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PRE-OPERATIONAL HTO/HT SURVEYS IN THE VICINITY OF THE CHALK RIVER LABORATORIES TRITIUM EXTRACTION PLANT

ÉTUDES, AVANT ENTRÉE EN SERVICE, DE CONCENTRATIONS DE HTO/HT DANS LE VOISINAGE DE L'INSTALLATION D'EXTRACTION DE TRITIUM DES LABORATOIRES DE CHALK RIVER

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RÉSUMÉ

On a effectué des études de concentrations de HT et HTO dans l'atmosphère, sous le vent installations nucléaires des Laboratoires de Chalk River, en novembre 1986 et mars, avril et septembre 1989 sous différentes conditions de température d'air, de direction de vent, de couverture végétale et d'enneigement. HT a représenté généralement 1-5 % de la quantité totale de tritium mais on a observé des valeurs allant jusqu'à 20 % probablement en raison de l'extraction préférentielle de HTO. Dans toutes les études, la plus forte persistance dans l'atmosphère de HT par rapport à HTO, a été évidente. Les quantités existantes de HT sont telles que les rejets chroniques de l'Installation d'extraction du tritium (IET) ne les fera pas augmenter de façon importante lorsqu'elle entrera en service. Ainsi, l'entrée en service de l'IET ne facilitera pas les études du comportment de HT dans l'environnement. Toutefois, des études à plus long terme de la répartition de HT provenant des installations existantes, en vaudraient la peine. On signale des quantités de HTO dans le sol et la végétation de l'aire d'études. D'autres études de la répartition du tritium entre l'air, le sol et la végétation dans les lieux soumis à une exposition chronique, seraient précieuses.

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ABSTRACT

Surveys of the concentrations of HT and HTO in the atmosphere downwind of the Chalk River Laboratories reactor facilities were carried out in 1986 November, and in 1989 March, April and September under different conditions of air temperature, wind direction, and snow or vegetative cover. HT usually amounted to 1-5% of total tritium, but values up to 20% were observed, probably resulting from preferential removal of HTO. In all of the surveys, the greater persistence in the atmosphere of HT than of HTO was evident. The existing levels of HT are such that they will not be augmented significantly by chronic releases from the Tritium Extraction Plant (TEP) when it comes into operation. Hence, operation of the TEP will not facilitate studies of the environmental behaviour of chronically released HT. However, longer term studies of the distribution of HT from the existing facilities would be worthwhile. Soil and vegetation HTO levels in the study area are reported. Further studies of the distribution of tritium between the air, soil and vegetation in areas subjected to chronic exposure would be valuable.

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1. INTRODUCTION

The Tritium Extraction Plant (TEP) at the Chalk River Laboratories (CRL) was built to reduce the tritium concentration in Atomic Energy of Canada Limited (AECL) heavy water, thus reducing occupational doses and emission of tritium to the environment. Since this facility will be handling high specific activity tritium in the elemental form, studies of the environmental behaviour of chronic and any acute releases of tritium may provide valuable information relevant to the environmental impact of fusion facilities. Pre-operational surveys in the vicinity have been carried out to obtain baseline data in view of the long-term and continuing release of tritium from nearby reactor facilities.

The maximum allowable operating gas inventory of the TEP is 1.2×10^{16} Bq (3×10^5 Ci) tritium, and the design limit for chronic release has been set at 5×10^{11} Bq (13 Ci) per week. The processing facilities are in double containment. Venting of all systems is to a short stack on the roof of the TEP, with no provision for tritium removal from the stack gases. Tritium, predominantly as HTO, is released from the NRX and NRU reactor buildings, from a D_2O upgrading plant and from the 50 m reactor stack sited on a height of land about 50 m above and 700 m distant from the reactor buildings. DT is generated in the reactors by radiolysis of the tritiated heavy water and, no doubt, some escapes. Determining the amount of HT and the variability of the atmospheric HT/HTO ratio are important objectives of the pre-operational surveys.

A survey of atmospheric HTO and HT, and plant and soil HTO concentrations, had been done in 1986 November, and results are included here with data from four surveys done in 1989 under different conditions of wind speed and direction, air temperature and snow or vegetative cover.

2. EXPERIMENTAL DETAILS

2.1 Sampling

Atmospheric sampling for HTO and HT was done using the technique of Östlund and Mason (1974). The principle of the method is to draw air containing 0.2% of added H_2 through Molecular Sieve (MS) 4A to remove all water vapour and associated HTO, and then through palladium-coated Molecular Sieve (Pd-MS) to combust the added H_2 and trap it and associated HT (Figure 1). Samplers constructed for the HT release experiments of 1986 and 1987 (Brown et al., 1988) were used. Air flow rates were measured with a rotameter at the beginning and end of each sampling period, and the mean was used to estimate total air sample volume. The volume of air sampled ranged from 0.1 to 1.8 m^3. Low volumes were due to poor battery performance in the surveys of March 29 and April 20, when several of the samplers were not operating when picked up. This introduces considerable uncertainty into the volume of air sampled, and hence the concentrations, but does not affect the HTO/HT ratios. Sampling intervals ranged between three and seven hours.

Subsequent to these surveys, tests indicated that slight oxidation of HT was occurring on the particular MS in some of the HTO traps. While this is of concern in experiments studying the oxidation of HT in the atmosphere, the extent of oxidation, less than 1% of the HT, is not great enough to affect significantly the HTO and HT concentrations observed in these surveys.

In each survey, eight samplers were deployed downwind of the reactor facilities, at distances of approximately 500, 1000, 1600 and 2000 m. The TEP is located at the northwest end of the Inner Area of Chalk River, 380 m from NRU, the major tritium-emitting building (Figure 2). With NRU at the centre, a 2000 m arc subtends McQuestion Point on the west, Perch Lake on the south and Point au Baptême on the east. Sampling locations were accessible by road, except for the western area between the stack and the river, where old roads and ski trails were used. Each sampler was started as it was placed in the field, so that the samplers did not run exactly concurrently. Seasonal conditions ranged from late winter with snow-cover to early autumn. As might be expected, winter conditions made deployment of the samplers considerably more difficult in the western area, where access was by snowmobile.

In 1989 September, passive samplers developed for occupational sampling were deployed, to test them in the field in association with the active samplers. The passive samplers consisted of 3 mL of tritium-free water in a plastic vial, having a defined orifice, providing controlled diffusion and exchange of atmospheric moisture. These samplers provide HTO concentrations only, integrated over the sampling period, which was 24 hours in this case.

2.2 Analysis

After sampling, the retained water was recovered from the MS traps by heating for two hours at 520°C on a vacuum line having liquid-N_2 cooled traps. To ensure complete recovery of the HTO, 1 to 2 mL of tritium-free water was flushed through each hot trap and combined with the original water recovered. Traps holding 40 or 100 g of MS and Pd-MS were used. When the activity level was high enough, it could be measured by dumping the MS and Pd-MS from a 40 g trap into 50 mL of distilled water, equilibrating for at least 48 hours and measuring a 2 mL aliquot of the water.

All tritium activity was measured by liquid scintillation counting. The major source of error in the final air concentration values was in estimating the total air volume sampled.

Free water tritium was recovered from soil and vegetation samples by azeotropic distillation with toluene.

2.3 Meteorological Data

Meteorological data were obtained from sensors at the 30 m level of a tower at Perch Lake (Figure 2), hence they indicate general conditions rather than ground-level conditions in the sampling areas. Mean air temperature, wind speed and direction were calculated for the sampling periods.

3. RESULTS

Table 1 shows the concentrations of atmospheric HTO and HT observed in all sampling periods, together with the mean meteorological conditions. The wind direction is the bearing, from north, or the origin of the wind. In contrast, the location of each sampling point is given by the angle from north to the line connecting the sampling point to NRU. To compare the two directions, 180° must therefore be added to the wind direction. Sigma Y, the standard deviation of the wind direction, indicates the atmospheric stability.

Air concentration data from the various surveys are presented in the figures identified below, showing:

a) the spatial distribution of HTO and HT concentrations, Figures 2, 4, 7, 10, 13, 16;

b) the decrease, with distance from NRU, of HTO and HT concentrations at stations approximately along the plume centre line, as suggested by the mean wind direction and the distribution of concentrations, Figures 3, 5, 8, 11, 14; and

c) meteorological data (three-minute mean values) for the day of the survey, with mean conditions indicated over the sampling period, Figures 6, 9, 12, 15.

3.1 Surveys of 1986 November 20, 1989 March 10 and September 19

The surveys conducted on these dates were over the same area to the northwest of the reactor buildings, with the wind from the ESE (Figures 2, 4, and 13). This area is lightly wooded with birch, poplar and the occasional pine and spruce. In 1986 November, the trees were bare and the ground was frozen but not yet snow-covered. The wind was stronger than usual, as a storm was just blowing in. Snow started to fall at the end of the sampling period. On 1989 March 10, the ground was completely snow-covered, and the weather was mild. On 1989 September 19, the trees were still in leaf and green. The higher concentrations of 1986 November likely resulted from a relatively high release rate, relatively stable atmospheric conditions, and a closer proximity of the sampling sites to the plume centre line.

