source | 15' | Not known | Not known | 600 | $9 \times 10^{-5}$ |

## TABLE III

## TEN-CURIE TRITIUM EXPERIMENT

| Instrument | Position | Distance from release point | Time of first response (sec) | Apparent velocity (cm/sec) | Time of maximum reading (sec) | Indicated maximum concentration ($\mu c/cm^3$) |
|---|---|---|---|---|---|---|
| 1. Model 128 multi-station system | a. Northwest beam tube, directly above release point | 5' | 13 | 11.7 | 19 | $1.4 \times 10^{-1}$ |
| | b. Northwest beam tube, directly below release point | 5' | 120 | 1.3 | 290 | $1.8 \times 10^{-3}$ |
| | c. North-northwest beam tube, 6'9" above floor | 8'5" | 33 | 7.8 | 52 | $4.1 \times 10^{-2}$ |
| | d. West beam tube, 6'9" above floor | 14'6" | 104 | 4.2 | 208 | $5.6 \times 10^{-3}$ |
| | e. Southwest beam tube, 6'9" above floor | 26'6" | 187 | 4.3 | 320 | $3.7 \times 10^{-3}$ |
| 2. Model 101 Sniffer | a. First floor, north stairway, ~18' below release point | 23'* | No appreciable response | | | |
| | b. Fourth floor, ~ 15' above release point | 25'6" | 80 | 9.7 | 180 | $2 \times 10^{-3}$ |
| | c. Floor of hallway | 35' | No appreciable response | | | |
| 3. T-289 | a. Northeast of source; equipped with 15' hose leading to 1' from point of release | 16' (including hose) | 10 | 48.8 | 10 | $>10^{-2}$ |

| | | | | | | | | |
|---|---|---|---|---|---|---|---|---|
| | | b. | Northeast of source and 30" above floor | 15' | 90 | 5.1 | 100 | $>10^{-2}$ |
| 4. | T-290 | a. | Northwest of source; equipped with 6' hose leading to 1' from point of release | 7' (including hose) | 20 | 10.7 | 20 | $>10^{-1}$ |
| | | b. | Northwest of source and 12" above floor | 15' | 60 | 7.6 | 540 | $8.5 \times 10^{-3}$ |
| | | c. | Fourth floor, ~ 15' above release point | 25.6" | 190 | 4.0 | 360 | $7 \times 10^{-4}$ |
| 5. | NR-1 | a. | North-northwest of source and 12" above floor | 3' | 30 | 3.0 | 90 | $1.5 \times 10^{-3}$ |
| | | b. | North-northwest of source and 12" above floor | 15' | 90 | 5.1 | 600 | $1.3 \times 10^{-3}$ |

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*This is the approximate distance tritium would have to travel from the release point to the instrument.

