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An Investigation of a Bubbler Tritium Sampler

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An Investigation of a Bubbler Tritium Sampler*

by

Allen M. Valentine

*Experimental work conducted during employment at Lawrence Radiation Laboratory, operated by the University of California under AEC Contract W-7405-Eng-48.

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AN INVESTIGATION OF A BUBBLER TRITIUM SAMPLER

by

Allen M. Valentine

ABSTRACT

Investigation of a bubbler tritium sampler indicated collection characteristics similar to reported human body uptake rates for tritium gas and tritiated water vapor. The bubbler collector consisted of a dry gas wash bottle with a fritted glass tip on the inlet and distilled water as the collecting medium.

Tests were made under both laboratory and field conditions, and the effects of varying the flow rate, water volume, and water temperature were determined. A flow-through ionization chamber instrument was used to measure the tritium concentrations sampled, and the collected tritium was determined by liquid scintillation counting.

The observed collection efficiency for tritiated water vapor was >90% for sampling rates from 8 to 10 lpm, water temperatures from 25 to 35°C, and water volumes from 30 to 50 cc. Tritium gas was collected with an efficiency of <0.1% with the same sampling rates and water volumes but with a water temperature of 18°C.

This collector can selectively sample tritiated water vapor in the presence of tritium gas, and allows the health physicist to make a better assessment of tritiated water vapor concentrations in air.

I. INTRODUCTION

The abundance of tritium has increased during recent years because larger quantities exist at most heavy water reactor facilities and tritium has become easier for scientists to obtain. Tritium is commercially available in many different forms, but tritium gas, tritiated water, and tritiated organic compounds are the most frequently used.

Health physics considerations increased as more scientists started handling tritium. The need for improving tritium air sampling systems and techniques became apparent be-

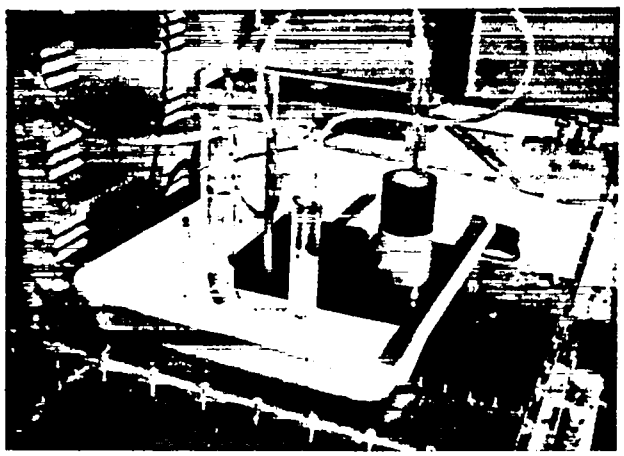

Fig. 1. Two bubbler collectors.

cause most tritium samplers could not selectively collect or differentiate between tritium gas and tritiated water vapor in air. The inability to selectively collect either tritium gas or tritiated water is undesirable because the body uptake rates[1] and maximum permissible concentrations in air (MPC_a)[2] for the two forms differ greatly. Tritiated water vapor is ~400 times more hazardous than tritium gas when present in the working atmosphere.

The collector evaluated in this report employs distilled water as the collecting medium[3] and selectively collects tritiated water vapor.

II. DESCRIPTION

A. General Techniques

The collector is called a "bubbler" because of the bubbling action that occurs when air passes through the water. The collector is a 125-cc dry gas wash bottle with a fritted glass dispersion tip on the inlet and from 30 to 50 cc of distilled water as the collecting medium. Two collecting bottles containing distilled water are shown in Fig. 1. A low-volume gas pump and flow meter combination is used to provide a known

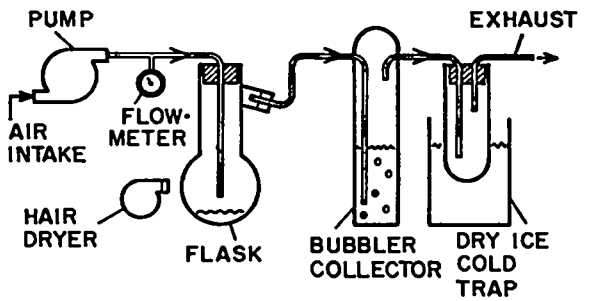

Fig. 2. A bubbler sampler for laboratory sampling of tritiated water vapor.

air flow through the system.