On 1986 November 20 and 1989 March 10, the % HT peaked about 800 m from NRU (Figures 3 and 5), demonstrating the greater persistence in the atmosphere of HT compared to that of HTO. In 1989 September (Figure 14), the concentrations of HT and % HT were unusually high. Most of the high % HT is attributable to low HTO levels. Perhaps this reflects the removal of HTO by the forest in leaf.

Passive HTO samplers were put out at the time of the 1989 September 19 surveys, and left out for a 24-hour sampling. Wind direction remained reasonably similar to that during the short-term sampling, but the wind speed

and air temperature went through a diurnal cycle. HTO concentrations shown in Figure 16 were somewhat higher than the short-term values.

3.2 Survey of 1989 March 29

This survey was done over the area to the south of the plant buildings (Figure 7). The plume passed over a considerable stretch of open and built-up area before reaching forested areas, where snow cover still existed. The terrain is broken up by a ridge running NW-SE between the plant area and Perch Lake. Sample locations (Figure 7) did not adequately cover the central part of the plume along the plant highway. The major peak in HT concentration at 415 m (Figure 8) may be due to a local source (Chemical Engineering Building), since elevated levels have been observed in this vicinity in other samplings.

3.3 Survey of 1989 April 20

During this survey, a west wind created a plume across buildings and the open area at the edge of the river, and out across the water to Pointe au Baptême (Figure 10). The HT persisted across the water to a much greater extent than the HTO (Figure 11), as shown by the continuing increase in the % HT.

3.4 HTO in Snow, Soil Cores and Vegetation

In 1986 March, HTO concentrations were measured in snow cores and the free water of pine needles collected around the plant area (Figure 17). In association with the air sampling of 1986 November 20, soil cores and pine needles were taken. The soil cores were sectioned and their HTO profiles measured. Data are presented in Table 2 and Figure 18. There is very good correlation in trend between the concentrations observed in pine needle free water and the soil HTO inventory, to a depth of 45 cm. Concentrations in the pine needles were significantly higher than in the soil moisture, reflecting the pickup of HTO directly from the atmosphere. The surface levels are maintained higher by exchange with atmospheric HTO, whereas the lower levels reflect the diluting effect when precipitation infiltrates.

4. DISCUSSION

The surveys have established the levels of HTO and HT occurring in the atmosphere around the Chalk River facilities, and the variability of the HT/HTO ratio. The usual range of HT was 1-5% of the total tritium, but considerably higher values have been observed in a number of instances. At times of frozen bare or snow-covered terrain (1986 November 20 and 1989 March 10, respectively), HT was a few percent of the total tritium and the percentage peaked at about 800 m from the source. On the other hand, on 1989 April 20 and 1989 September 19, the percentage of HT continued to rise with distance. This may be attributable to the more rapid removal of HTO from the plume by the open water of the river on 1989 April 20, and by the leafed forest on 1989 September 19.

Our measurements show that release of HT from all facilities is about 2% of the 2 x 10^{13} Bq/month total tritium release to the atmosphere (i.e., about 4 x 10^{11} Bq/month). With a derived release limit of 2.2 x 10^{12} Bq/month for the TEP, the operational chronic release will probably be kept to less than 2 x 10^{10} Bq/month. Thus, operation of the TEP will not augment local environmental levels of HT significantly. However, the studies of the relative distribution of HT and HTO from the existing facilities have given useful information on the behaviour of chronically released HT in the environment. If discriminating passive samples were available, time-averaged behaviour over a broader area could be studied advantageously.

Further studies of the distribution of tritium in the air, soil and vegetation of the area subjected to chronic exposure would be valuable. These should be designed keeping in mind the requirement for parameters in the modelling of the behaviour of chronically released tritium.

5. ACKNOWLEDGEMENTS

The authors wish to acknowledge the able assistance with the sampling programs provided by P. Jay, J. Jirovec, T. O'Kane, and M. Wood, all of Chalk River Laboratories, and Z. Franic, IAEA fellow from the Institute for Medical Research and Occupational Health, Zagreb, Yugoslavia.

6. REFERENCES

Brown, R.M., G.L. Ogram and F.S. Spencer (1988). "Field Studies of HT Oxidation and Dispersion in the Environment II. The 1987 June Experiment at Chalk River". CFFTP-G-88007.

Östlund, H.G. and A.S. Mason (1974). "Atmospheric HT and HTO: I. Experimental Procedure and Tropospheric Data 1968-72". Tellus XXVI, 91.

Table 1. Tritium-in-air measurement at CRL before TEP operation

DATE	LOCATION from NRU (Degrees)	(m)	HTO (Bq.m-3)	HT (Bq.m-3)	% HT of total T	Meteorological Data (Mean Over Sampling Interval)
	319	400	499.00	2.41	0.48	
	290	400	748.00	9.00	1.19	Air Temp -7°C
1986	295	500	898.00	7.65	0.84	Wind Dir. 125 deg
NOV	290	800	43.40	2.42	5.28	Wind Spd 6 m.s-1
20	290	1060	27.20	0.94	3.34	
	278	1270	38.70	1.59	3.95	
	289	1600	42.60	0.76	1.75	
	280	475	155.80	3.50	2.20	
	270	800	79.80	1.90	2.33	Air Temp 2.9°C
1989	294	880	35.30	0.60	1.67	Wind Dir. 129 deg
MARCH	288	880	56.00	1.40	2.44	Sigma Y 20.7 deg
10	278	1270	58.50	1.30	2.17	Wind Spd 3.2 m.s-1
	274	1380	75.20	1.10	1.44	Snow depth 39 cm
	289	1580	31.10	0.50	1.58	
	276	1800	33.10	0.70	2.07	
	298	225	113.90	2.90	2.48	
	204	415	40.20	9.50	19.11	Air Temp 3.0°C
1989	157	550	5.50	0.20	3.51	Wind Dir. 18.3 deg
MARCH	260	745	11.20	0.30	2.61	Sigma Y 16.6 deg
29	203	1360	4.00	0.70	14.89	Wind Spd 3.3 m.s-1
	161	1500	0.00	0.50	100.00	Snow depth 10 cm
	248	1670	6.50	0.20	2.99	
	202	2250	0.20	0.20	50.00	
	324	180	18.30	1.10	5.67	
	154	325	132.20	1.50	1.12	Air Temp 8.9°C
1989	116	375	228.30	4.00	1.72	Wind Dir. 300 deg
APRIL	204	415	3.70	0.40	9.76	Sigma Y 26.2 deg
20	149	765	35.40	0.80	2.21	Wind Spd 3.2 m.s-1
	131	850	137.90	6.00	4.17	
	176	900	7.00	1.20	14.63	
	123	1800	9.10	1.10	10.78	
	319	350	180.00	7.80	4.15	
	306	410	162.00	3.70	2.23	Air Temp 21.1°C
1989	278	1270	----	4.80	----	Wind Dir. 131 deg
SEPT	274	1380	6.90	0.80	10.39	Sigma Y 18.4 deg
19	289	1600	7.80	1.40	15.22	Wind Spd 3.3 m.s-1
	302	1800	24.30	2.60	9.67	
	276	1800	5.70	1.40	19.72	

Table 2. Tritium (HTO) in vegetation and soil cores downwind of NRU on 1986 November 20.

