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The Canadian Experimental HT Release of June 10, 1987, U.S. Measurements

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THE CANADIAN EXPERIMENTAL HT RELEASE OF JUNE 10, 1987, U.S. MEASUREMENTS

by
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ABSTRACT

In June 1987, an experiment was performed at the Chalk River Nuclear Laboratories in Ontario, Canada, to study the oxidation of elemental tritium (HT) released to the environment. The experiment involved a 30-minute release of 3.54 TBq (95.7 Ci) of HT to the atmosphere at an elevation of one meter. Scientists from six countries participated in the experiment.

The U.S. contribution involved measurements of HT and tritiated water (HTO) concentrations in air during and following the release and after the release, sampling of vegetation for tritium content. Multiple sampling with two different types of air samplers was used. Both types measured the tritium concentration in air by removal of HTO followed by conversion of the HT to HTO, which was collected separately.

The air measurements showed HT concentrations downwind of the release in general agreement with classical atmospheric diffusion (Gaussian) up to the maximum distance measured (400 m). The HTO/HT ratios were shown to slowly increase downwind (~ 4×10^{-5} at 50 m to almost 10^{-3} at 400 m) as conversion of HT took place. After the release, HTO concentrations in the atmosphere remained elevated. Vegetation samples were also taken since the vegetation and associated soil system have been implicated in the oxidation of HT. Freeze-dried water from vegetation samples was found to be low in HTO immediately after the release suggesting a low direct uptake of HTO in air by vegetation. The tritiated water concentration increased during the first day, peaking during the second day (about 15-30 kBq/L of water at 50 m from the source), and decreasing by the end of the second day.

This pattern suggests oxidation in the soil followed by plant uptake through sorption of soil water. This was confirmed by measurements taken by other groups at the experiment site. The HTO in vegetation decreased with distance downwind with the same pattern as the HT measured during the release indicating that the oxidation of HT was linearly related to the HT concentration in the atmosphere during the exposure period. An adequate description of the process can be made through the observed phenomenon of HT deposition into the soil with subsequent rapid oxidation by soil bacteria.

Measurements of organically bound tritium (OBT) in the vegetation were also made. For the tritium release of 30 minutes the OBT accummulated by the vegetation was small compared to the organic hydrogen already present in the existing vegetation. The OBT in the vegetation continued to accumulate during the period following exposure (about 0.4 Bq/g dry weight at 50 m after two days). This was, presumably, a consequence of the photosynthetic fixation of the tritium in the tissue water following the release.

INTRODUCTION

Despite its high specific activity, tritium is usually considered a radionuclide of relatively low risk to human health. It emits a low energy beta that does not penetrate the dead layer of the skin; thus there is no direct emersion dose to radiosensitive cells. However, as an isotope of hydrogen, tritium can be incorporated into the body as water or other organic material where the beta particle can do biological damage. Another important property of tritium is its mobility in living organisms and their environment. In the form of tritiated water, the mobility leads to rapid uptake and transport in the body. However, this same mobility works against isotopic concentration in the body or in any part of the environment.

Nuclear facilities release tritium to the environment in the form of elemental tritium (HT) or tritiated water (HTO) and in lesser amounts as other compounds such as tritiated methane. HT is less of a health concern than HTO because it has a low solubility and is not readily metabolized by humans. As a result the (occupational) biological hazard from an exposure to HT in air is a factor of 25 000 lower than an equal exposure to HTO.[1] Fusion reactors will have large quantities of tritium in their inventories, mainly in the form of HT (or DT) but also including some HTO. The dose commitment to the population surrounding the fusion reactor sites is dependent on the proportion of the released tritium in the forms HT and HTO and the conversion of HT to HTO after release.

Past experience has suggested that conversion of HT in the free atmosphere is very slow.[2 - 9] While radiation-induced conversion in air may be high at high tritium concentrations, at the levels to be expected in the open atmosphere after an accidental release, conversion is slow. Estimates of the residence time of HT in the atmosphere average about 6.5 years.[9]

Oxidation of HT has been demonstrated in soil where many microorganisms are capable of metabolizing hydrogen.[10-16] The rate of oxidation varies with soil moisture and temperature, but oxidation appears to occur in most soils.[14,17,18] In many cases the rate of oxidation appears to be limited by diffusion of hydrogen into the soil. The rate of oxidation of HT in soil is roughly consistent with the residence time of HT in the atmosphere above the soil.

Elemental tritium is also oxidized in higher plants although the rate of oxidation is much slower than in soils and the mechanism of oxidation is not understood.[19-21] Plants also act as the conduit for moving tritium oxidized in the soil to the atmosphere.[22] In spite of the great deal that is known about the oxidation rate in soils and plants, there is very little experience with HT releases to the atmosphere. Most of the existing experience has come from accidental releases which had varying amounts of HTO in the release.[23] The HTO component makes interpretation of these data difficult although it does allow calculation of the upper bound for tritium oxidation. These calculations are consistent with the oxidation rates measured by other means and with the estimated HT residence time in the atmosphere.[24]

About eighteen years ago a series of experiments was conducted in France by scientists interested in atmospheric diffusion. The scientists used tritium as a tracer in these tests. Unexpectedly, the experiments suggested that elemental tritium was rapidly converted to tritiated water in the atmosphere downwind of the release site. Since the experiments were not designed to look at tritium conversion, the results were not disseminated widely but were nevertheless released in a report.[25] The results showed an apparent HT-to-HTO conversion half time of about 22 minutes. It became clear that if the results were valid, the population doses projected for fusion reactor releases would be greatly increased over what had been expected from earlier experience with HT gas.

The international fusion program calls for the building of a DT-burning fusion reactor. The dose to the surrounding population from fusion power reactors is projected to come from routine and possible accidental tritium releases in addition to releases of activation products in both gaseous and particulate forms. Since the inventory of tritium will be high (kilograms), the risk assigned to tritium releases is an important factor in calculating the total risk of a fusion facility. For these reasons, the European Community supported a series of experiments to look further into tritium conversion in the environment.

One of the experiments was conducted at the Chalk River Nuclear Laboratories (CRNL), in Ontario, Canada in June, 1987. Canadian scientists who took part in the experiment were mainly from Ontario Hydro, Ltd. and CRNL. The Europeans were represented by groups from France, Germany, and Sweden. When the Canadians extended an invitation to scientists from Japan and the United States to participate in the experiment, both countries accepted. The Japanese sent a research team from the Japan Atomic Energy Research Institute. There were two U.S. participants: Roland Jalbert (LANL) and Charles Murphy Jr. (SRL). (A third U.S. scientist, who sampled air for HT and HTO during and after the tritium release, also participated but his data are not included here due to uncertainties resulting from problems involving HTO recovery from the silica gel samplers and possible conversion of HT in the HTO samplers.)

The U.S. team focused its attention on measuring HT and HTO concentrations in air during and after the release in addition to analyzing vegetation samples after the release for free-water tritium and organically bound tritium (OBT). The latter activity provided vegetation that had been exposed under controlled conditions and which can be used to study the incorporation of tritium into vegetation organic matter, the subject of ongoing SRL research.

THE EXPERIMENT

A detailed description of the field site, pertinent meteorological data and the release itself may be found elsewhere.[2 6] Briefly, the release site was an open field of approximately 400 meters on the long axis and 200 meters at the widest point on the other axis. Most of the field was covered with grass and moss with occasional small trees. At the end of the field farthest from the release site, the surface soil had been removed leaving an area with very little vegetation. The field was surrounded by a stand of mixed conifer and hardwood trees (Figs. 1-3).

