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U.S. TEAM MEASUREMENTS DURING THE JUNE 1987 EXPERIMENTAL HT RELEASE AT THE CHALK RIVER NUCLEAR LABORATORIES, ONTARIO, CANADA

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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 HT in the environment. The experiment involved a 30-minute release of 100 Ci of HT to the atmosphere at an elevation of one meter. The HTO/HT ratios were shown to slowly increase downwind ($\sim 4 \times 10^{-5}$ at 50 meters to almost 10^{-3} at 400 meters) as conversion of HT takes place. For several days after the release, HTO concentrations in the atmosphere remained elevated. Freeze-dried water from vegetation samples was found to be very low in HTO immediately after the release suggesting a very low direct uptake of HTO in air by vegetation. The tritiated water concentration increased during the first day, peaking during the second day (about 400-600 pCi/ml of water at 50 meters from the source) and decreasing by the end of the second day. The organically bound tritium continued to accummulate during the period following exposure (about 10 pCi/gm dry weight at 50 meters after two days).

INTRODUCTION

The second in a series of recent major experimental tritium release experiments was performed in June 1987 at the Chalk River Nuclear Laboratories in Ontario, Canada. The purpose of these experiments was to study the fate of elemental tritium in the environment. Scientists from six countries took part in the experiment. United Stated participants focused their 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).

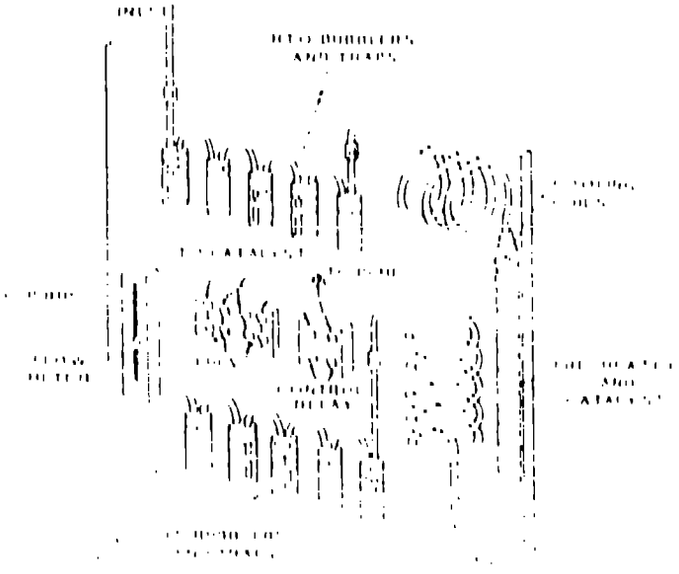
THE EXPERIMENT

A detailed description of the field site, pertinent meteorological data and of the release itself may be found elsewhere.[1] Briefly, the release site was an open field 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 point of release, 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 (Fig. 1).

Fig. 1. Downwind view of the experimental field.

One hundred curies of HT were released at an elevation of 1 meter from the ground between 1520 and 1550 EDT on 10 June 1987. Wind speed during the release varied between [illegible] 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 centerline (300°) during the first 5-10 minutes of the release to [illegible] 25° north of the centerline during 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 [illegible] 1 m/s. Light rain began to fall at about 1500 of the second day (11 June) and continued on and off during the remainder of the day. This did not affect the U.S. air measurements, which had been concluded by that time.

water-bubbler-type air samplers were signed and built by the Los Alamos National boratory (LANL) especially for this experi- nt. Each sampler consisted of three small bblers in series (for HTO) followed by a ated catalyst, three more bubblers (for HT), flowmeter and pump (Fig. 2). The bubblers re 20-mL low-background-glass liquid scintil- tion vials filled with 10 mL of water. The talyst cartridge consisted of a copper tube lled initially with brass chips followed by a ecious metal catalyst heated to 400°C. llowing the catalyst, cooling coil and HT bblers were a flowmeter and an air pump. The bbler flow rate of 1.0 Lpm was checked after ch sampling run. The catalyst heaters were rned on by plugging them into a local power tlet at least 30 minutes before the sampling gan. The sampling during the release was ntrolled by starting and stopping the pumps notely through cables. Only the first two O vials were used for the HTO determination order to eliminate the possibility of adding ckstreaming tritium from the catalyst. For e HT determination, the total tritium col- cted during the release was determined by ding all the tritium collected in the HT als during the sampling runs including the o runs after the end of the release. In dition, the remaining tritium trapped on the talyst and associated plumbing was leached t at Los Alamos and added to the tritium llected in the vials. The solubility of the in the HTO bubblers limited the HTO/HT ratio at could be determined to a minimum of about . Three LANL samplers were placed close gether at 50 meters downwind from the source ar the center line. Three more, also close gether, were located at 96 meters and the maining two at 183 meters downwind and 18 meters apart. The reason for bunching the samplers at 50 and 96 meters was to attempt to get a time profile of the tritium air concentration during the release.