	Soil Cores					Pine Needles
Location	Area (cm2)	Depth (cm)	+ M.C. (%)	HTO (Bq.L-1)	Inventory (Bq.cm-2)	# TFWT (Bq.L-1)
	4	0-5	31	9972	14.41	
319°	3	5-20	23	5696	23.87	23861
400 m	2	20-35	23	6287	47.21	
				Total	85.49	
	4	0-5	31	11595	13.19	
290°	3	5-20	18	5171	18.17	
400 m	3	20-45	15	4935	24.27	
				Total	55.63	
	4	0-5	33	12766	15.54	
295°	3	5-20	18	7146	23.07	16313
500 m	2	20-35	12	6439	25.45	
				Total	64.06	
	4	0-5	55	2510	6.48	
290°	3	5-20	23	1714	10.74	8543
800 m	3	20-45	21	1267	11.64	
				Total	28.86	
	4	0-5	20	3190	3.78	
290°	3	5-20	14	1414	4.17	5934
1060 m	3	20-45	11	913	5.70	
				Total	13.65	
	4	0-5	27	2328	3.26	
278°	3	5-20	15	1234	3.75	6956
1270 m	3	20-45	14	1032	5.90	
				Total	12.91	

+ Moisture Content
# Tissue-Free Water Tritium

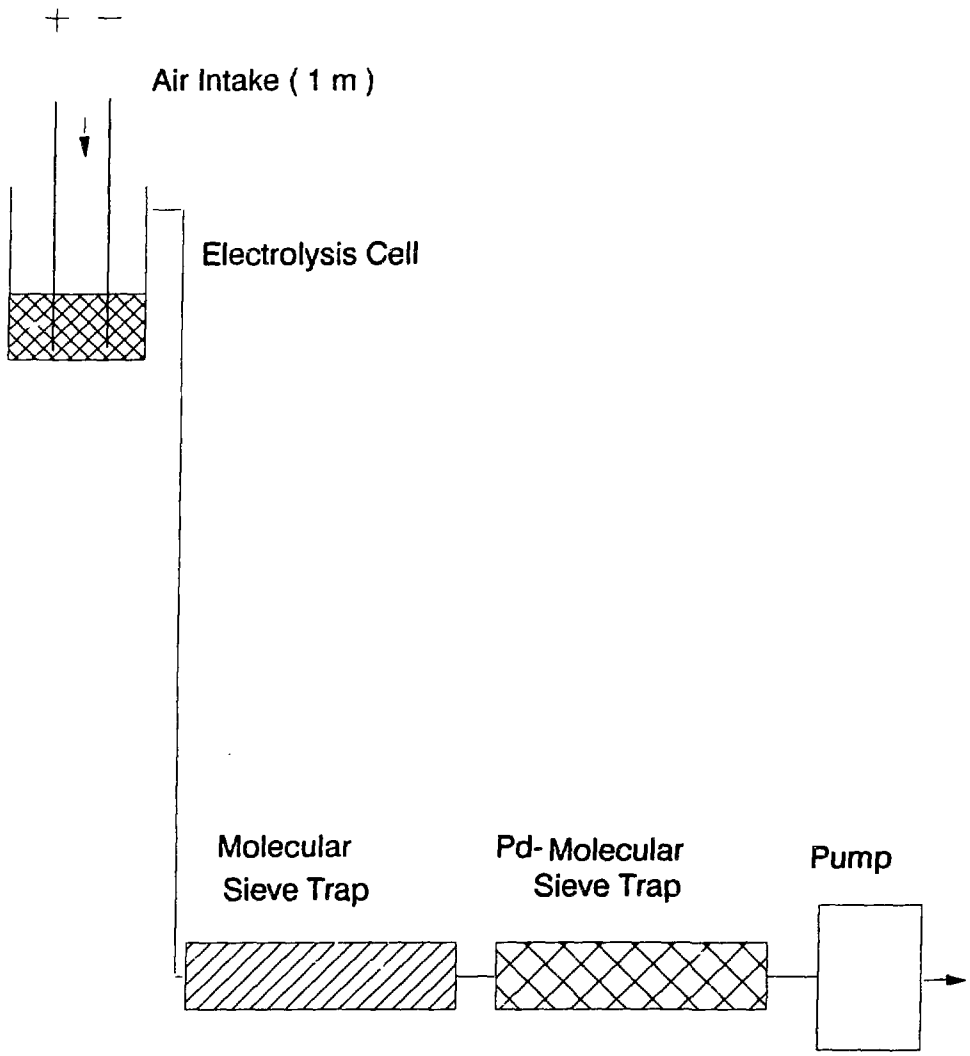

Figure 1. Sampler for atmospheric HT and HTO.

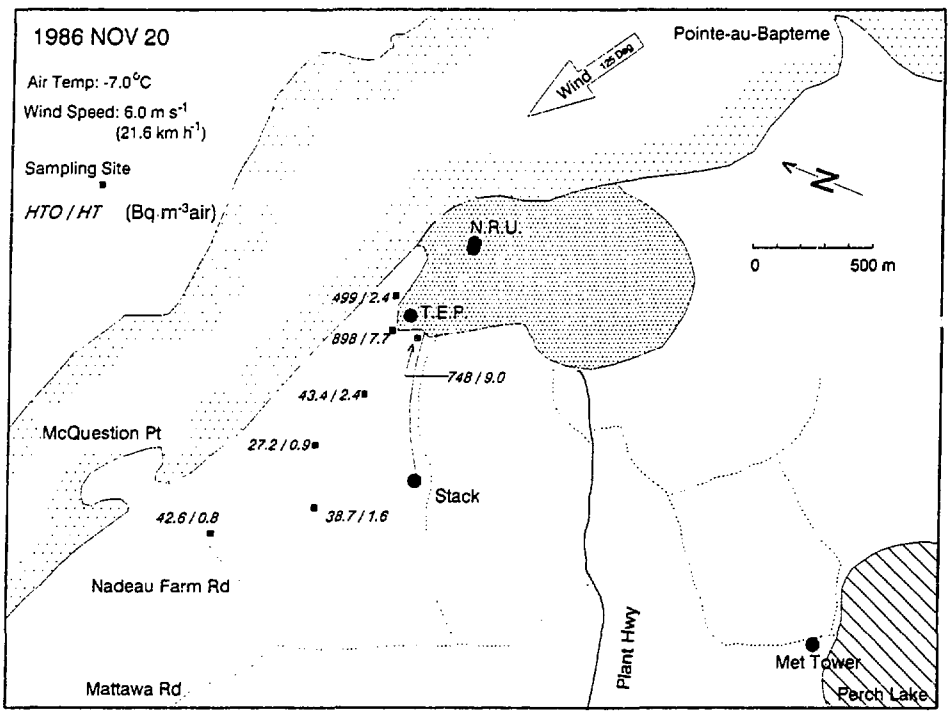

Figure 2. Atmospheric sampling for HTO and HT on 1986 November 20.