Tritium is collected in the distilled water when air containing tritium is bubbled through the collector. The tritium activity in the distilled water is determined by accurately measuring the total water volume after sampling and counting a 500-λ aliquot of this water and tritium in a liquid scintillation counter. The counter has a known counting efficiency for tritium under the counting conditions.

The collection efficiency of the collector for tritium gas and tritiated water vapor is evaluated by making laboratory tests with known tritium concentrations and field tests with tritium concentrations determined by an ionization chamber instrument. The ionization chamber instrument has beem empirically calibrated for tritium. The field tests included studies of the effort of flow rate, water volume, and water temperature on collection efficiency.

B. Tritiated Water Vapor Sampling

1. Laboratory Sampling. Measured volumes of tritiated water of known specific activity are evaporated and circulated through a bubbler collector (Fig. 2). Hot air from a hair dryer is used to give uniform evaporation for the desired sampling times. A constant air flow is maintained through the system during each of four tests;

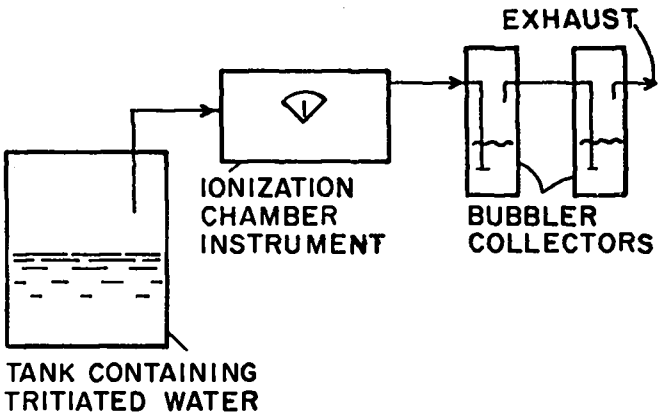

Fig. 3. Field sampling system for tritiated water vapor.

however, the rate is different for each test.

The volumes of bubbler water and cold trap condensate are measured and analyzed for tritium after each test to determine the collection efficiency.

2. Field Sampling. The term "field sampling" was used to denote sampling of air that contained unknown tritium concentration. Air containing unknown but measurable concentrations of tritiated water vapor was sampled from metal tanks partially filled with tritiated water (Fig. 3).

An ionization chamber instrument with a built-in gas pump and flowmeter was used to provide air flow through the system and to measure the tritium concentration during each test. For most tests, the air with tritiated water vapor first passed through the ionization chamber instrument after being drawn from the tanks and then through two bubbler collectors in series. The second collector retained the activity lost from the first and indicated how efficient the first was for the tritium sampled.

c. Tritium Gas Sampling

A two-container system was used for bubbler collector tests with tritium gas (Figs. 4 and 5). The two containers were gas-tight Plexiglas boxes with volumes of 0.05 and 1.09 m^3. One curie of tritium gas was first released in the small box, and ~3 mCi was later transferred into the larger box. The small box was then isolated from the system, and circulation was established through the ionization chamber instrument and large box until the tritium gas concen-