On 10 June 1987, 3.54 TBq (95.7 Ci) of HT was released at one end of the field and at an elevation of 1 meter from the ground between 1520 (H hour) and 1550 EDT (H + 30 min). Wind speed during the release varied between ~2 and 3.5 m/s with a mean value of about 2.4 m/s. The direction varied from about eight degrees south of the center line (300° - the direction of the "x" axis) during the first 5-10 minutes of the release to ~ 25° north of the center line during

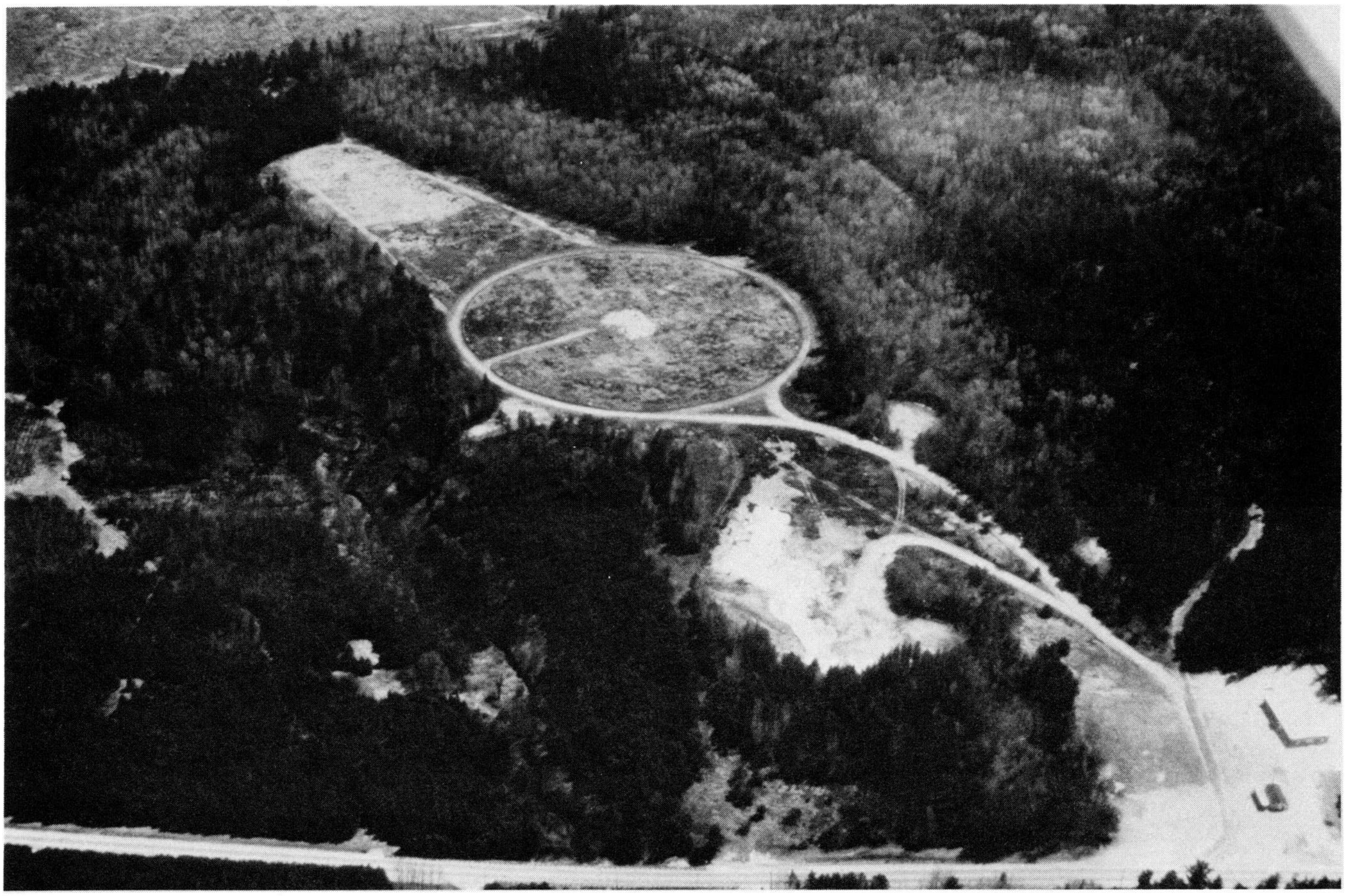

Fig. 1. Aerial view of experimental site at CRNL.

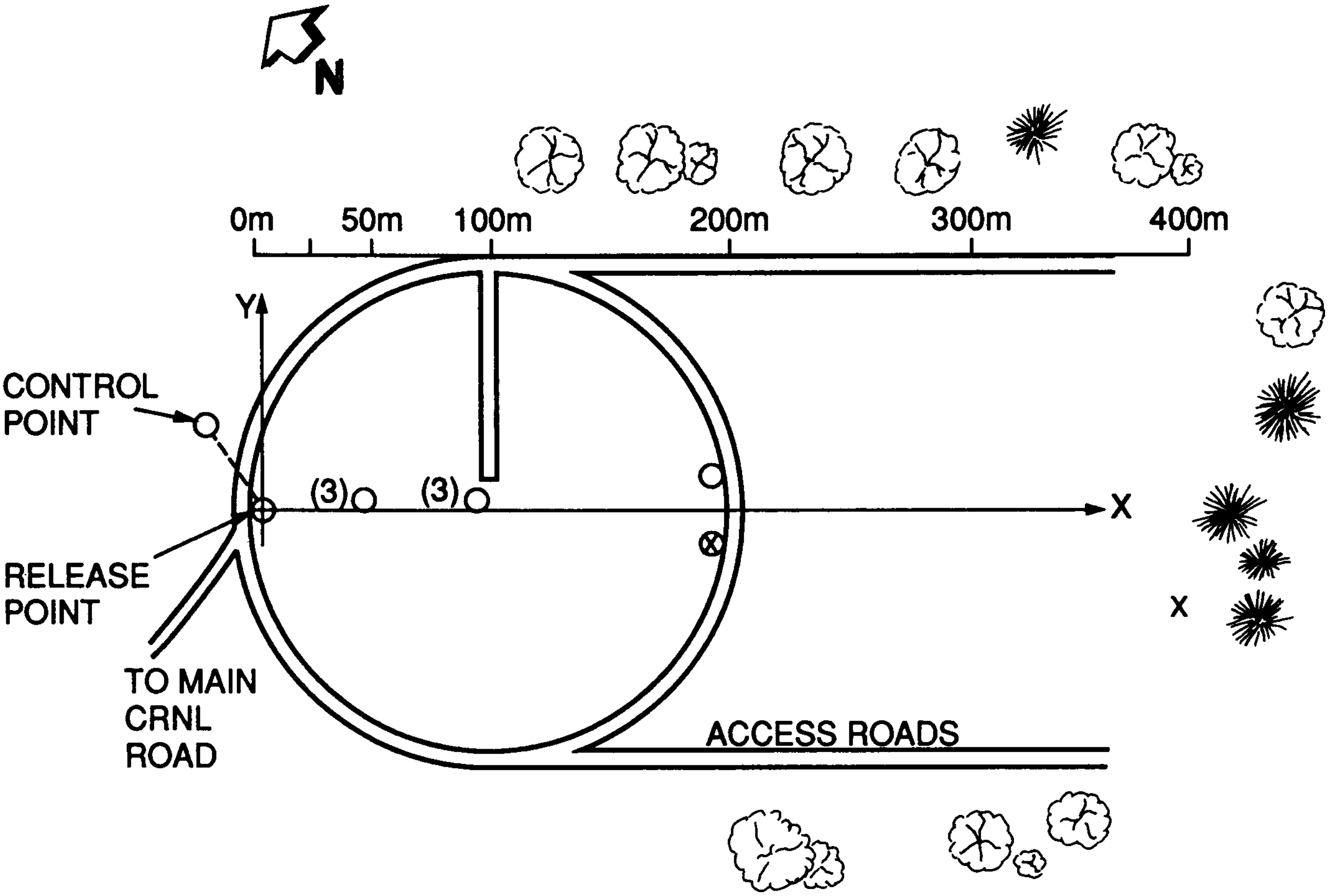

Fig. 2. Map of the experimental area with location of LANL (O) and SRL (X) air samplers.