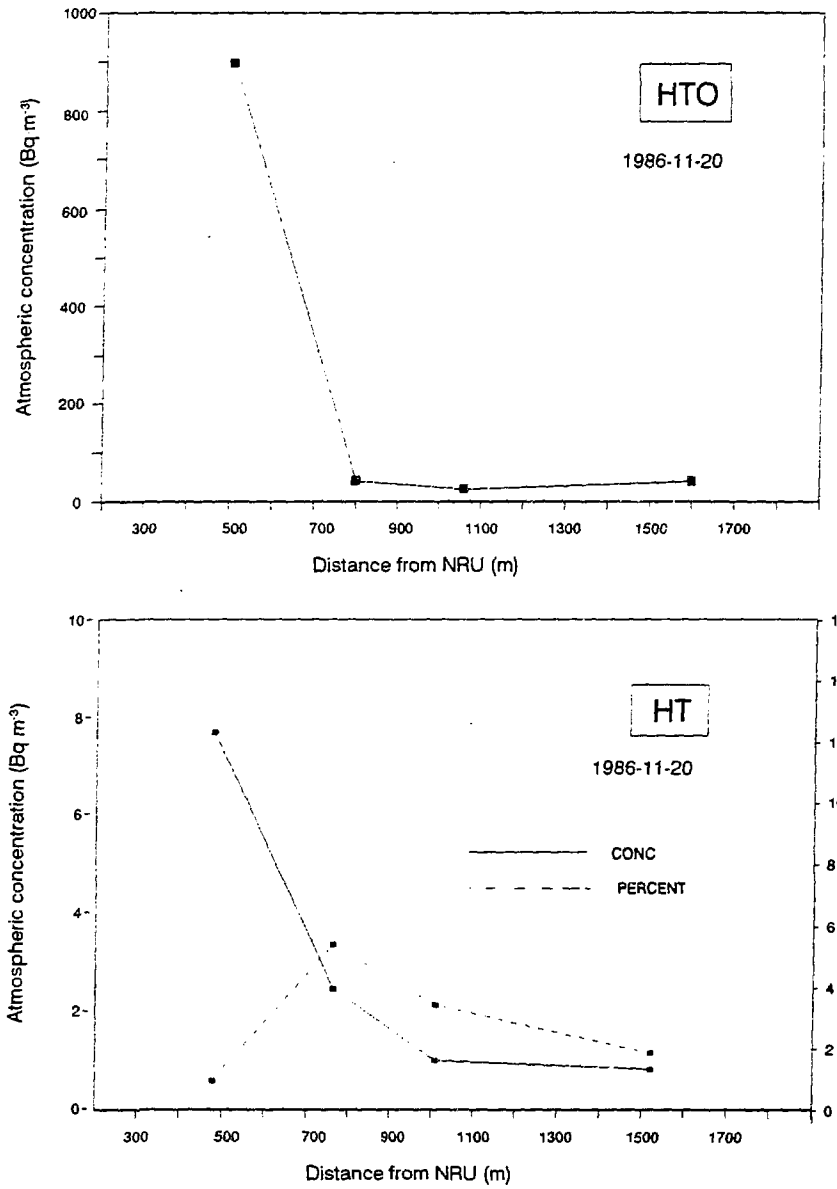

Figure 3. Tritium concentrations along plume centre line. 1986 November 20

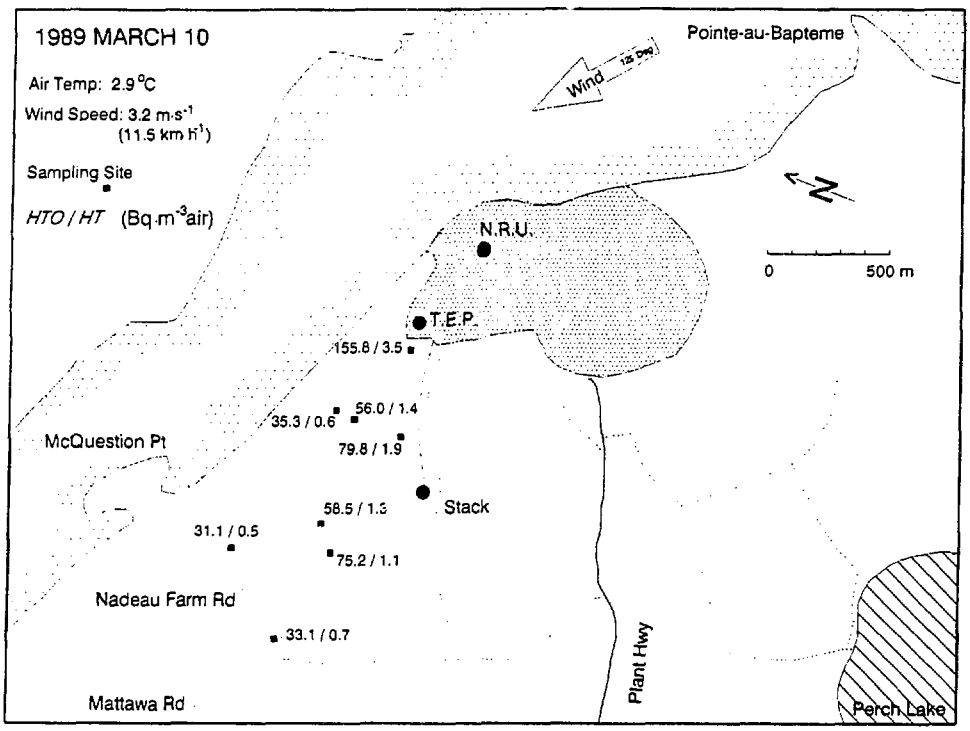

Figure 4. Atmospheric sampling for HTO and HT on 1989 March 10

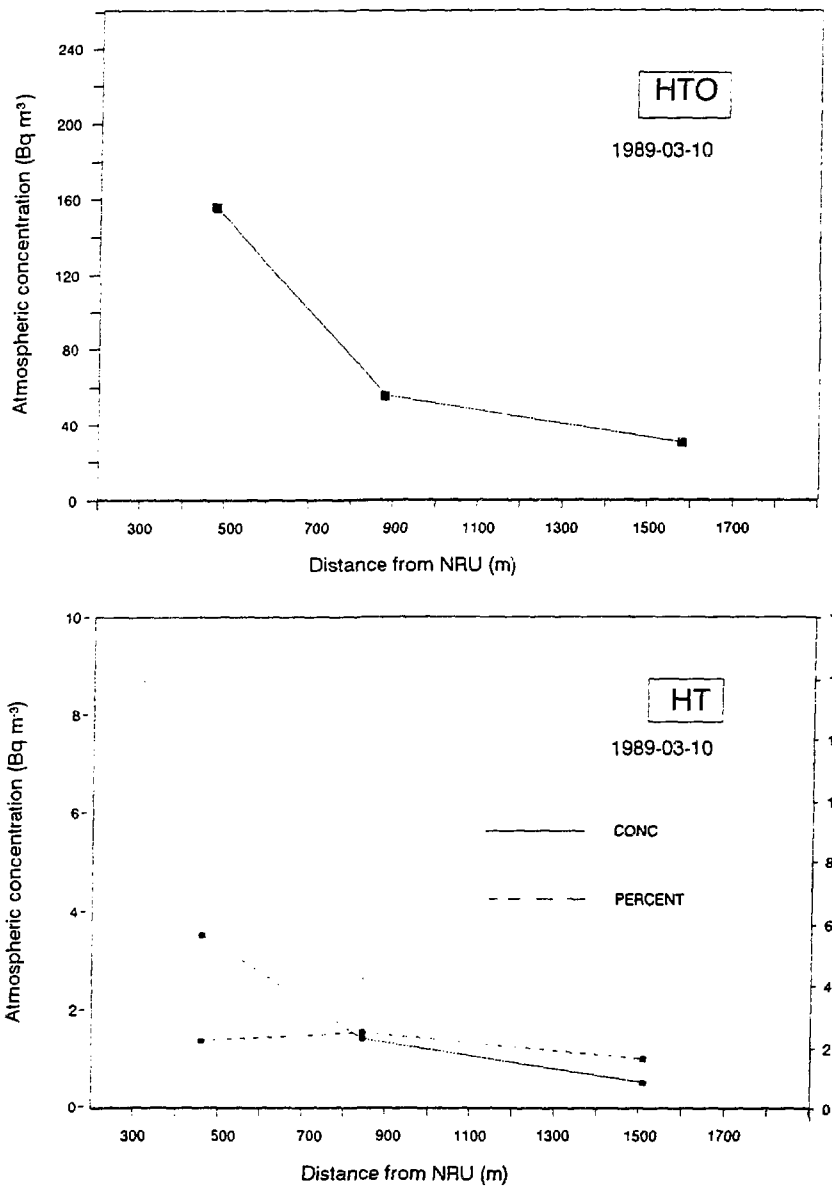

Figure 5. Tritium concentrations along plume centre line. 1989 March 10

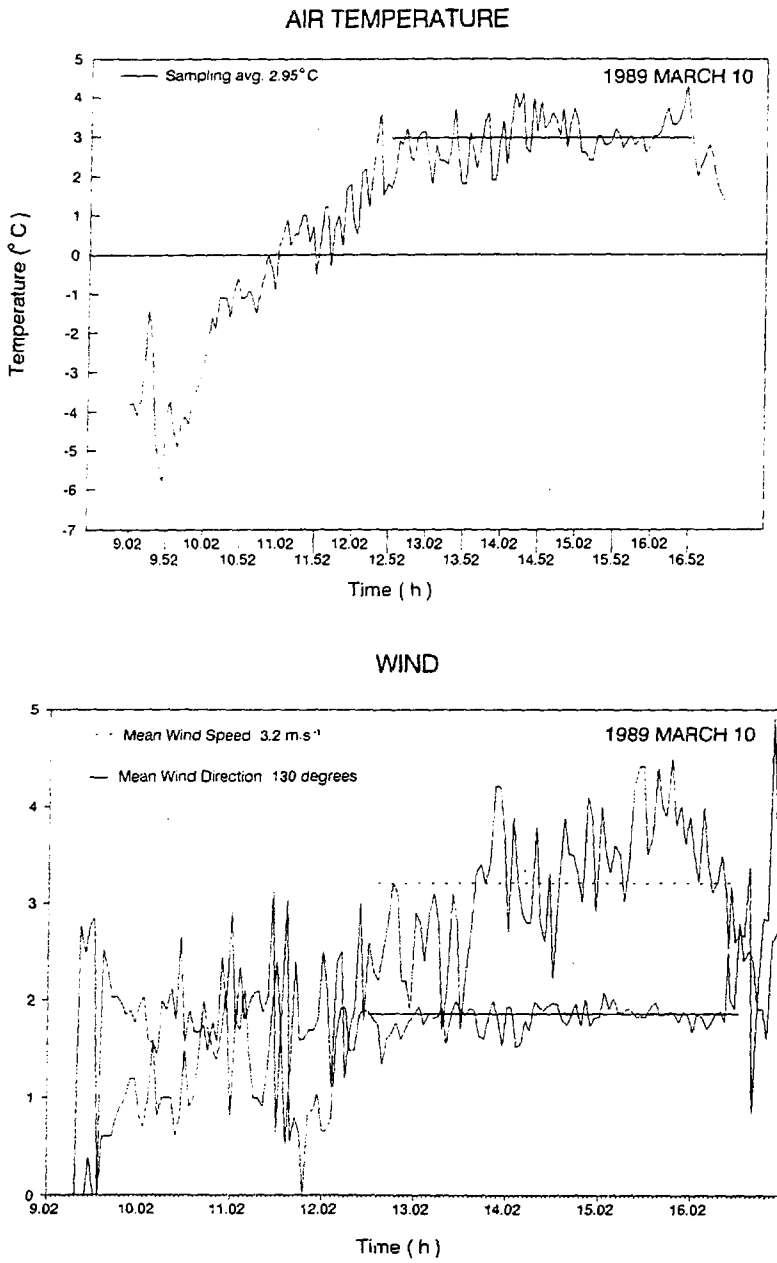

Figure 6. Meteorological data for 1989 March 10.

