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AECL-11421

Atmospherically Dispersed Radiocarbon at the Chalk River Laboratories

Carbone radioactif dispersé dans l'atmosphére aux Laboratoires de Chalk River

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January 1996 janvier

VOL 27 № 15

AECL

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RÉSUMÉ

Un faible pourcentage du carbone radioactif total produit par les réacteurs expérimentaux NRX et NRU aux Laboratoires de Chalk River s'est échappé de la cheminée principale et s'est dispersé dans l'atmosphère sur le site. Les contrôles effectués en 1982-1983 et 1993-1994 ont indiqué que les niveaux atmosphériques à plus de 50 m de la cheminée ne sont jamais supérieurs à 600 $Bq.kg^{-1}$ de carbone au-dessus du niveau du fond naturel de rayonnement et chutent aux niveaux atmosphériques quasi normaux, aux limites du site éloignées d'environ 7 km. Un facteur de dispersion > $1,2 \times 10^6$ $m^3.s^{-1}$ à une distance d'environ 0,75 km du point de rejet est calculé d'après un contrôle récent à l'intérieur de la cheminée. Une analyse des anneaux d'accroissement, ou cernes, des arbres du site a donné l'occasion de chercher les corrélations entre les données de ^{14}C et une production électrique en été et (ou) les pertes de modérateur.

Recherche sur l'environnement
Laboratoires de Chalk River
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ABSTRACT

A small percentage of the total radiocarbon produced by the NRX and NRU experimental reactors at the Chalk River Laboratories has been vented from the main reactor stack and atmospherically dispersed across the site. Surveys conducted in 1982-83 and 1993-94 have shown that atmospheric levels more than 50 m from the stack are never greater than 600 $Bq.kg^{-1}$ carbon above the natural background level, falling to near-global atmospheric levels at the site boundaries roughly 7 km away. A dispersion factor > 1.2×10^6 $m^3.s^{-1}$ at ~ 0.75 km distance from the point of emission is calculated on the basis of recent in-stack monitoring. Analysis of growth rings in on-site trees has provided an opportunity to search for correlations of ^{14}C output with summer power production and/or moderator losses.

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1. INTRODUCTION

1.1 Background

Because of its long half-life of 5730 years, and its rapid exchange with stable carbon in the atmosphere, natural radiocarbon is an enormously valuable tool for a wide range of disciplines, from archeology and anthropology to oceanography and hydrology. Its applications grew rapidly following the first identification of its potential as a dating device by Libby et al. in 1949, increasing even more sharply following the conclusive demonstration by Cain and Suess (1976) that the $^{14}C/^{12}C$ of vegetation is in equilibrium with that of the atmosphere in which it is grown.

The primary source of ^{14}C is cosmogenic production in the stratosphere and troposphere, largely as a result of the interaction of high-energy particles with nitrogen in the atmosphere. Fluxes and energy spectra of cosmic particles, and hence production rates for ^{14}C, are influenced by a number of external forces, such as the earth's geomagnetic field, solar disturbances, etc.; these variances in production have been studied in detail by the radiocarbon dating community, whose results depend on a precise value for the $^{14}C:^{12}C$ ratio at the time of growth of the object under study. More substantial perturbations in atmospheric levels have been caused by the burning of fossil fuels since the start of the Industrial Revolution, releasing to the atmosphere large quantities of CO_2 containing no radiocarbon, and resulting in a dampening of the natural cosmogenic signal. This is known as the Suess effect (1955).

Natural production of radiocarbon has been augmented substantially in the 20th century by anthropogenic contributions. The above-ground testing of nuclear weapons, which commenced in the early 1950s and continued intermittently until 1980, was sufficient to almost double the concentration of ^{14}C in the atmosphere during the peak year (1963). Fallout of the radiocarbon injected into the stratosphere during those years is still measurable, but has steadily declined with a half-life of approximately 12 years.