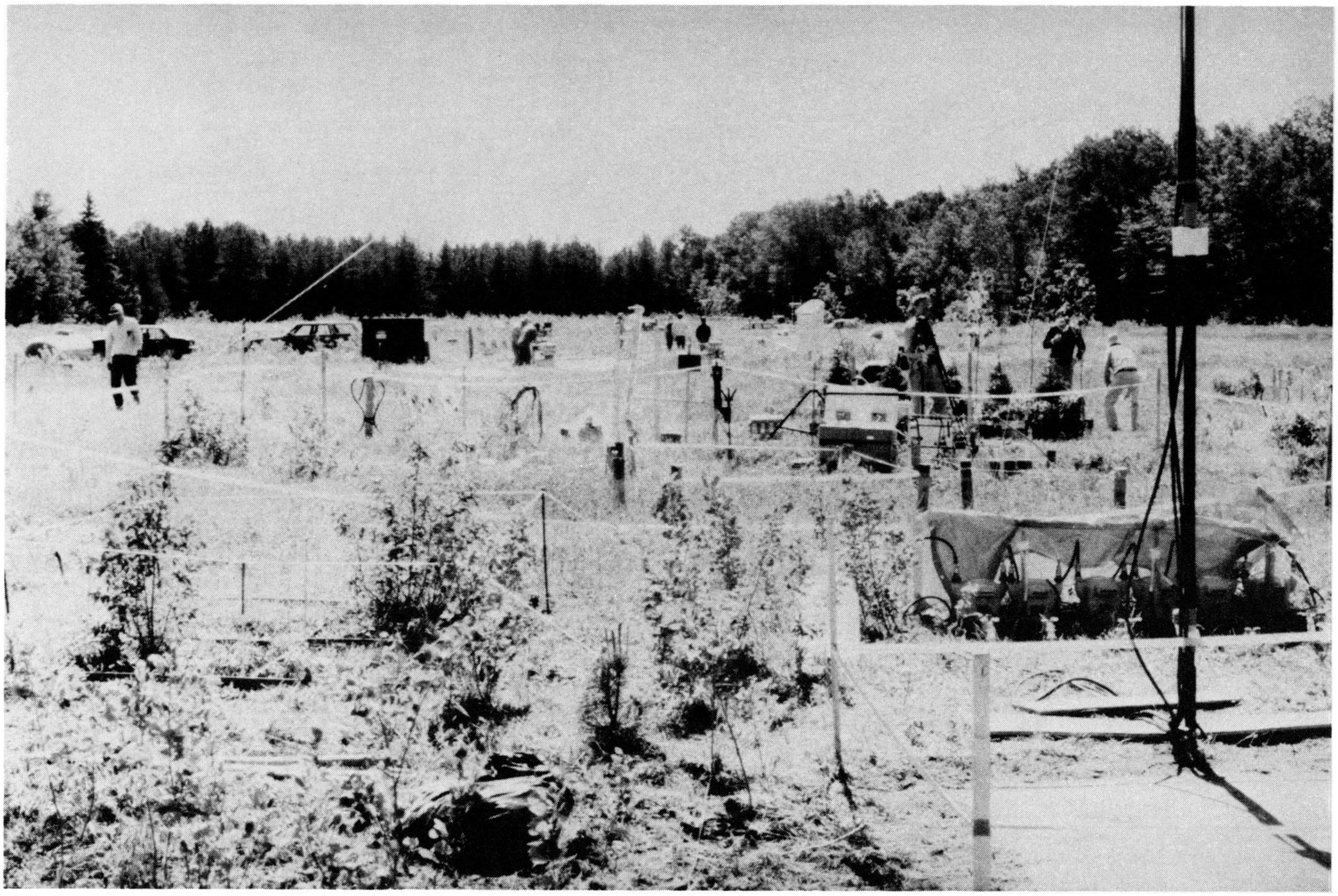

Fig. 3. Downwind view of the field taken about 50 m from the point of release.

the remainder of the release. During the evening and through most of the following day when additional air and vegetation measurements were made, the wind direction did not markedly change, remaining within 240° and 340°. The wind speed likewise maintained its strength (2-3 m/s) except at night (2130, 10 June, to 0800, 11 June) when it dropped to < 1 m/s. Light rain began to fall at about 1500 of the second day (11 June) and continued intermittently during the remainder of the day. This did not affect the U.S. air measurements, which had been concluded by that time.

SAMPLING METHODS

LANL Air Samplers

Water-bubbler-type air samplers were designed and built especially for this experiment. Each sampler consisted of three small bubblers in series (for HTO) followed by a heated catalyst, three more bubblers (for HT), a flowmeter and pump (Figs. 4, 5). The bubblers consisted of 20-mL low-background glass liquid scintillation vials filled with 10 mL of low activity water furnished by CRNL. The catalyst cartridge was constructed of a 28 cm long x 1.3 cm diameter copper tube filled initially with brass chips (serving as the preheater) followed by 10 g of an Englehard precious metal catalyst (on an alumina substrate). The tube was heated by a heat tape, which, with light fiber glass insulation, resulted in a catalyst temperature of ~ 400°C. The catalyst cartridge was preceded and followed by several turns of copper tubing to prevent heating of the bubblers. Following the catalyst, cooling coil and HT bubblers were a flowmeter (with valve) and an air pump located on the ground. The flowmeter's needle valve was used to maintain a sampling flow rate of 1.0 Lpm, which was verified by external calibration. The flow rate of each sampler was checked after each sampling run. The catalyst heaters were turned on by plugging them into a local power outlet at least 30 minutes before the sampling began. The actual sampling was controlled by starting and stopping the pumps remotely through cables and solid-state relays located at the samplers.

Three LANL samplers were placed close together at 50 m downwind from the source. Three more, also close together, were located at 96 m and the remaining two at 183 m downwind and 18 m apart (Fig. 2). The reason for bunching the samplers at 50 and 96 m was to get a time profile of the tritium air concentration during the release. The attempt was marginally successful because of the low sampling flow rate (1 L/min) and counting problems discussed below.