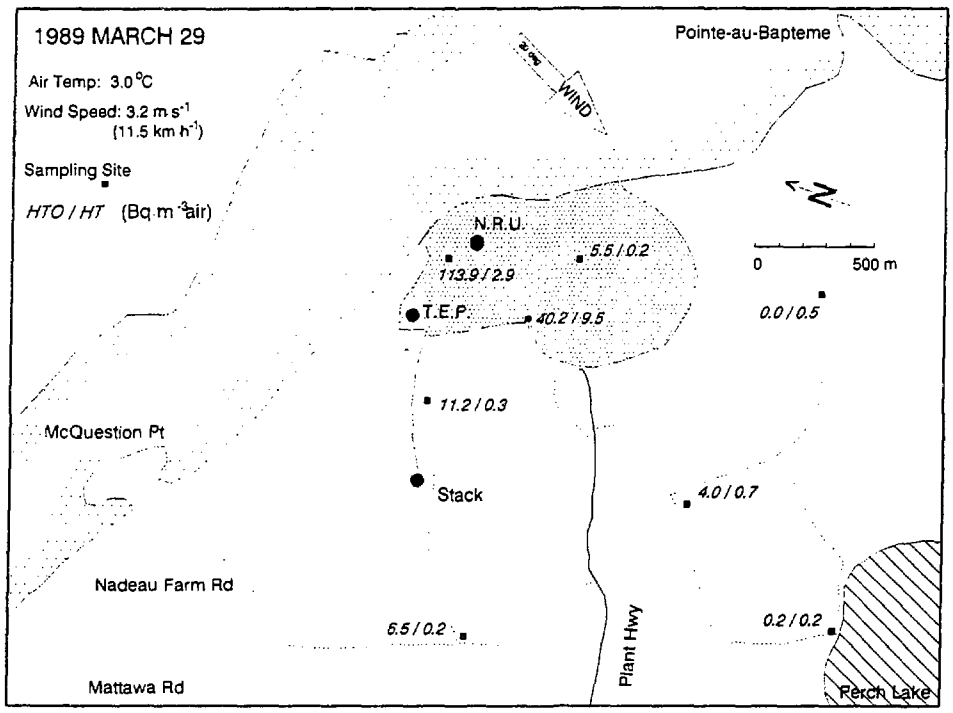

Figure 7. Atmospheric sampling for HTO and HT on 1989 March 29

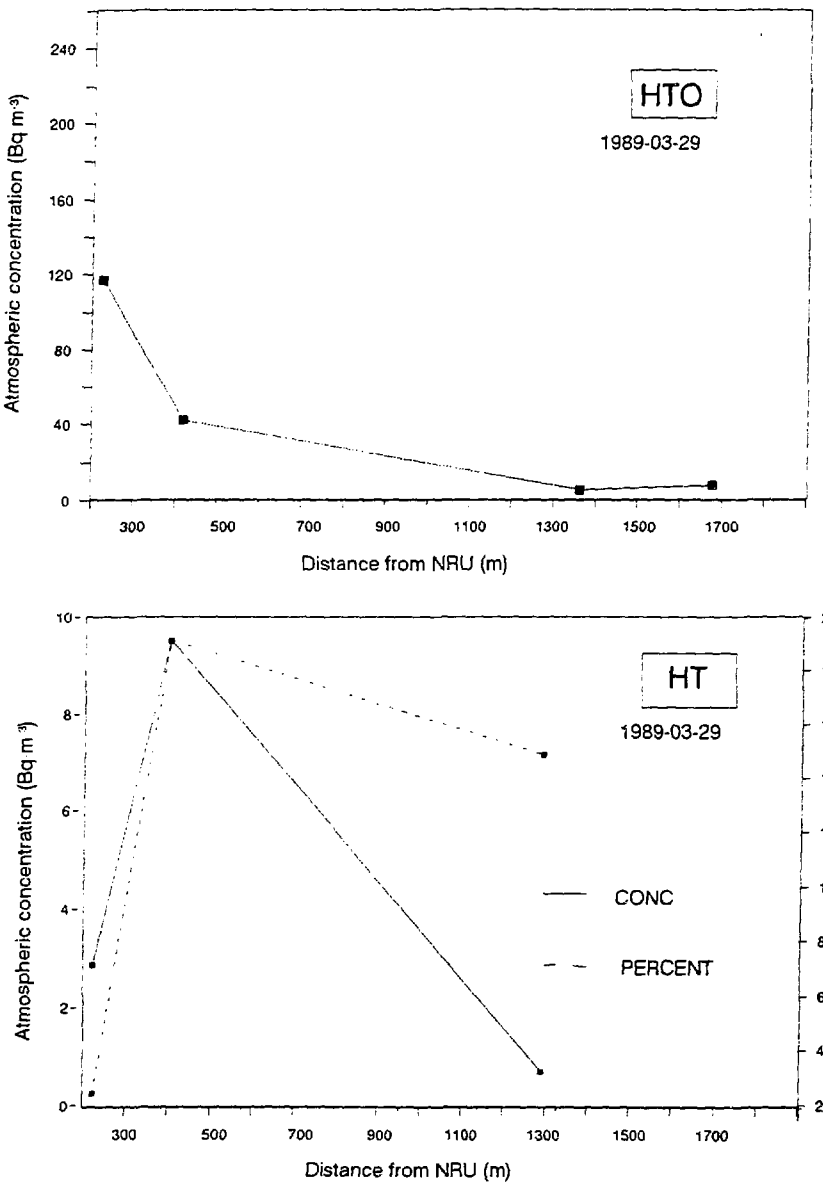

Figure 8. Tritium concentrations along plume centre line. 1989 MARCH 29.

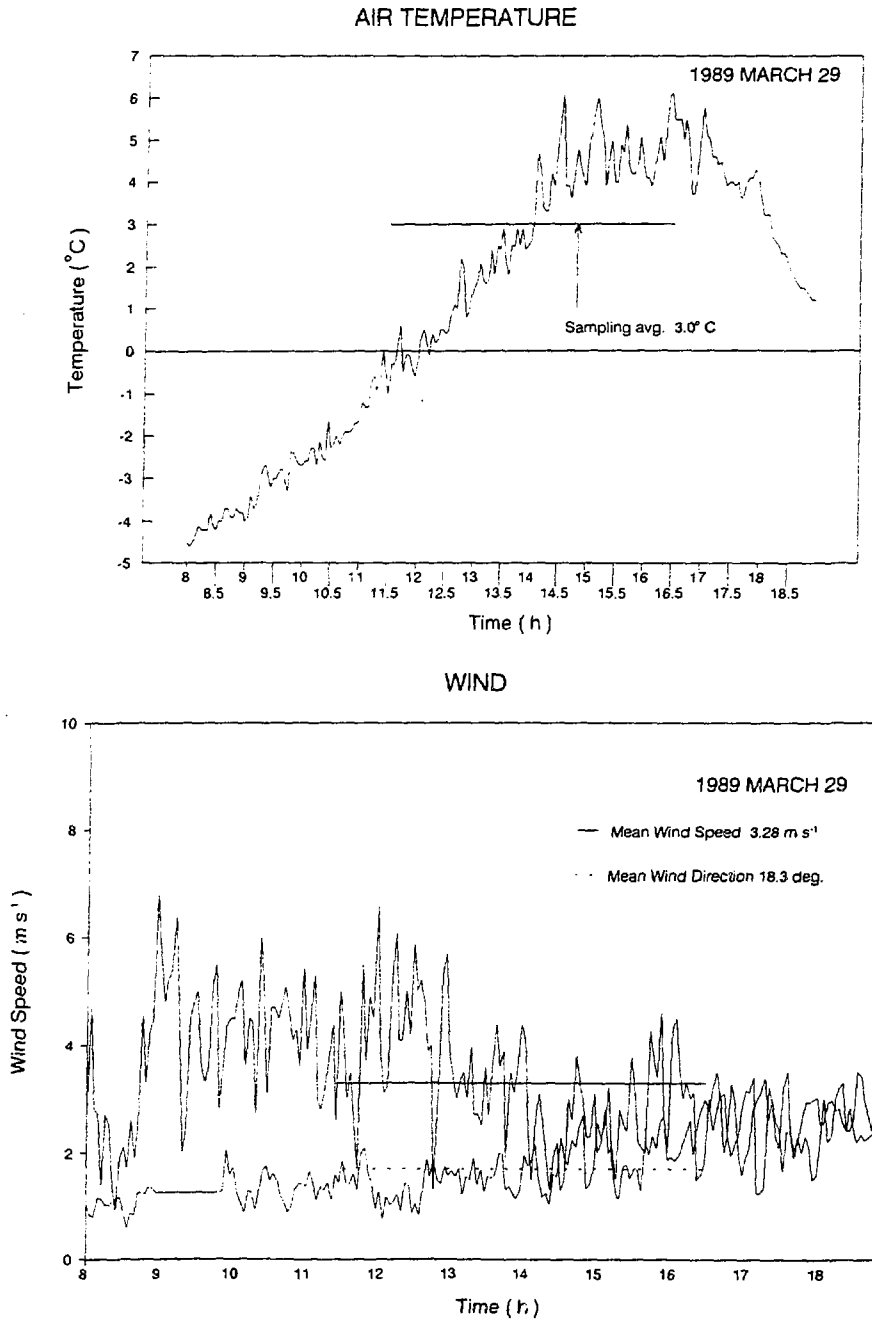

Figure 9. Meteorological data for 1989 March 29.