A smaller contribution to global ^{14}C results from activation reactions occurring during nuclear power-plant operations. The three principal reactions are as follows;

$^{14}N(n,p)^{14}C$ (*1*) $\qquad$ $^{17}O(n,\alpha)^{14}C$ (*2*) $\qquad$ $^{13}C(n,\gamma)^{14}C$ (*3*)

For a heavy-water reactor (HWR), calculations by Walton (1994) and others have shown reaction (1) to be most important in fuel, cladding and cover gas, reaction (2) in moderator and/or coolant, and reaction (3) in the annular gas, when CO_2 is used for that purpose. In all cases, 99% of the ^{14}C produced in the moderator and primary heat-transport system is removed on ion-exchange resins. However, $^{14}CO_2$ produced in the annular gas and in the cover gas, or released to that blanket from the moderator, will be vented via the reactor stack. The HWR's that have operated at the Chalk River Laboratories since the 1950s are experimental reactors, and differ very considerably in design from the CANDU HWR. These reactors do not contain an "annular gas", per se, but there are occasional opportunities during

operation for air to enter void spaces in the J rod annuli. These spaces will be vented during routine purging of the cover gas. In addition, some portion of moderator evaporative losses are released via the building roof vents, providing another potential route for $^{14}CO_2$ emissions.

The net specific activity of ^{14}C in the atmosphere in 1950 has been determined to be 13.56 dpm/g (226 $Bq.kg^{-1}$) carbon. This value has been defined by the radiocarbon community to be "modern carbon", and all subsequent measurements have been compared to it, as percent Modern Carbon (pMC). By 1963 atmospheric levels reached ≈200 pMC (~450 $Bq.kg^{-1}$); however, the combined effect of atmospheric washout and increased fossil-fuel burning has reduced present-day levels to approximately 110 pMC (~250 $Bq.kg^{-1}$).

1.2 Purpose and Scope of this Study

Although in absolute terms the contribution to global ^{14}C from nuclear reactors is small, radiocarbon has the potential to be a significant contributor to radiation dose in local situations, and it is largely for this reason that further information on production and emission rates, dispersion and mixing in the atmosphere in the immediate vicinity of the station, uptake in vegetation, etc., are required for site monitoring. Since the primary route for exposure to ^{14}C in both human and animal populations is via ingestion, local and regional dose predictions must be based on accurate measurements of vegetation.

In 1982-1983, a systematic survey of the levels of ^{14}C in the atmosphere and vegetation on-site was performed. This survey was repeated and enlarged in 1993-1994, and the results of both studies are reported here.

Unlike ^{14}C produced by cosmic rays and nuclear-weapons testing, emissions from nuclear power stations are not necessarily uniformly distributed in the vicinity of the reactor, and assumptions about the validity of the record of emissions preserved in vegetation must be verified. Consequently, in addition to reporting here on the collection and analysis of atmospheric and vegetative samples, we also present comparisons of short-term (two weeks or less) and whole-season data, comparison of different species of vegetation, and comparison of day versus night air sampling.

A dispersion factor has been calculated from correlation of data from two week-long sessions of in-stack monitoring (Rao and Young, in preparation), separated by a year, with environmental monitoring during the same periods at 0.75 km from the stack in the principal wind direction.

Data from this study have also been used to test the suitability of tree rings as passive biomonitors of long-term reactor operations (King, Repta and Enright, in preparation.).

2. EXPERIMENTAL

2.1 Field Methods

2.1.1 Site

The Chalk River Laboratories (CRL) occupy a site of approximately 100 km^2, 200 km northwest of Ottawa, on the shore of the Ottawa River (Figure 1). Aside from the relatively small area occupied by the laboratory buildings, most of the area is covered in mixed vegetation, consisting of pine and spruce, maple, birch, poplar and alder, with occasional open patches of grasses, and some wetland areas which provide a suitable habitat for ferns, cedar, etc.

2.1.2 Sample Collection

Sampling sites for both surveys were laid out on a rough grid, wherever possible choosing locations readily accessible to the roads that traverse the property (Figures 2 and 3). The availability of suitable vegetation also influenced the choice of sites.