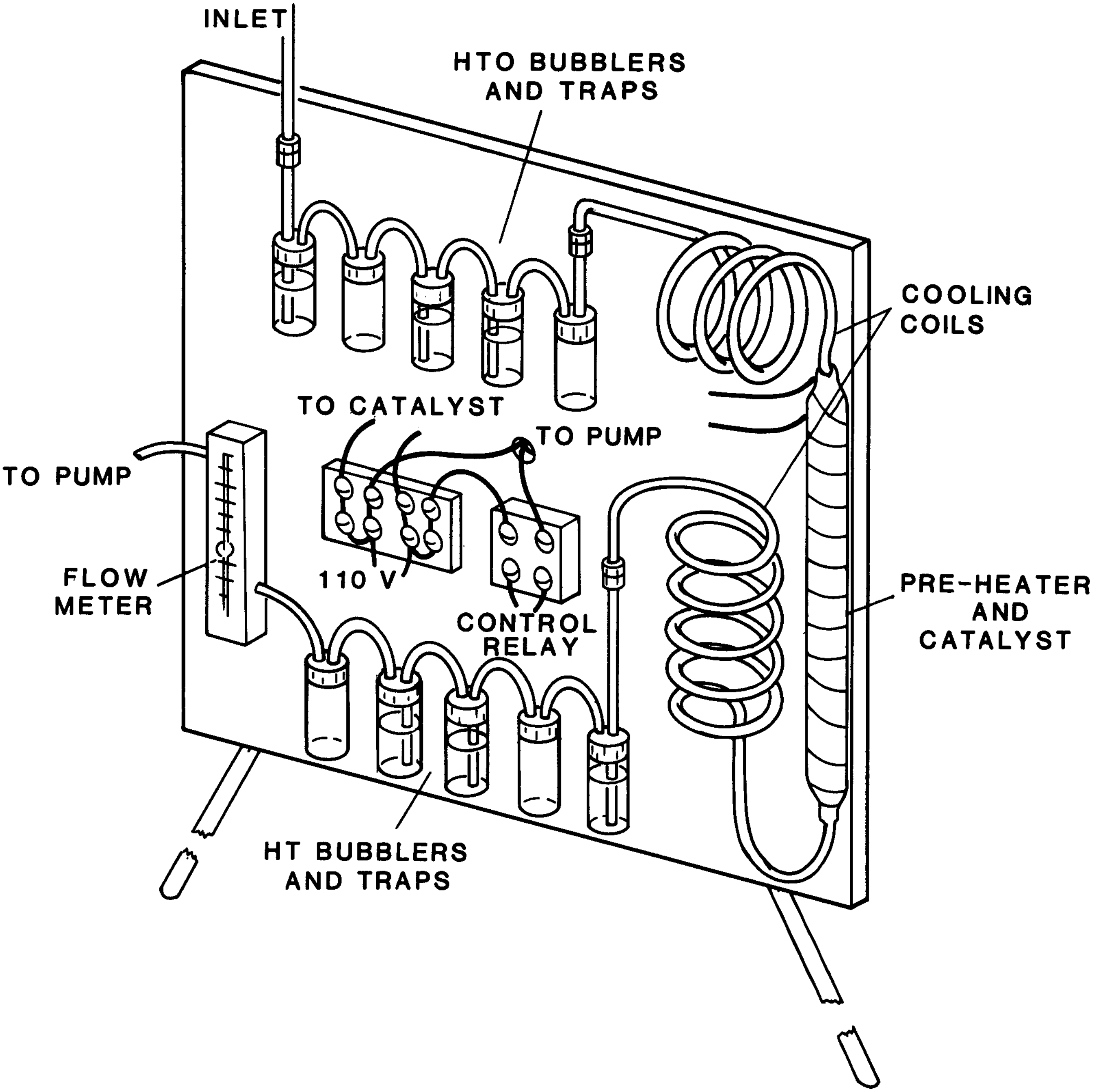

Fig. 4. Drawing of LANL field air sampler.

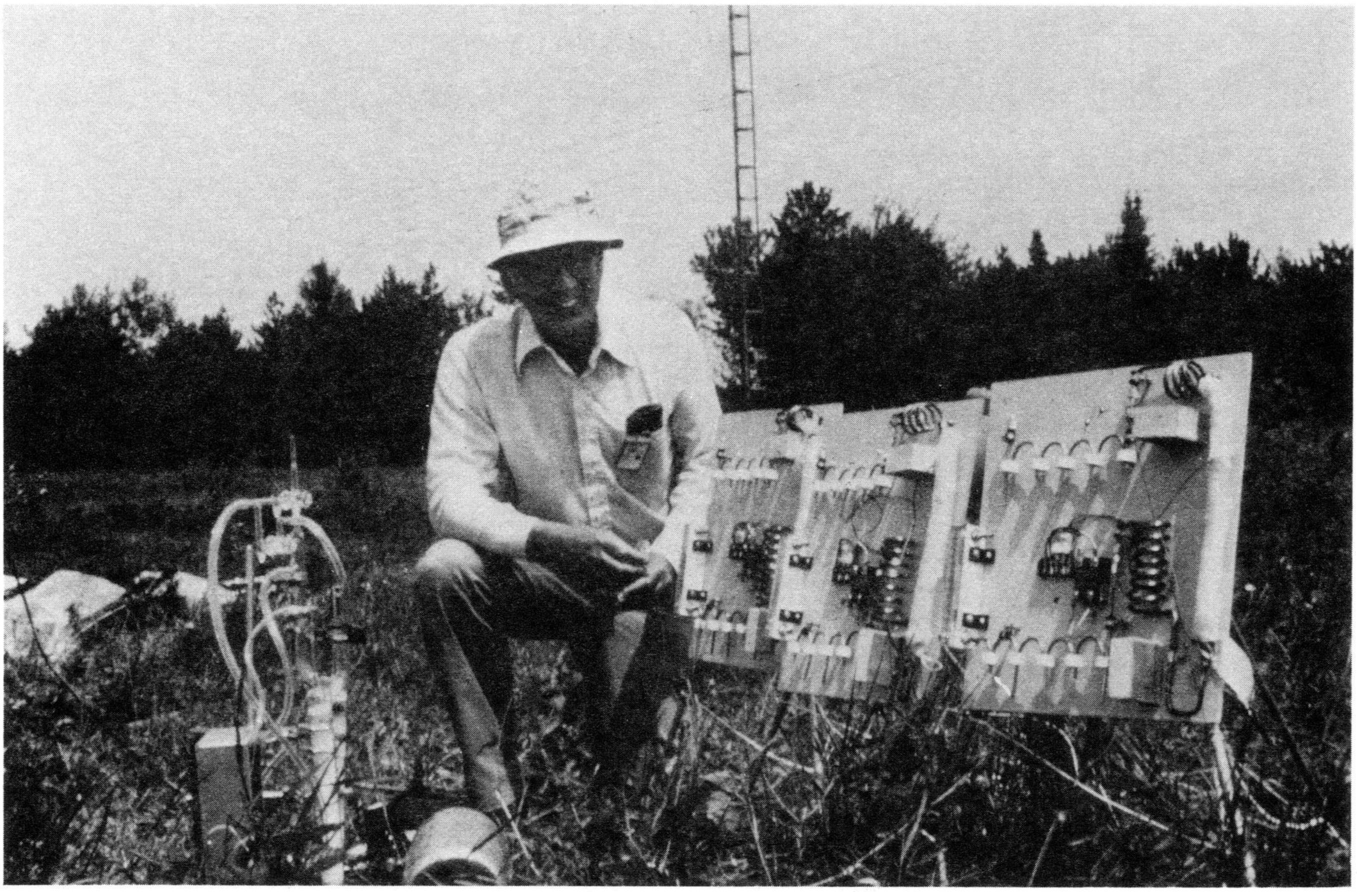

Fig. 5. Photograph of LANL air samplers located at 50 meters. The wooden blocks are used for stacking the samplers during shipment.

After each run the vials were removed and replaced with fresh vials prior to the next run. Once in Los Alamos, the amount of water in each vial, which affects counting efficiency, was determined by weighing, after which 10 mL of cocktail was added to the water prior to counting. Only the first two HTO vials were used for the HTO determination in order to eliminate the possibility of adding backstreaming tritium from the catalyst. For the HT determination, the total tritium collected during the release was determined by adding all the tritium collected in the HT vials during the experiment including the two runs after the end of the release. In addition, the remaining tritium trapped on the catalyst and associated plumbing was leached out at Los Alamos and added to the tritium collected in the vials. The small bubblers were found to be about 90% efficient in collecting the HTO at the flow rate of 1 L/min. The solubility of the HT in the HTO bubblers limited the HTO/HT ratio that could be determined to a minimum of about 10^{-5}. The cocktail used was chosen because of its good efficiency: 25% with a mixture of 10 mL water and 10 mL cocktail. Unfortunately it produced twice the usual background counting rate (36 c/min), which was unexpected, and resulted in lower sensitivity and poorer statistics.