g. 2. Sketch of LANL field air sampler.

Air sampling by the Savannah River Laboratory (SRL) was done with two HTO/HT samplers (Fig. 3) based on the Oslund-Mason design modified for high volume sampling.[2] The system consists of a pump which draws 50 liters of air per minute through two collection cylinders each filled with 350 grams of Davison 4A molecular sieve. Hydrogen carrier gas is added to the sampled air stream (150 cc/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 samplers were run for a period of one hour. At the end of a sampling period the pump was stopped, the collection cylinders were removed and sealed, and fresh collection cylinders were 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 samplers were located near the centerline at 183 m and 350 m from the release point.

Fig. 3. Test area looking upwind toward the release point. An SRL air sampler is shown in the foreground.

After the release, samples of vegetation were taken at 50, 180, 350 and 400 meters downwind from the point of release and sealed in plastic bags. At SRL the samples were vacuum distilled to remove the free water, which was collected in a cold trap and saved for later counting. The dry samples were then combusted in a furnace using an oxygen and argon gas mixture. The activity of the OBT was determined by liquid scintillation counting.

RESULTS

Table 1 shows the results of the air

TABLE 1. Results of LANL and SRL air sampling during and after the release.

Sampler Number	Location[1] x,y,z (m)	Sampling Time[2] H + (hr:min)	[HT] Bq/m^3	[HTO] Bq/m^3	[HTO]/[HT]
LANL 1	50.5,3.5,0.9	H+(0:1 to 0:10.6)	3.0E+06	1.6E+02	5.2E-05
2	50.5,3.5,0.9	H+(0:10.6 to 0:20.1)	3.2E+06	1.1E+02	3.5E-05
3	50.5,3.5,0.9	H+(0:20.1 to 0:29.6)	1.7E+06	5.9E+01	3.4E-05
4	96,2.0,0.9	H+(0:2 to 0:11.6)	1.3E+05	1.1E+02	8.7E-04
5	96,2.0,0.9	H+(0:11.6 to 0:21.1)	5.7E+05	3.8E+01	6.7E-05
6	96,2.0,0.9	H+(0:21.1 to 0:30.8)	5.6E+05	9.8E+01	1.8E-04
7	183,-10,1.0	H+(0:2 to 0:32)	5.1E+04	2.5E+01	4.9E-04
8	183, 8,1.0	H+(0:2 to 0:32)	6.5E+04	8.5E+01	1.3E-03
1	50.5,3.5,0.9	H+(1:50 to 2:58)	-	2.6E+02	-
2	50.5,3.5,0.9	H+(1:50 to 2:58)	-	2.4E+02	-
3	50.5,3.5,0.9	H+(2:58 to 3:58)	-	2.0E+02	-
4	96,2.0,0.9	H+(1:50 to 2:58)	-	8.3E+01	-
5	96,2.0,0.9	H+(1:50 to 2:58)	-	7.0E+01	-
6	96,2.0,0.9	H+(2:58 to 3:58)	-	1.0E+02	-
7	183,-10,1.0	H+(1:50 to 2:58)	-	1.4E+01	-
8	183,8,1.0	H+(1:50 to 2:58)	-	4.0E+01	-
1	50.5,3.5,0.9	H+(18:59 to 19:59)	-	2.9E+02	-
2	50.5,3.5,0.4	H+(18:59 to 19:59)	-	3.6E+02	-
3	50.5,3.5,0.07	H+(18:59 to 19:59)		4.6E+02	
4	96,2.0,0.9	H+(18:59 to 19:59)	-	7.4E+01	-
5	96,2.0,0.4	H+(18:59 to 19:59)	-	9.0E+01	-
6	96,2.0,0.09	H+(18:59 to 19:59)	-	1.1E+02	
7	183, 10,1.0	H+(18:59 to 19:59)		2.4E+01	
8	183,8,1.0	H+(18:59 to 19:59)		3.4E+01	-
SRL 1	180,10,0.75	H (24:32 to 23:51)	5.00E 01	5.22E+00	1.04E+01
2	400,30,0.5	H (24:28 to 23:47)	3.81E 01	4 48E+00	1.17E+01
1	180,10,0.75	H+(00:00 to 00:38)	2.47E+05	2.89E+01	1 17E 04
2	400,30,0.5	H+(19:12 to 19:43)	5.00E+04	4.00E+01	8.00E 04
1	180,10,0.75	H+(19:09 to 19:40)	2.11E+01	8.25E+00	3.19E 01
2	400,30,0.5	H+(19:15 to 19:44)	5.87E+00	3.12E+00	5.32E 01