Atmospheric CO_2 collection

Passive samplers, consisting of trays of NaOH sheltered from animal intrusion and precipitation, were deployed at each of the chosen sites for two-week periods. Details of the apparatus are given in Milton and Brown (1993). Ten sites were selected for the 1982 survey (Figure 2), with the solutions being collected every two weeks into plastic storage bottles and replaced with fresh solution, over a ten-week period. The grid was altered slightly and enlarged in 1993 (Figure 3) to 31 sites for the two-week period of June 1-15; this number was subsequently reduced to two, which were sampled at two-week intervals for the remainder of the season. These two locations were within 1 km of the reactor stack, in the principal wind directions. Air sampling in 1994 was also concentrated at these two sites, but included a few of the earlier, more distant sites, to check for any substantial changes between the two years.

Active samplers, consisting of two 300 mL glass bubblers in series, connected to a Brailsford pump and 12-volt battery, were also deployed at these sites over a one-week period in 1994 May, during which time in-stack monitoring was also taking place. A sampling time of 24 hours, at flow rates of ≈0.5 L/min, was required to collect sufficient CO_2 for analysis.

In order to evaluate the importance of changes in the ^{14}C specific activity of air between daylight (growing) hours and those at night, samplings were performed at these two sites to measure the two periods separately. Two samplers fitted with timed closing devices were borrowed from Otlet and Walker (1992, private communication) in 1993 August for this purpose; during 1994 samplers of a slightly different design, fabricated at CRL, were deployed at these same sites (Figure 4).

Vegetation Collection

Whenever possible during the 1982 survey, poplar leaves were collected at the sites chosen, usually at the same time that air samplers were emptied and refilled with fresh solution. In early September of that year, wild grass samples were collected from 36 sites on laboratory property and in the surrounding area, to serve as representative of the entire growing season.

Similarly, leaf samples were picked at 15 sites at the end of the two-week intensive survey in 1993 June; ten of these were poplar and five were maple. Following that period, vegetation was collected over the whole season at the two all-season air sampling sites. In addition, samples were collected from a number of tree species at one site, both coniferous and deciduous.

Care has been taken throughout this study to avoid sample contamination. Plastic disposable gloves were worn during leaf collection, with samples being placed directly into new plastic bags following picking. On their return to the laboratory, all samples were rinsed in double distilled water, dried in an oven at approximately 80°C, crushed and stored in plastic bottles (Milton and Brown, 1993).

Meteorological Data

A wind rose showing annual average wind direction for each of the survey years was provided by the meteorological group, Environmental Research Branch, as well as information on wind speed and direction during the times of the two sets of stack-emission measurements used for calculation of dilution factors. Temperature and precipitation data were obtained from the data collection service within the Branch.

2.2 Analytical Methods

The chief analytical techniques utilized in this study have been described by Milton and Brown (1993). Details of minor changes in these procedures have been reported elsewhere (Kramer, Milton and Repta, in press, 1995).

The accuracy of our results has been checked by measurement of NBS oxalic acid and secondary reference materials obtained from the International Atomic Energy Agency (IAEA), and the level of precision of individual analysts assessed by repeated measurement of one well-homogenized vegetation sample.

Regional ^{14}C backgrounds were measured during both surveys. Samples were obtained from Eganville, 84 km southeast of CRL, for this purpose in 1982; a number of sites well beyond the CRL site were surveyed in 1994, and the results in both cases were compared to the published data for Gatineau Park, Québec (Lowden and Dyck, 1977; McNeely, 1994).

In a few cases samples have been analyzed at the University of Waterloo Isotope Laboratory for ^{13}C, to estimate the degree of isotopic fractionation occurring during air-sample collection and photosynthetic processes.

3. RESULTS AND DISCUSSION

3.1 Regional Background

In 1982 the specific activity of ^{14}C at Eganville, Ontario was determined to be 269±9 $Bq.kg^{-1}$ C.

A more detailed survey in 1994 of levels in southeastern Ontario, well removed from any nuclear facilities, gave an average value of 252±8 $Bq.kg^{-1}$ C.