SRL Air Samplers

Air sampling by SRL was done with an SRL HTO/HT sampler (Figs. 6, 7) based on an Oslund-Mason design modified for high volume sampling.[27] The system consists of a pump which draws 50 liters of air per minute through two collection cylinders each filled with 350 g of Davison 4A molecular sieve. Hydrogen carrier gas is added to the sampled air stream (150 mL/min) before the air enters the first cylinder, which contains uncoated molecular sieve for trapping the HTO. The second cylinder contains palladium-covered molecular sieve for HT. The palladium catalyzes the oxidation of hydrogen (including HT) to water, which is collected on the sieve. The hydrogen gas reportedly increases the catalyst efficiency in addition to making the subsequent recovery of HTO from the coated molecular sieve more easily quantitative.

Both types of molecular sieve were received from the manufacturer in sealed metal cans. The cans were opened as near to the time of sample collection as practical and the sieve poured into the cylinders. At the end of a sampling period of one hour, the pump was stopped, the collection cylinders were removed and sealed, and fresh collection cylinders installed on the samplers. The used molecular sieve was transferred in the field to glass jars which were sealed for later laboratory analysis. The two SRL air samplers were located close to the field center line at 50 m and 400 m (Fig. 2).

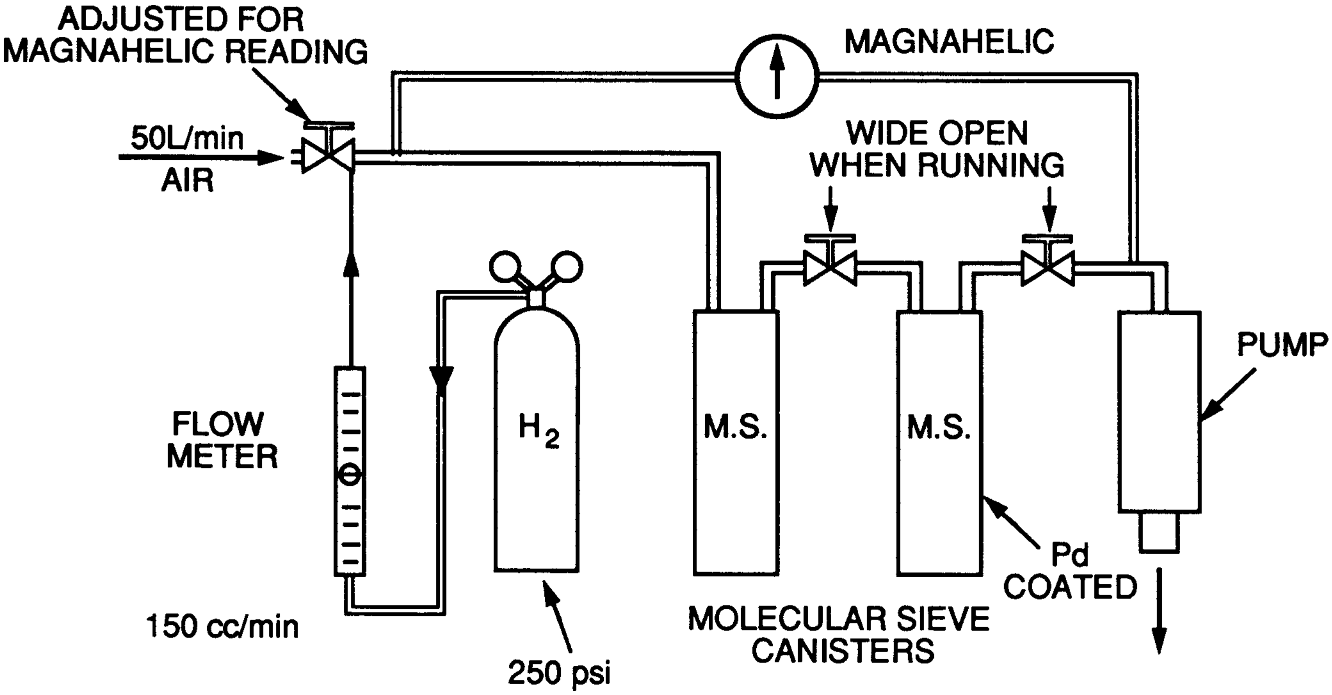

Fig. 6. Sketch of the SRL high-volume air sampler.

Fig. 7. Savannah River Laboratory air sampler. The view is of the field looking upwind toward the release point.

At the Savannah River Laboratory, the water was removed from the sieve by heating in a dry gas stream and trapping the water on a cold finger. The tritium activity of the samples was measured by liquid scintillation counting.

Vegetation Analysis

After the release, samples of vegetation were taken at 50, 180 and 400 m downwind from the point of release. The samples were collected in jars, frozen and shipped frozen to SRL, where about 50 g of the vegetation samples was subsampled for analysis. The remaining material was stored as a backup in case confirmation of the results was needed later. The samples were vacuum-distilled to remove the water, which was collected in a cold trap and saved for later counting. The dry vegetation was subjected to a stream of water-saturated air at 60°C for 48 hours to recover any remaining free HTO. Subsamples were taken at daily intervals (3 total) to test for removal of free exchangeable tritium. The results of the water-vapor-exchange procedure indicated that the procedure removed all of the free tritium. The dry samples were then combusted in a furnace using an oxygen and argon gas mixture. The water of combustion was collected in a cold trap and subsequently analyzed by liquid scintillation counting. Because of the possibility of quenching of the combustion samples prepared for scintillation counting, the water samples were counted twice using an internal tritium spike. All OBT data presented used the combustion results following 4-8 hours of "vapor washing."

RESULTS

The LANL results are shown in Tables 1 and 2. The counting standard deviations are listed as well as the additional uncertainties due to impurities accidentally added to a few of the samples at Los Alamos and to an uncertain sampling flow rate of sampler No. 8. Table 3 shows the results of air sampling by SRL. The results of the first two SRL samples taken before the release can be considered background for the combination of location and the samplers.

All the air samples taken during the release show the expected decrease in concentration of HT with distance from the release. The samples taken during the release show an increase in the concentration of HTO significantly greater than would be predicted by small-scale conversion experiments in air.[2-5] The increase is assumed to come mainly from the oxidation of the HT in the soil. However, the increase is much smaller than suggested by the early French experiment[25] but consistent with what might be expected from previous soil experiments with tritium.

TABLE 1

Sampler Number	Location(1) x,y,z (m)	Sampling Time(2) H + (hr:min)	[HT](3) Bq/m^3	[HTO] Bq/m^3	[HTO]/[HT]	[HTO] Statistical and other uncertainties (Bq/m^3)	
						σ	(4)
1	50.5,3.5,0.9	H+(0:1 to 0:11)	3.0E+06	1.6E+02	5.2E-05	±9E+00	-
2	50.5,3.5,0.9	H+(0:11 to 0:20)	3.2E+06	1.1E+02	3.5E-05	±1.0E+01	-
3	50.5,3.5,0.9	H+(0:20 to 0:30)	1.7E+06	5.9E+01	3.4E-05	±7E+00	-
4	96,2.0,0.9	H+(0:2 to 0:12)	1.3E+05	1.1E+02	8.7E-04	±1.0E+01	-
5	96,2.0,0.9	H+(0:12 to 0:21)	5.7E+05	3.8E+01	6.7E-05	±7E+00	-
6	96,2.0,0.9	H+(0:21 to 0:31)	5.6E+05	9.8E+01	1.8E-04	±7E+00	-
7	183,-10,1.0	H+(0:2 to 0:32)	5.1E+04	2.5E+01	4.9E-04	±3E+00	-
8	183,8,1.0	H+(0:2 to 0:32)	6.5E+04	8.5E+01	1.3E-03	±6E+00	±9E+00

Notes: (1) See Fig. 1 for location of samplers.