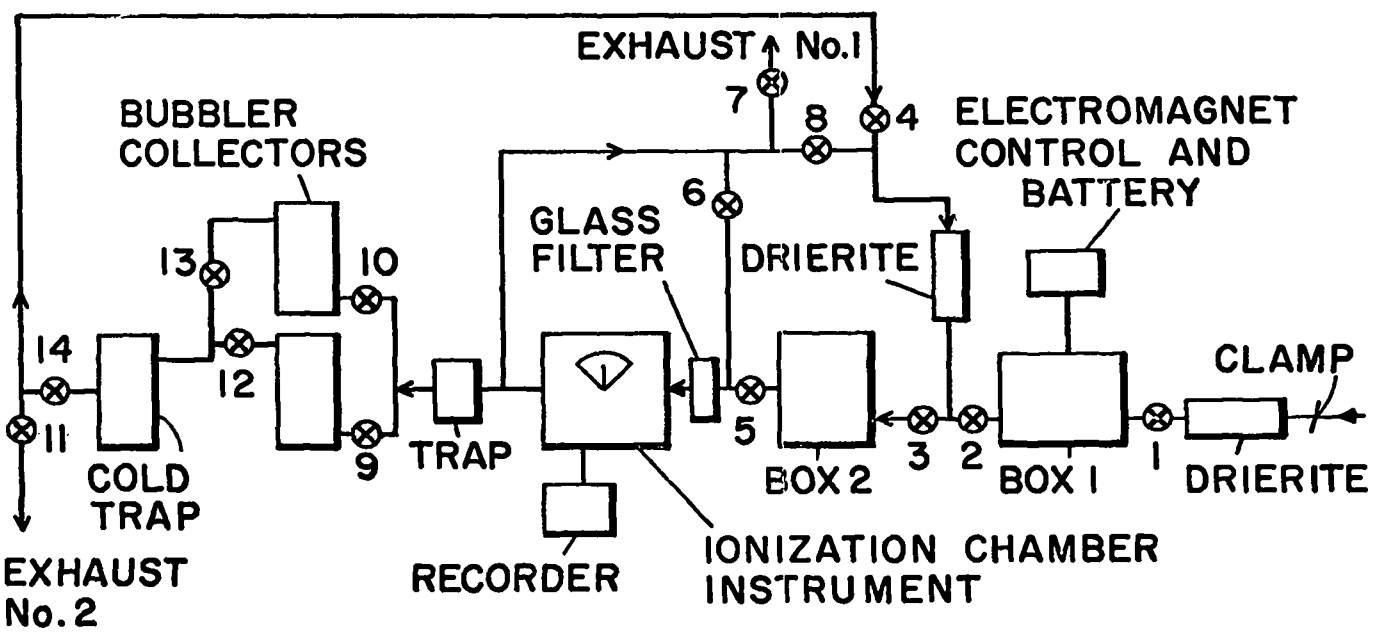

Fig. 4. Schmatic of a bubbler tritium gas sampler.

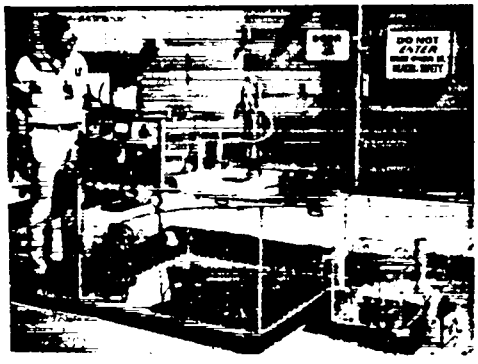

Fig. 5. A Bubbler tritium gas sampler.

tration was uniform. Before the gas was released, dry air had been circulated through the system to minimize the oxidative conversion of tritium gas to tritiated water. The ionization chamber instrument was used to measure tritium concentrations in the air sampled from the large box during the collector tests.

III. RESULTS

A. Tritiated Water Vapor Tests

1. Laboratory Sampling. The results and sampling conditions for the four laboratory tests are given in Table I. From 91 to 95% of the activity passing through the bubbler collector was collected. Water was visibly blown out of the bubbler when flow rates greater than 20 l/min were attempted.

2. Field Sampling

(a) Sampling Time Dependence. Samples were collected for from 5 to 20 minutes, and the results are given in Table II. For each sample, two bubbler collectors were connected in series.

The first bubbler collector retained >90% of the total tritium circulated through it for all four samples. Less than 3% of the total tritium collected was in the second bubbler when the sample time was 15 minutes or less. For a sample time of 20 minutes, 8% of the tritium was in the second collector.