Notes: [1] X is the distance from the source along the centerline. Y (+) is toward the north, Z is the vertical distance from the ground to the sampler intake.

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

sampling by LANL and SRL. The first two S.L samples were taken before the release and can be considered background for the combination of location and the samplers. All of the 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 with distance. The increase in the HTO/HT ratio is greater than would be predicted by HT to HTO conversion rates in air determined by small scale laboratory experiments. However, the rate of oxidation is much smaller than suggested by an early French experiment but consistent with what might be expected from oxidation in the soil. 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 HT oxidized and remaining in the soil and vegetation. This has important implications for dose calculations resulting from releases of HT during the seasons when soil bacteria are active.

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 slow release of HT sorbed on the metal surfaces of the air samplers.

Table 2 shows the location of the vegetation samples and the results of the analysis.

TABLE 2. Tritium activity in vegetation samples.

Location of Sample x,y (m)	Sampling Date Mo., Day	Sampling Time[1] (Hours)	Free HTO Bq/L	OBT Tritium Bq/L
50,10	6,10	-6.83	2.25E+01	3.91E+02
50,10	6,10	0.68	1.7[illegible]E+03	4.64E+02
50,10	6,11	17.67	3.00E+04	2.07E+03
50,10	6,11	23.77	1.83E+04	3.00E+03
50,10	6,19	186.17	2.52E+03	1.13E+03
180,0	6,10	-7.13	2.52E+01	
180,0	6,10	0.57	2.12E+02	
180,0	6,11	17.37	2.13E+03	
180,0	6,11	23.44	1.22E+03	
400,-15	6,10	-7.33	6.90E+01	
400,-15	6,10	0.50	9.70E+01	
400,-15	6,11	17.17	5.60E+02	
400,-15	6,11	23.33	1.70E+02	
50,0	6,10	-6.93	6.72E+01	6.77E+02
50,0	6,10	0.65	4.46E+03	6.58E+02
50,0	6,11	17.57	1.50E+04	9.85E+02
50,0	6,11	23.66	1.33E+04	1.02E+03
50,0	6,19	186.17	2.37E+03	8.43E+02
180, -40	6,10	-7.03	6.72E+01	
180, -40	6,10	0.61	1.47E+02	
180, -40	6,11	17.47	1.69E+03	
180, -40	6,11	23.55	9.69E+02	
350, 100	6,10	7.23	2.79E+01	
350, 100	6,10	0.54	5.47E+01	
350, 100	6,11	17.27	3.17E+02	
350, 100	6,11	23.22	1.69E+02	

[1] Measured from end of release 1550 (EDT), 10 June 1987.

results for the free water HTO samples are illustrated in Fig. 4. As expected, the concentration of HTO in the vegetation decreases with distance from the release point. This is a 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 is very low immediately following the release. This suggests that little HTO in air is picked up by plant leaves through exchange and that oxidation of HT in the vegetation is small. Five hours later there is a significant increase in the vegetation HTO concentration. The source of the HTO is the soil where oxidation had taken place during the release. The last samples taken at 50 meters 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 HT in the soil is depleted.