(2) H refers to time release began: 15:20 (EDT), 10 June 1987.

(3) For [HT], $\sigma < 1\%$. For sampler No. 8 at H+(0:2 to 0:32) add an uncertainty of 0.70E+4 due to flow rate drop. This does not affect [HTO]/[HT] since both values are similarly affected.

(4) Uncertainty due to change in flow rate.

Table 1. Results of LANL air sampling during the release (10 June 1987).

TABLE 2

Sampler Number	Location(1) x,y,z (m)	Sampling Time(2) H + (hr:min)	[HTO] Bq/m^3	[HTO] Statistical and other uncertainties (Bq/m^3)		
				σ	(4)	(5)
1	50.5,3.5,0.9	H+(1:50 to 2:58)	2.6E+02	±3E+00	-	-
2	50.5,3.5,0.9	H+(1:50 to 2:58)	2.4E+02	±2E+00	-	-
3	50.5,3.5,0.9	H+(2:58 to 3:58)	2.0E+02	±2E+00	-	-
4	96,2.0,0.9	H+(1:50 to 2:58)	8.3E+01	±2E+00	-	-
5	96,2.0,0.9	H+(1:50 to 2:58)	7.0E+01	±1E+00	-	-
6	96,2.0,0.9	H+(2:58 to 3:58)	1.0E+02	±2E+00	-	-
7	183,-10,1.0	H+(1:50 to 2:58)	1.4E+01	±1E+00	-	-
8	183,8,1.0	H+(1:50 to 2:58)	4.0E+01	±2E+00	±4E+01	-
1	50.5,3.5,0.9	H+(18:59 to 19:59)	2.9E+02	±3E+00	-	-
2	50.5,3.5,0.4	H+(18:59 to 19:59)	3.6E+02	±4E+00	-	-
3	50.5,3.5,0.07	H+(18:59 to 19:59)	4.6E+02	±5E+00	-	±4.6E+01
4	96,2.0,0.9	H+(18:59 to 19:59)	7.4E+01	±2E+00	-	±7E+00
5	96,2.0,0.4	H+(18:59 to 19:59)	9.0E+01	±2E+00	-	±9E+00
6	96,2.0,0.09	H+(18:59 to 19:59)	1.1E+02	±2E+00	-	±1.1E+01
7	183,-10,1.0	H+(18:59 to 19:59)	2.4E+01	±2E+00	-	±2E+00
8	183,8,1.0	H+(18:59 to 19:59)	3.4E+01	±2E+00	±5E+01	±3E+00

Notes: (5) Uncertainty due to cocktail poisoning. See Table 1 for explanations of other notes.

Table 2. Results of LANL air sampling following the release (10-11 June 1987).

TABLE 3

Sampler Number	Location(1) x,y,z (m)	Sampling Time(2) H + (hr:min)	[HT] Bq/m^3	[HTO] Bq/m^3	[HTO]/[HT]	[HT] Error(3) σ Bq/m^3	[HTO] Error(3) σ Bq/m^3
1	180,10,0.75	H-(24:32 to 23:51)	5.00E-01	5.22E+00	1.04E+01	7.40E-03	3.70E-02
2	400,30,0.5	H-(24:28 to 23:47)	3.81E-01	4.48E+00	1.17E+01	3.70E-03	3.70E-02
1	180,10,0.75	H+(0:00 to 0:38)	2.47E+05	2.89E+01	1.17E-04	1.73E+03	8.51E-01
2	400,30,0.5	H+(-0:2 to 0:41)	5.00E+04	4.00E+01	8.00E-04	5.00E+02	4.07E-01
1	180,10,0.75	H+(19:09 to 19:40)	2.11E+01	8.25E+00	3.19E-01	4.07E-01	3.70E-02
2	400,30,0.5	H+(19:15 to 19:44)	5.87E+00	3.12E+00	5.32E-01	1.48E-02	1.48E-02

Notes: (1) See Figure 1 for location of samples.

(2) H refers to time release began: 15:20 (EDT), 10 June 1987.

(3) Statistical as well as other errors are included.

Table 3. Results of SRL Air Sampling.

Measurements taken on the following day show that the concentration of HTO in the air remains elevated over that found before the release. The source of this HTO is likely to be the HT oxidized and remaining in the soil and the vegetation. This also has important implications when calculating potential population doses following a release of HT to the environment.

The concentrations of HT also remained slightly elevated. This could result from some HT absorbed into soil and vegetation. However, it is also possible that it is an artifact caused by the slow release of HT sorbed on the metal surfaces of the air samplers.

Vegetation Sampling

Pine needles and maple leaves were sampled at 400 m whereas grass was sampled at the shorter distances. Table 4 shows the location of the vegetation samples and the results of the analysis. The results for the two sampling directions from the release point are illustrated in Fig. 8. As expected, the concentration of HTO in the vegetation decreased with distance from the release point. This was the result of the lower air concentration to which the soil and vegetation were exposed during the release.

At all of the sites the concentration in the vegetation was low immediately following the release. This suggests that little HTO in air was picked up by plant leaves through exchange and that oxidation of HT in the vegetation was small. Seventeen hours later there was a significant increase in the vegetation HTO concentration. The source of the HTO was the soil where oxidation had taken place during the release. The last samples taken at 50 m were air-dried before vacuum distillation and may not be as reliable as the earlier samples but they do follow the expected pattern of decrease as the HTO in the soil is depleted.

Only at the 50-m site were the concentrations of OBT high enough to be detectable above background using our techniques. The OBT concentration in the organic material was higher than that found in the plant free-water. This may reflect the history of tritium releases in the area. It is also possible that this was another demonstration of higher tritium combustion than free-water tritium under natural environmental conditions.[28]

The OBT increased during the first two days after the release (Fig. 9). Organically bound tritium was still increasing after the vegetation free-HTO had begun to decrease. Organically bound tritium has a much slower turnover than HTO in the vegetation free water. It is likely that most of the OBT was the result of photosynthetic fixation of tritium from the plant

TABLE 4

Location of Sample x,y (m)	Sampling Date Mo., Day	Sampling Time(1) (Hours)	Free HTO Bq/L	Free HTO Counting Error (σ) Bq/L	OBT Tritium Bq/L	OBT Tritium Counting Error (σ) Bq/L
50,0	6,10	-6.93	6.72E+01	3.61E+01	6.77E+02	3.89E+02
50,0	6,10	0.65	4.46E+03	4.67E+02	6.58E+02	2.25E+02
50,0	6,11	17.57	1.50E+04	1.52E+03	9.85E+02	2.32E+02
50,0	6,11	23.66	1.33E+04	1.35E+03	1.02E+03	5.14E+02
50,0	6,19	186.17	2.37E+03	2.68E+02	8.43E+02	2.26E+02
50,10	6,10	-6.83	2.25E+01	3.26E+01	3.91E+02	3.14E+02
50,10	6,10	0.68	1.75E+03	1.97E+02	4.64E+02	2.63E+02
50,10	6,11	17.67	3.00E+04	3.03E+03	2.07E+03	3.91E+01
50,10	6,11	23.77	1.83E+04	1.85E+03	3.00E+03	3.08E+02
50,10	6,19	186.17	2.52E+03	2.76E+02	1.13E+03	1.26E+02
180,0	6,10	-7.13	2.52E+01	3.28E+01		
180,0	6,10	0.57	2.12E+02	4.67E+01		
180,0	6,11	17.37	2.13E+03	2.36E+02		
180,0	6,11	23.44	1.22E+03	1.45E+02		
180,-40	6,10	-7.03	6.72E+01	3.61E+01		
180,-40	6,10	0.61	1.47E+02	4.10E+01		
180,-40	6,11	17.47	1.69E+03	1.92E+02		
180,-40	6,11	23.55	9.69E+02	1.20E+02		
350,-100	6,10	-7.23	2.79E+01	3.30E+01		
350,-100	6,10	0.54	5.47E+01	3.51E+01		
350,-100	6,11	17.27	3.47E+02	5.91E+01		
350,-100	6,11	23.22	1.69E+02	4.29E+01		
400,-15	6,10	-7.33	6.90E+01	3.62E+01		
400,-15	6,10	0.50	9.70E+01	3.85E+01		
400,-15	6,11	17.17	5.60E+02	7.29E+01		
400,-15	6,11	23.33	1.70E+02	4.29E+01		

(1) Measured from end of release: 15:50 (EDT), 10 June 1987.