(b) Water Volume Dependence. A minimum water volume is desirable because it decreases the dilution of the tritium collected and allows higher air flow rates through the collector. Increased air flow is possible because it becomes more difficult to blow water out of the bubbler collector. The size of the dry gas wash bottle also dictates the maximum water volume that can be used for a given flow rate.

The water volume was decreased from 50 to 30 cc in 5-cc increments to determine the effect of water volume on the collection efficiency. The results given in Table III indicate that for these test conditions less that a 5% difference in the tritium activity

TABLE I

RESULTS OF LABORATORY SAMPLING
OF TRITIATED WATER VAPOR

Sampling Conditions	Test No. 1	Test No. 2	Test No. 3	Test No. 4
Initial activity in flask (μCi)	0.563	0.563	0.563	0.563
Flow rate (l/min)	5	10	15	20
Sampling time (min)	11	12	12	14
Initial volume in flask (ml)	2	2	2	2
Initial bubbler volume (ml)	100	100	100	100
Final bubbler volume (ml)	101	101	98.5	98
Volume collected in trap (ml)	0.8	1	1.6	1.9
Activity collect in bubbler (μCi)	0.536	0.535	0.512	0.526
Activity collected in trap (μCi)	0.001	0.002	0.004	0.006
Activity collected in bubbler (%)	95	95	91	93

TABLE II

SAMPLING TIME DEPENDENCE OF A BUBBLER SAMPLER FOR TRITIATED WATER VAPOR[a]

Sample Time (min)	Total Activity Collected (μCi)	Activity in Second Collector (%)	Indicated Air Concentration (μCi/m³)
5	1.81	1.7	42
10	3.42	1.8	40
15	4.54	2.9	35
20	7.00	7.8	38

[a]Flow rate, 8.5 l/min.
Water volume, 50 ml.
Water temperature, 26°C.
Ionization chamber measured concentrations from 36 to 44 $\mu Ci/m^3{}_a$.

collected was found for volumes between 30 and 50 cc. The collection efficiency was >90% for all samples.

(c) Water Temperature Dependence. Ice- and hot-water baths were used to maintain the bubbler collector water at temperatures between 8 and 35°C. There was some difficulty in maintaining a constant water temperature for the extreme temperatures, and reported temperatures are averages. The maximum change during the 10-minute sampling time was 5°C.

The results are given in Table IV and

TABLE III

WATER VOLUME DEPENDENCE OF A BUBBLER SAMPLER FOR TRITIATED WATER VAPOR[a]

Water Volume (ml)	Total Tritium in Bubbler (μCi)	Indicated Air Concentration (μCi/m³)
30	5.52	130
35	5.60	132
40	5.72	135
45	5.40	127
50	5.60	132

[a]Sample time, 5 min.
Flow rate, 8.5 l/min.
Water temperature, 26°C.
Ionization chamber instrument measured an average concentration of 138 $\mu Ci/m^3{}_a$.

TABLE IV

WATER TEMPERATURE DEPENDENCE OF A BUBBLER SAMPLER FOR TRITIATED WATER VAPOR[a]

Average Water Temperature °C	Average Water Temperature °F	Indicated Air Concentration (μCi/m³)
8	46	28
14	57	34
18	64	37
26	79	43
32	90	43
35	95	42

[a]Sampling time, 10 min.
Flow rate, 8.5 l/min.
Water volume, 50 cc.
Ionization chamber instrument measured a concentration of 44 $\mu Ci/m^3{}_a$.

indicate collection efficiencies of >90% for water temperatures between 20 and 35°C. Between 25 and 35°C, there was less than a 10% difference in collection efficiency, and this was later found to be within the experimental error for sampling at a constant temperature.

(d) Uniformity of Sample Results. The uniformity of bubbler sample results was evaluated by having two experimenters collect samples from two different sources of tritium on separate days. The results of these independent sample evaluations are given in Table V. The sample results agreed to within ±8% and ±19% for a sample time of 5 minutes and concentrations of ~20 MPC_a and 10 MPC_a, respectively. Some of the ±19% variance was caused by changing tritium concentrations during sampling and by poor counting statistics due to the low tritium activity collected during the 5-minute sample time.