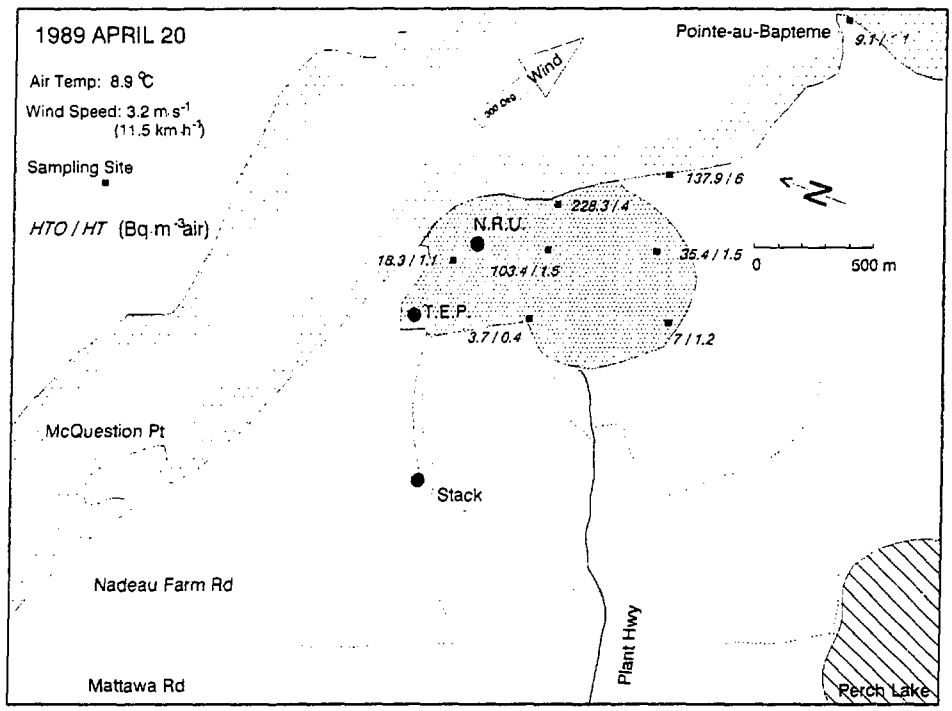

Figure 10. Atmospheric sampling for HTO and HT on 1989 April 20.

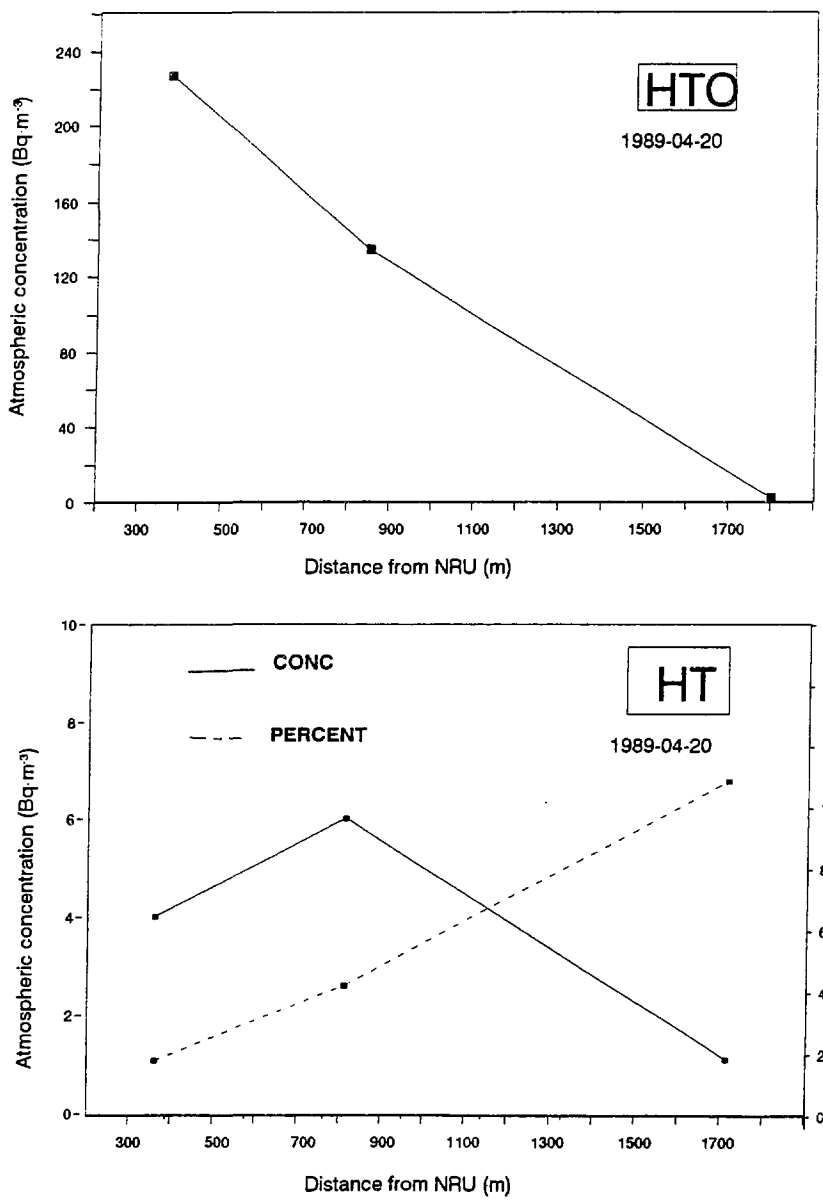

Figure 11. Tritium concentrations along plume centre line. 1989 April 20.