Table 4. Tritium activity in vegetation samples.

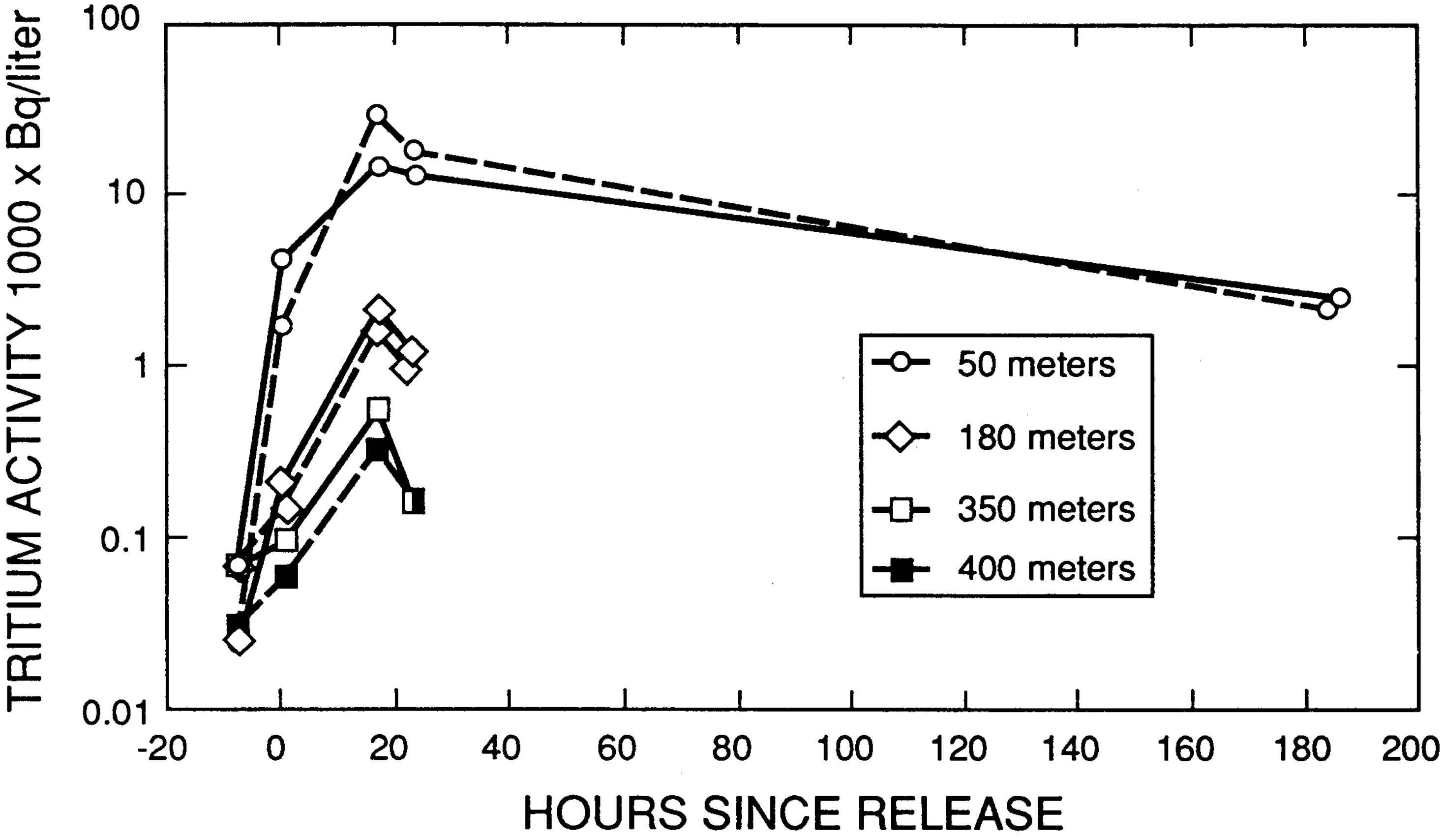

Fig. 8. Vegetation free-HTO at four distances. At a given distance the solid lines represent the northernmost samples. (Refer to Table 4 for location of samples).