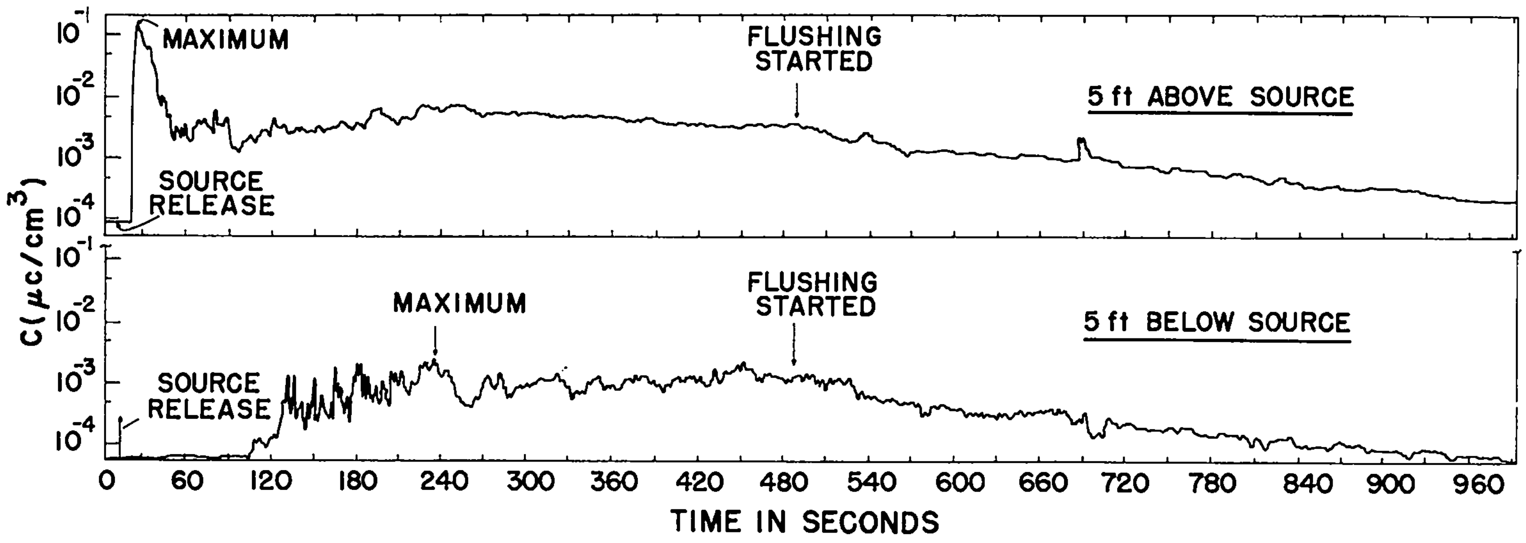

Figure 13. Graphs from the Varian recorders used with the Model 128 detectors on the northwest beam tube in the 10-curie tritium experiment.

## DISCUSSION OF RESULTS

Interpretations have been applied to the diffusion process by a series of curves developed to provide determination of concentration, velocity, and maximum concentration as functions of time and distance.

Based on theoretical diffusion considerations, the concentration velocity of tritium in air has a practical maximum value less than 0.4 cm/sec (Figure 5). The measured concentration velocity was dependent on the air currents in the room and was as much as three orders of magnitude greater than the theoretical value.

When the diffusion process is the only one considered (using a 1-curie source), a maximum concentration of $10^{-2}$ $\mu c/cm^3$ would occur in $1.6 \times 10^4$ sec at a distance of 195 cm (Figure 4). In the 1-curie tritium source experiment ( Table II), a maximum concentration of $7 \times 10^{-2}$ $\mu c/cm^3$ occurred in about 16 sec at a distance of 206 cm. The difference in maximum concentration and the large factor of $10^3$ in time for short distances are due to air turbulence and initial motion of the source gas. Diffusion is insignificant when compared to the mixing forces of air motion.

In a building of $8 \times 10^3$ cubic meters volume, a 10-curie source evenly diffused with air would give a concentration of 1.25 x $10^{-3}$ $\mu c/cm^3$. Immediately above the point of release, a fairly steady concentration of about 5 x $10^{-3}$ $\mu c/cm^3$ was obtained (Figure 13), four times greater than that from strict diffusion calculations. For the first 2 min, tritium concentrations at floor level immediately below the point of release were 100 times less than those 10 ft above. During the next 6 min, there were large fluctuations at floor level, with average concentrations 1/5 as large as those 10 ft above. Variations in concentrations on the floor were large due to the uneven mixing of tritium and air within the interval of observed time.

After flushing of the building air started, the time required for an indicated 50% decrease in concentration varied from 2.5 to 6.5 minutes, and obviously was dependent on the local air movement near each detector. In these experiments with sources of 1 and 10 curies, the room was clear of measureable amounts of tritium in 20 to 30 min, a time consistent with the time for a 50% decrease.

Tables II and III show that, at all distances, the times for first responses and for maximum concentrations vary greatly. These data emphasize that the hazard from tritium gas cannot be evaluated accurately by random sampling. For personnel monitoring, the detector should be as close to the breathing zone as possible and in a position to take advantage of any existing air motions. In some applications it would be useful to attach a short piece of hose to the intake of the detector in order to get a more rapid and accurate indication by placing the sampling point closer to the source of tritium.

# APPENDIX

## OBSERVATIONS MADE DURING THE EXPERIMENTS WITH TRITIUM SOURCES OF 1 AND 10 CURIES

| | 1-curie Experiment | 10-curie Experiment |
|---|---|---|
| Time between source release and start of flushing | 18.5 min | 8 min |
| Time elapsed before indicated 50% decrease in concentration | 3.5-4.5 min | 2.5-6.5 min |
| Time required for Model 128 to show nearly steady concentration | 6.5 min | 5-6 min |
| Indicated approximate steady concentration by Model 128 | $6.5 \times 10^{-4}$ μc/cm$^3$ | $5 \times 10^{-3}$ μc/cm$^3$ |
| Calculated concentration in full-room volume | $1.25 \times 10^{-4}$ μc/cm$^3$ | $1.25 \times 10^{-3}$ μc/cm$^3$ |

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