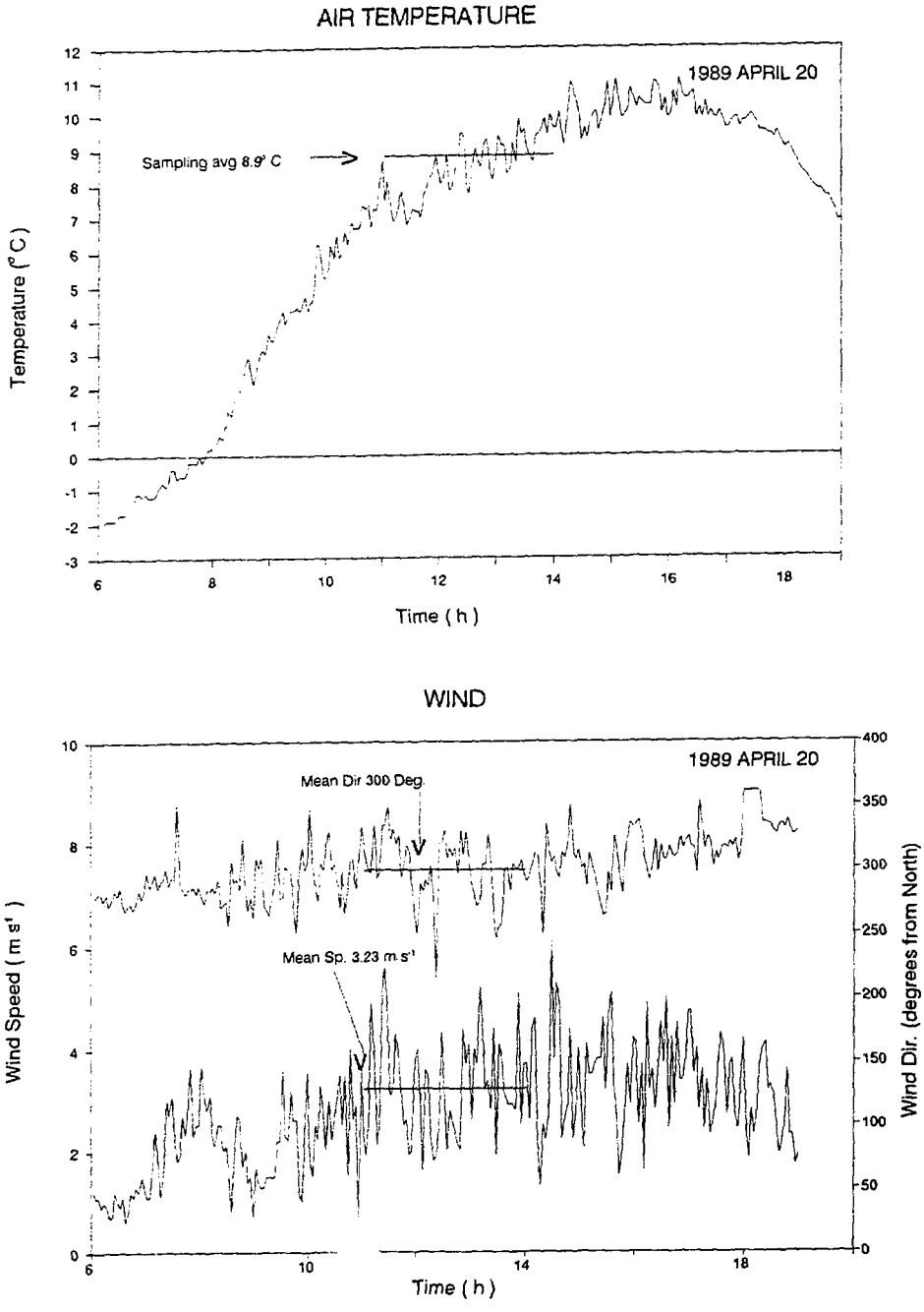

Figure 12. Meteorological data for 1989 April 20.

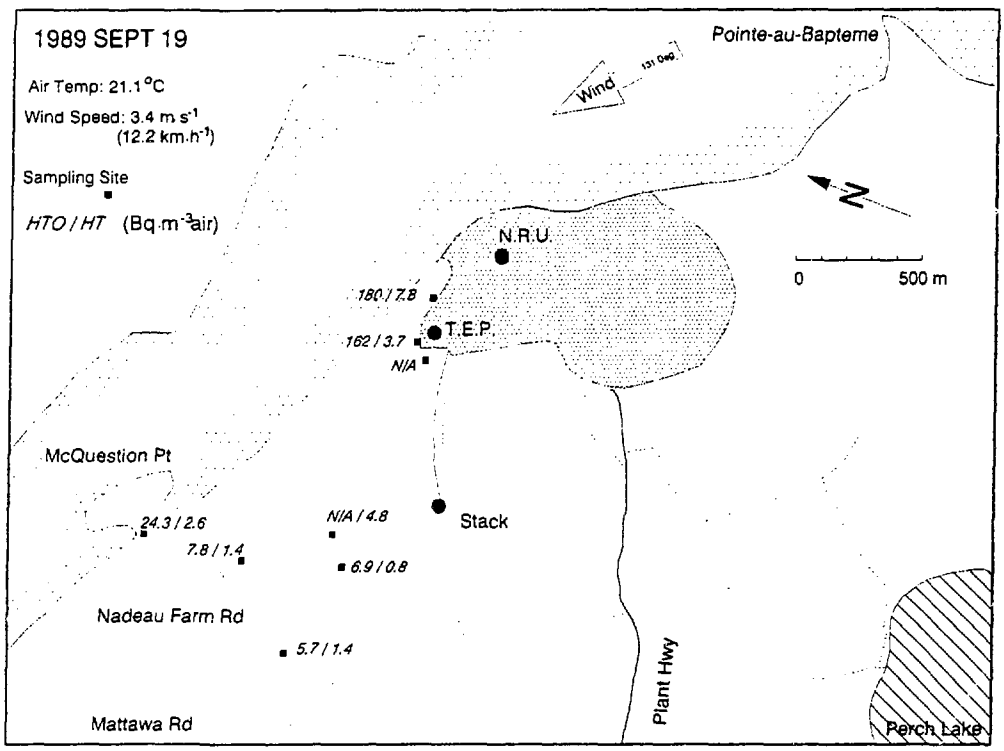

Figure 13. Atmospheric sampling for HTO and HT on 1989 September 19.