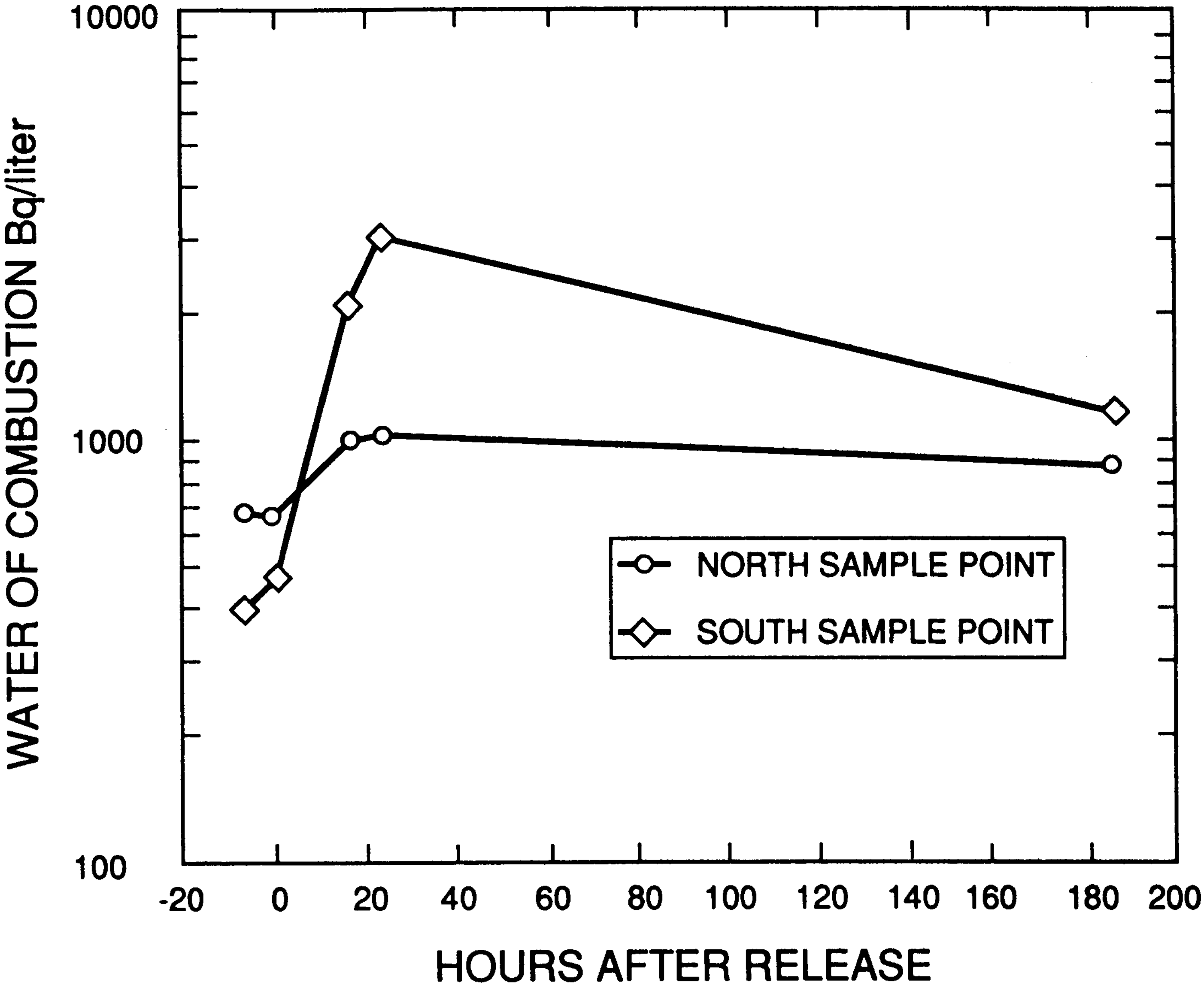

Fig. 9. Organic tritium content of vegetation at 50 m.

HTO.[29, 30] Therefore it is not surprising that by the second week, the concentration of OBT had decreased considerably. The decrease can be explained by the transport of some of the labeled compounds to other parts of the plant and the dilution of the labeled plant material with new growth which was not as highly labeled because of the decreasing HTO content of the vegetation.

Figure 10 shows the relationship of HT concentration in air measured by SRL during the release and the peak HTO concentration in the vegetation after the release. The slopes of the concentrations (on log-log paper) are nearly identical as would be expected if soil HT oxidation were proportional to air HT concentration. However, the slope of leaf HTO concentrations would be much less steep if the air-leaf pathway were the more important one for the leaf free-HTO.

CONCLUSIONS

Although there is a discrepancy at 180 m between the LANL and SRL air samples, the results of all the air analyses are internally consistent. It is clear that oxidation rates are higher than would be predicted by laboratory tritium conversion experiments but much lower than had apparently resulted from the early French experiment.[25] It appears from the present experiment and other recent similar release experiments in Canada and France in addition to small scale laboratory tritium and field experiments involving soils that soil bacteria play an important role in elevating the concentration of HTO in the area of an HT release. In spite of the uncertainties in the HTO data presented here, there appears to be little doubt that the numbers are valid within a factor of 2 at the most.

The OBT in the grass leaf organic matter immediately after exposure to HT is not detectably different from that in the controls. Therefore any direct uptake of HT into vegetation organic matter must take place at a very slow rate. The concentration of bound tritium in the vegetation increases slowly after the release. The increase in OBT after the release can be explained by photosynthetic fixation of tritium from the HTO entering the plant from the soil. The decrease in OBT concentration a week following the release can be explained by a combination of tritiated products of photosynthesis being translocated from the leaves and the dilution of tritiated organic material with organic matter of lower tritium content produced after the peak HTO activity.

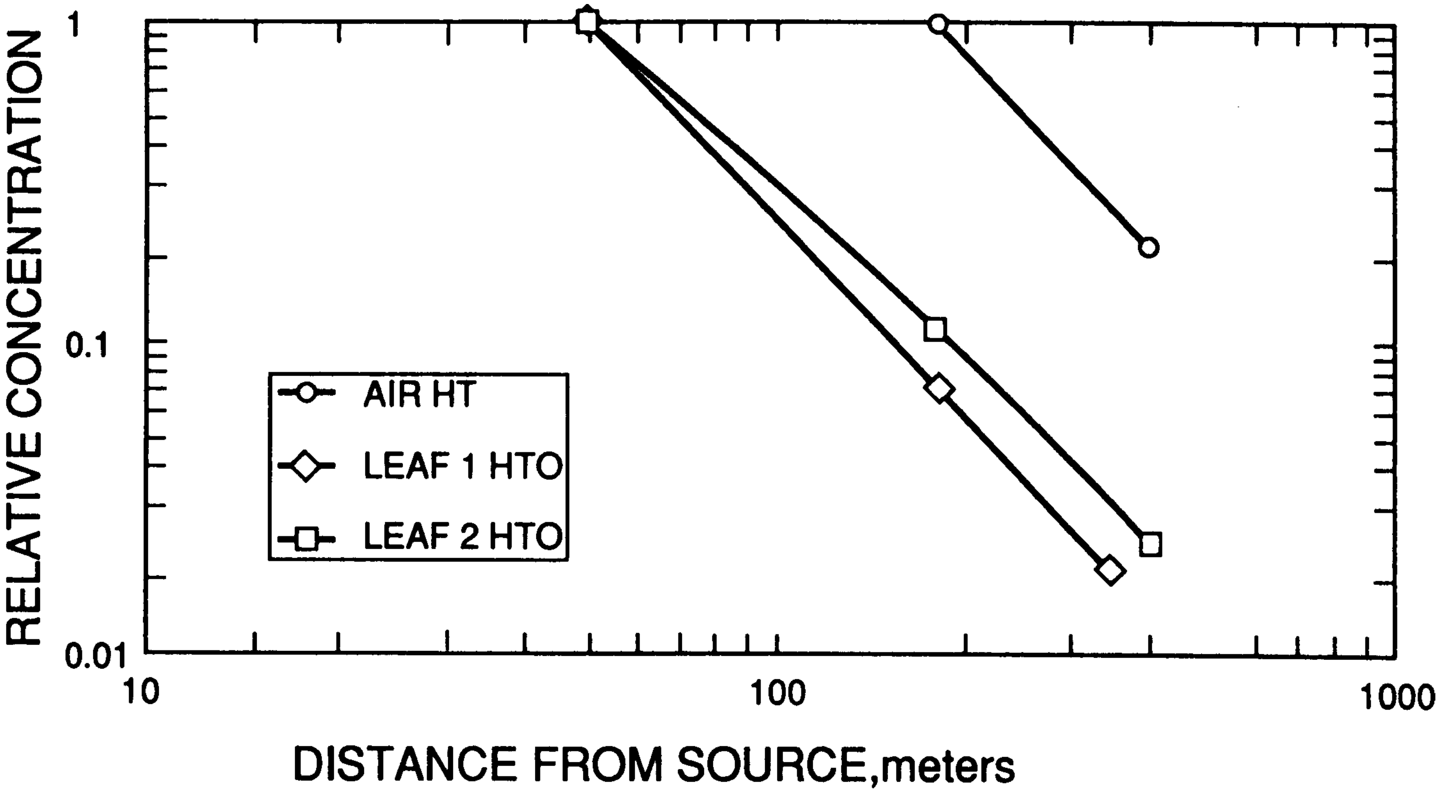

Fig. 10. Relative concentrations of tritium in air and vegetation free-HTO.

The concentration of HTO in the vegetation following the release suggests that direct oxidation in the vegetation takes place very slowly. The increase in HTO in the vegetation found some hours after the release must enter the plant from some other source. The source of water, and thus HTO, is most likely to be the soil.

The results of the measurements do not suggest the existence of any phenomenon which would cause very high rates of HT oxidation in the environment. They do confirm that relatively low rates of oxidation do occur in conjunction with the biotic components of terrestrial eco-systems. In general, the oxidation rates and HTO concentrations agree quite well with what would have been expected from recent past experiments.

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