In order to compare these values with the data of McNeely, Geological Survey of Canada (GSC) (1994), it is necessary to convert them to pMC, as in Table 1 (for details of the calculation, see Stuiver and Polach (1977)).

Table 1. Measurements of Background Levels of ^{14}C in Ontario Vegetation

Laboratory	1983 Average pMC	1993 Average pMC
GSC	128.4	114.6
CRL*	119.8	112.6

* Average ^{13}C values quoted by McNeely were used for CRL calculations, since no local data were available.

The regional background value (one measurement) used for 1983 appears to be somewhat low; however, the difference between the two laboratories in 1993 was less than 2%, indicating a lack of systematic bias in measurement.

3.2 Analytical Precision

On the basis of the data listed in Table 2(a), ^{14}C measurements made at CRL are ~5% low on average. The most likely cause of such a bias is an error in the value ascribed to our laboratory working standard.

The data listed in Table 2(b) indicate good reproducibility for high-specific-activity samples. Although the precision is lower for low-activity samples, it is still acceptable.

Table 2a. CRL Results for IAEA Quality Assurance Materials and NBS Oxalic Acid

Sample	Date Analyzed (No. of replicates)	Our Value pMC	Consensus Value pMC	% Deviation from Consensus Value
NBS Oxalic II	1982 (2)	130.0	135.7	-4.2
NBS Oxalic II	1992 (2)	131.9	135.7	-2.8
IAEA Sucrose	1992 (3)	135.7	150.6	-9.9
IAEA Cellulose	1992 (3)	123.7	129.4	-4.4
IAEA Cellulose	1993 (8)	122.7	129.4	-5.2

Table 2b. Precision of Replicate Measurements Obtained by Three CRL Analysts

Analyst	No. of Samples Analyzed	High Specific-Activity Sample 1 std. dev. of mean	Low Specific-Activity Sample 1 std. dev. of mean
1	6		4.9%
	2	2.1%	
2	16	1.8%	
	14		6.9%
3	7		6.3%

3.3 Measured ^{14}C Concentrations in Vegetation and Air

The concentrations of ^{14}C observed in vegetation in 1982-83 are listed in Tables 3a and 3b. Those observed in 1993-94 are listed in Tables 4a and 4b.

Table 3a. Carbon-14 Concentrations in Vegetation, 1982

Site No. Fig. 2	Location	Poplar Leaves		Grass	
		Collection Date	(Bq.kg^{-1} C)	Collection Date	(Bq.kg^{-1} C)
60	Eganville, 84 km SE of CRL	82/08/16	253±9	82/08/19	271±9
24	Deep River			82/08/31	280±7
1	Balmer's Bay Gate	82/06/09	314±9	82/08/30	314±11
2	1 km east of gate			"	344±9
3	2.2 km east of gate		348±9	"	325±9
4	3 km east of gate			"	339±9
5	3.6 km east of gate	82/06/09	362±7	"	380±7
6	4.7 km east of gate			"	418±9
7	5.6 km east of gate	82/07/12	486±11	"	520±11
8	Road to Ottawa River from Balmer's Bay (BB) Road	82/06/21	362±14	"	332±9
9	Reactor Stack	82/06/22	1017±9	"	1607±14
51	Stack base, east side			82/08/11	1309±14
52	Stack base, west side			"	1103±18
49	17 m SE of stack			"	518±14
50	28 m ESE of stack			"	735±9
10	Public Information Centre	82/05/27	463±11	82/08/30	563±11
11	Emergency Basin, BB Road	82/08/06	364±11	"	319±11

Table 3a. Carbon-14 Concentrations in Vegetation, 1982 (Continued)