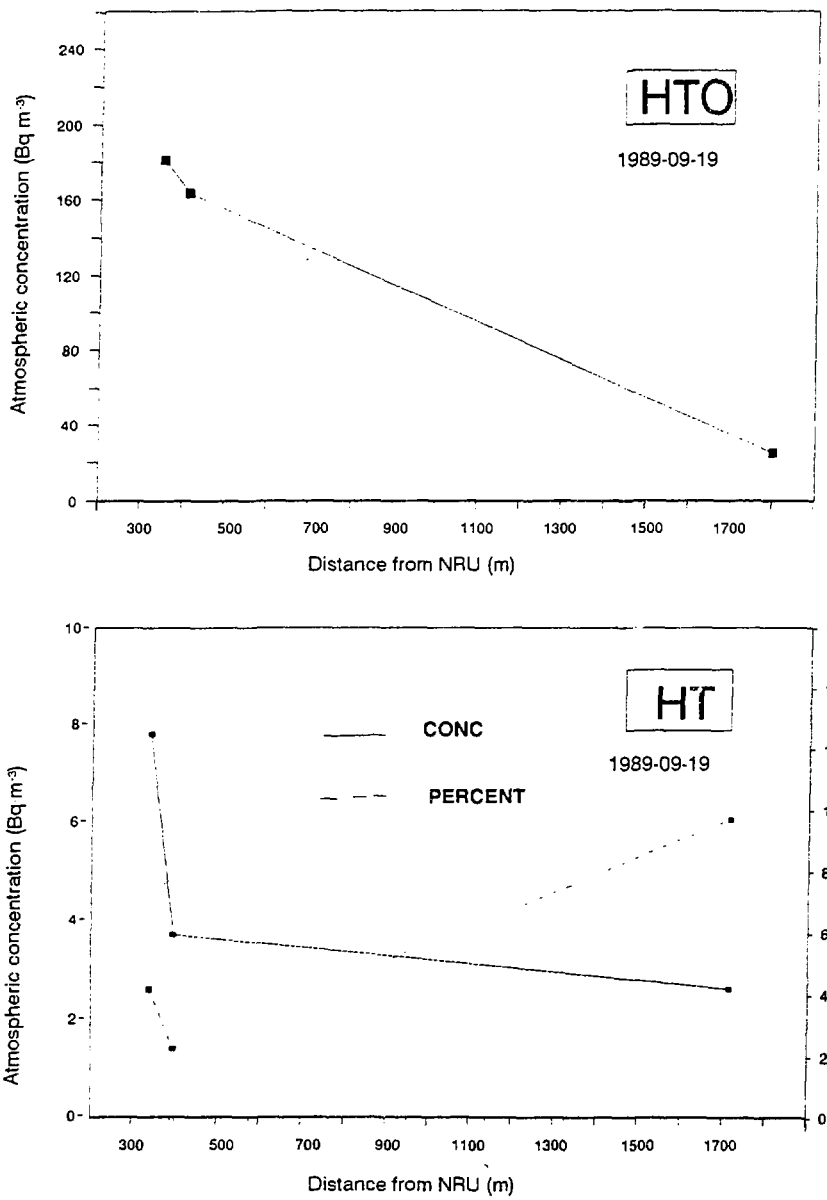

Figure 14. Tritium concentrations along plume centre line. 1989 September 19

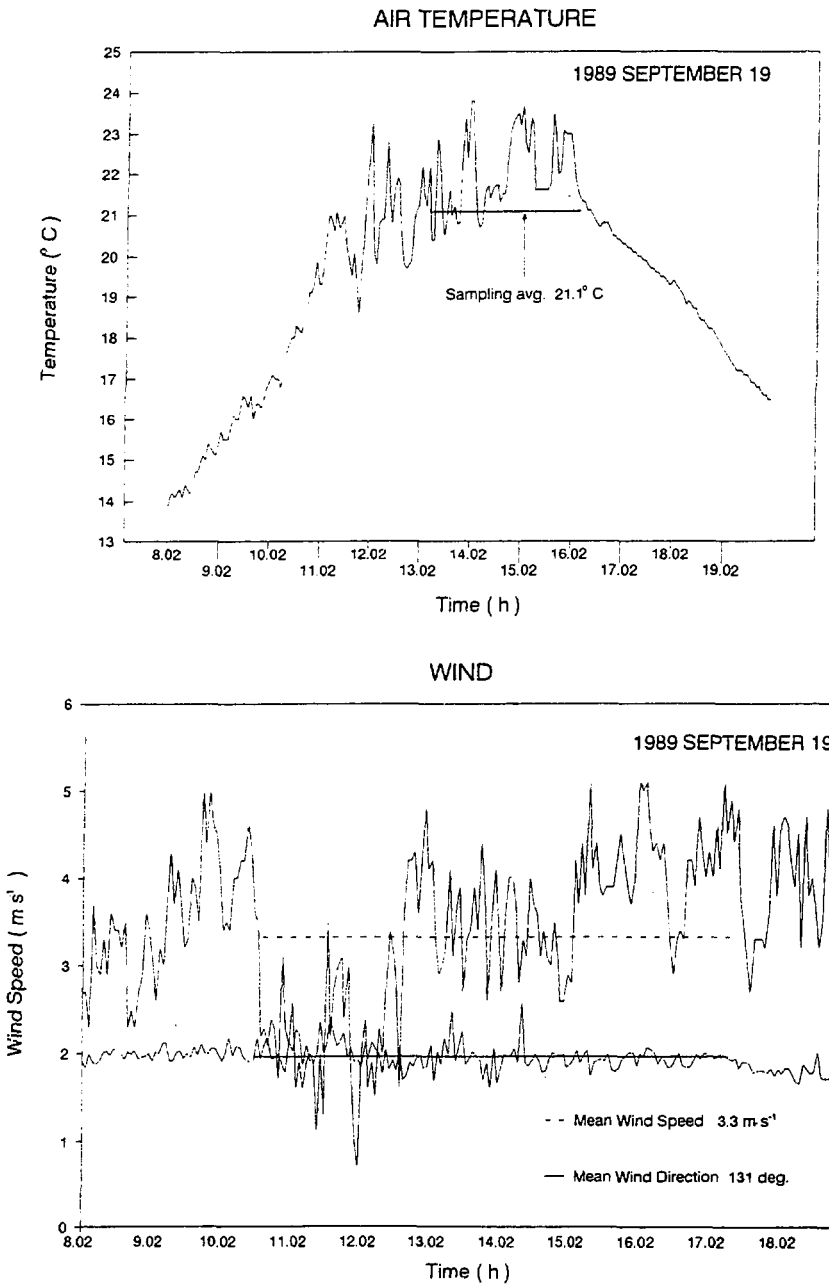

Figure 15. Meteorological data for 1989 September 19.

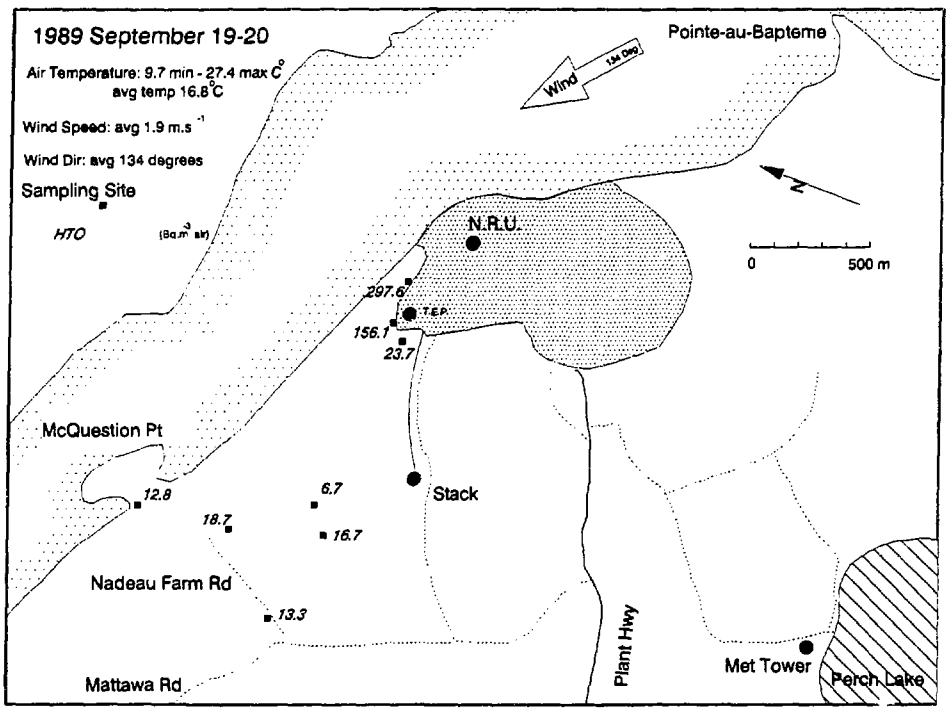

Figure 16. Sampling for HTO-in-air using passive samplers.
(over 24-h interval)

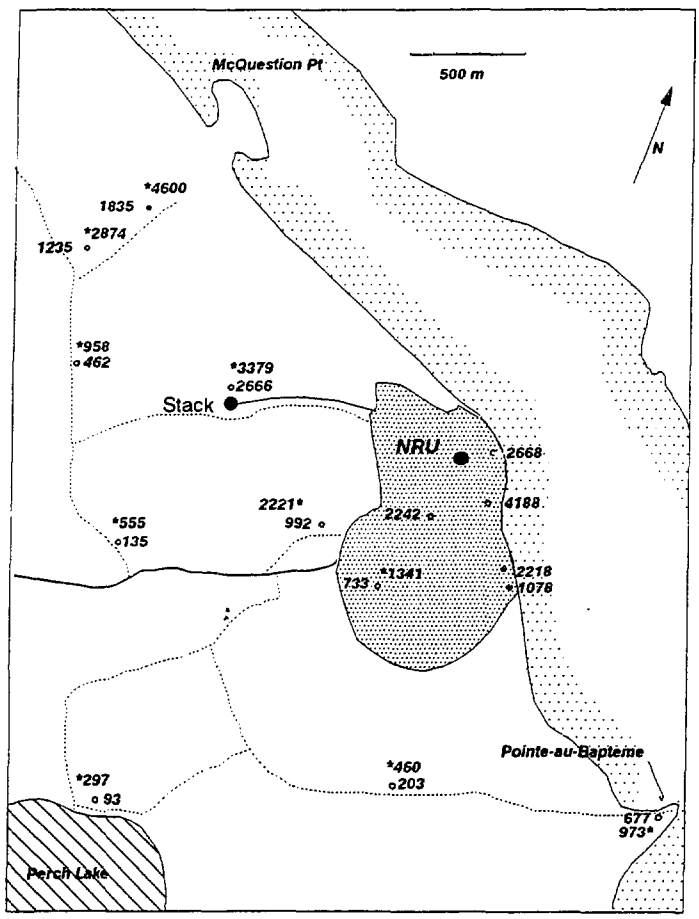

Figure 17. Tritium concentrations ($Bq.L^{-1}$) in snow cores [o] and tissue-free water of pine needles [*] in 1986 March.

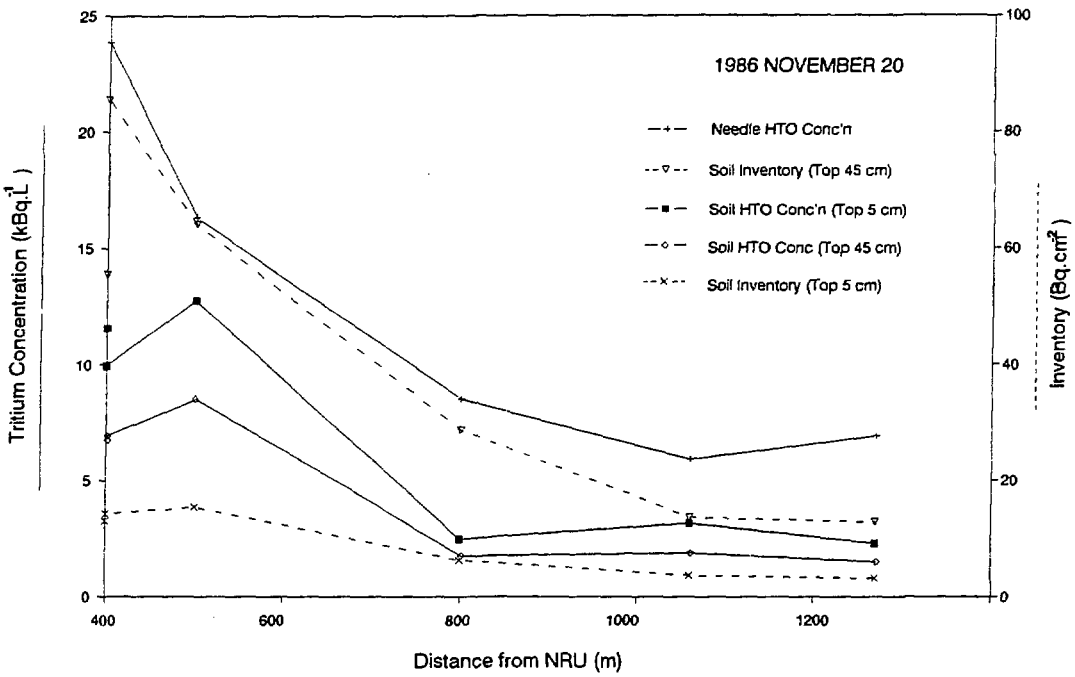

Figure 18. Variation with distance from NRU of HTO inventory and concentration in soil and pine needles.

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