B. Tritium Gas Tests

Nine bubbler samples were collected within 4 hours after the tritium gas was released in the gas sampling system. The re-

TABLE V

UNIFORMITY OF RESULTS OBTAINED WITH A BUBBLER TRITIUM SAMPLER[a]

Experimenter A		Experimenter B	
Sample No.	Measured Air Concentrations ($\mu Ci/m^3$)	Sample No.	Measured Air Concentrations ($\mu Ci/m^3$)
69	32	74	110
70	36	75	130
71	41	76	120
72	44	77	120
73	34		
Average	37		120

[a]Sampling time, 5 min.
Flow rate, 8.5 l/min.
Water temperature, ~26°C.
Water volume, 50 ml.
Air temperature, ~20°C.

TABLE VI

TRITIUM GAS SAMPLE RESULTS[a]

Total Tritium Circulated through Bubbler (μCi)	Total Tritium Collected in Bubbler (μCi)	Collection Efficiency (%)
120	2.7×10^{-2}	0.023
240	4.7×10^{-2}	0.020
230	4.4×10^{-2}	0.019
116	2.8×10^{-2}	0.024
116	2.4×10^{-2}	0.021
192	3.6×10^{-2}	0.019
156	3.6×10^{-2}	0.023
150	3.5×10^{-2}	0.023
154	3.2×10^{-2}	0.021

[a]Bubbler water temperature, 18°C.
Gas-air temperature, 18°C.
Sample time, 5 to 10 min.
Flow rate, 8.5 l/min.
Bubbler water volume, 30 to 40 ml.
Ionization chamber instrument measured concentrations from 1800 to 3060 $\mu Ci/m^3$.

sults are given in Table VI, and for these samples less than 0.025% of the tritium circulated through the bubbler collector was collected. The large box was isolated with the remaining tritium in it after the nine samples were collected. The box was left undisturbed and unsheltered for 11 days before being resampled. This was done so that the extent of tritium oxidation could be observed and to allow rechecking of the bubbler collection efficiency. After 11 days, a single 10-minute sample was collected with two bubbler collectors in series. The first collector retained 0.1% and the second retained 0.02% of the tritium circulated through the first collector.

IV. DISCUSSION

The initial purpose of the study was to determine the performance of the bubbler collector for sampling tritium field conditions. The numbers reported are qualitative and the collection efficiencies are reported as >90% for tritiated water vapor and <0.1% for tritium gas. However, the efficiency for tritiated water is probably >95% for most sampling conditions.

The bubbler collector selectively collects tritiated water vapor from a tritium atmosphere, and can be conveniently used to sample air from work areas and exhaust stacks. However, radionuclides other than tritium can be retained in the bubbler water if they are passed through the bubbler collector. An erroneous evaluation of the tritium concentrations could result if these other radionuclides, if present, are not removed from the bubbler water that is counted for tritium.

The time-consuming task of distilling or "milking" the water out of cold trap or Drierite collection system is not required for the bubbler collector. The bubbler collector evaluated in this study would normally be used to collect samples for less than a 30-minute period and would not be easily adaptable for use as a continuous collection system.

The bubbler collector can be advantageously used for specific applications in

the field of tritium health physics because of its simplicity, apparent reliability, and short time requirement between collection and final analysis.

V. SUMMARY

The bubbler collector was tested under varying conditions, and the collection efficiency for tritiated water vapor was consistently >90% and that for tritium gas, <0.1%. Varying the flow rate did not markedly affect the collection efficiency as long as the water did not visibly blow out of the bubbler collector. Changing the water volume in the collector did not affect the collection efficiency when sufficient water was used to give good bubbling action. The optimum water temperature range was from 25 to 35°C. However, at 15°C the collection efficiency for tritiated water vapor changed only from >90 to ~80%. Uniformity of sampling results was within ±10% for concentrations of 20 MPC_a and a 5-minute sample time.

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