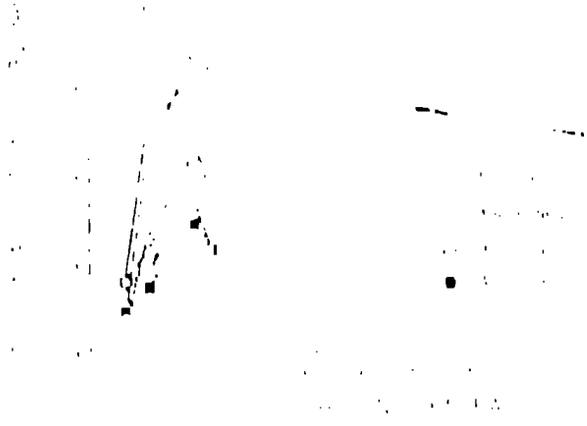


Fig. 4. Vegetation free HTO at three distances. At a given distance the solid lines represent the northernmost samples. (Refer to Table 2 for location of samples.)

Only at the 50 meter site were the concen-
ations of organically bound tritium high
ough to be detectable above background using
r techniques. The OBT concentration in the
ganic material is higher than that found in
e plant free-water. This may reflect the
story of tritium releases in the area. It is
so possible that this is another demonstra-
on of higher tritium of combustion than
ee-water tritium under natural environmental
nditions. The organically bound tritium
creased during the first two days after the
lease (Fig. 5). Organically bound tritium is
ill increasing after the vegetation HTO has
gun to decrease. Organically bound tritium
s a much slower turnover than HTO in the
getation free-water. It is likely that most
the OBT tritium is the result of photosyn-
etic fixation of tritium from the plant HTO.
erefore it is not surprising that by the
cond week, the concentration of organically
und tritium has decreased considerably. The
crease can be explained by the transport of
me of the labeled compounds to other parts of
e plant and the dilution of the labeled plant
terial with new growth which is not as highly
beled because of the decreasing HTO content
the vegetation.

ig. 5. Organic tritium content of vegetation
50 m.

NCLUSIONS

Apart from the discrepancy at 180 m
tween the LANL and SRL air samples, the
sults of all the air analyses are internally
nsistent. It is clear that [illegible]
re higher than would be predicted by [illegible]
ry tritium conversion experiments but much
wer than had apparently resulted during the
rly French experiment. It appears from the
resent experiment and other recent similar
elease experiments in Canada and France in
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 or 3 at the most.

Figure 6 shows the relationship of HT concentration in air measured by SRL during the release and HTO concentration in the vegetation five hours after the release. The slopes of the concentrations (on log-log paper) are nearly identical to those expected if soil HT oxidation is proportional to air HT concentration. This adds confidence to the rate of decrease in HT measured by the air monitors. Indirectly it also adds credence to the assertion that conversion in air of HT downwind of the release is occurring at a slow rate.

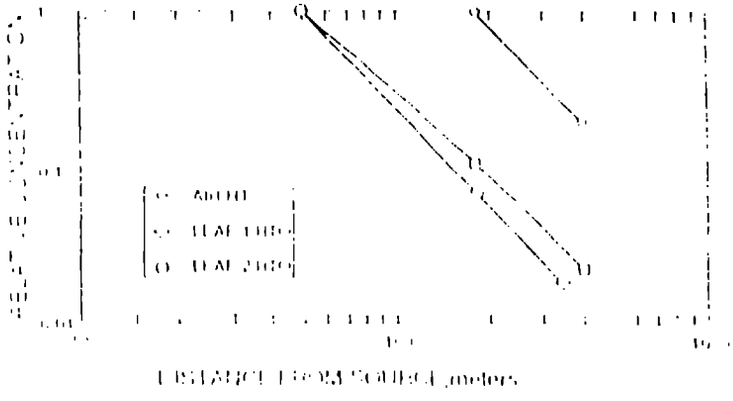


Fig. 6. Comparison of the decrease of HT with distance during the release and the decrease of peak plant free HTO.

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