Site No. Fig. 2	Location	Poplar Leaves		Grass	
		Collection Date	(Bq.kg^{-1} C)	Collection Date	(Bq.kg^{-1} C)
12	Power line close to stack			"	405±9
13	Maskinonge Lk, Plant Road			"	287±9
14	Bldg. 560	82/06/07	285±11	82/08/30	314±14
15	1 km SW BB Road			82/08/31	353±11
16	1.8 km SW BB Road			"	375±9
17	Waste Man. Site C, Gate 33			"	920±14
44	" "			82/11/11	870±11
45	" "			"	400±9
46	100 m west of Gate 33			"	680±9
47	5 m west of Gate 31			"	3094±23
48	30 m N of Duke Stream Weir			"	1157±15
18	Upper Bass Lake			82/08/31	359±11
19	Lower Bass Lake	82/08/06	344±11	"	328±7
20	1.2 km SE Lower Bass Lake			"	391±7
21	0.8 km NW of Plant Road			"	251±9
22	4 km NNW of Main Gate			"	219±14
23	2 km east of Plant Road			"	325±7
25	CRL Library	82/05/26	382±11	82/09/08	524±5
26	Bus garages, CRL			"	411±7

Table 3a. Carbon-14 Concentrations in Vegetation, 1982 (Concluded)

Site No. Fig. 2	Location	Poplar Leaves		Grass	
		Collection Date	(Bq.kg^{-1} C)	Collection Date	(Bq.kg^{-1} C)
27	Bldg. 513, outside fence	82/05/13	610±11	"	443±7
28	Road to Power Plant	82/05/26	355±11	"	997±14
29	CRL Power Plant			"	368±9
30	Perch Lake, east side	82/06/21	321±5	82/09/03	344±9
31	Perch Lake, Inlet 5			"	386±9
32	Pte. au Baptême		319±11	"	319±9
33	East Gate			"	334±11
34	Perch Creek Weir			"	396±7
35	Chalk Lake, west shore			"	334±16
36	11 km SE of stack, Harrington Bay			82/10/04	294±11
37	5.5 km ESE of stack, Oiseau Bay			"	292±11
38	CRL wharf			"	380±9
39	1.5 km NE of stack, Quebec side			"	283±9
40	Cook's Cove, Quebec, opposite BB			"	285±14
41	McSourley Lake near Stonecliffe			82/10/28	280±14

Errors quoted based on counting statistics only (1 σ).

Table 3b. Carbon-14 Concentrations in Vegetation at Areas "B" and C", 1983

Site No. Figs. 10,11	Location	Grass	
		Collection Date	($Bq.kg^{-1}$ C)
53	Waste Management Site B	83/06/02	11402±34
54	" "	"	6791±38
55	" "	"	2005±14
56	" "	"	3017±14
57	" "	"	2619±18
58	Waste Management C, Beside Open Trench	"	36257±63
59	Waste Management C, Outside Gate	"	2118±18

See note at bottom of Table 3a regarding errors.

Table 4a. Carbon-14 Concentrations in Air and Vegetation, Collection Date 93/06/15

Site No. Figure 3	Location	Air	Poplar Leaves
		($Bq.kg^{-1}$ C)	($Bq.kg^{-1}$ C)
1	Bldg. 560	261.3±20.	
2	Maskinonge Lake	959.1±13.	
3	Upper Bass Lake	331.2±18.	
4	Balmer's Bay Gate	275.6±27.*	254.6±11.
5	Area F Road at Turnaround	303.9±19.	
6	Perch Lake (SE side)	287.0±27.	
7	Acid Rain Monitoring Site	441.1±44.*	320.8±14.
8	Twin Lakes Intersection	359.7±14.	
10	Perch Creek Weir	386.5±27.	
11	Stack Road at Fork	739.5±37.*	825.8±41.*
12	Snow Collection Site, NW of Stack	336.4±34.*	
13	Mattawa Road at Stack Road	402.4±40.*	
14	Cyclist Path at Powerline	278.7±28.	
15	Snow Collection Site, SW of Stack	423.6±42.*	331.1±13.
16	Ski Trail, NE of Stack	359.5±36.*	
17	Grey Silo on Public Information Centre Road	827.7±41.*	
18	Point McQuestin	456.7±46.*	
19	Pointe au Baptême	275.8±28.*	
20	Farm Access Road	381.4±38.*	340.8±15.
21	Environ. Res. Branch (Bldg. 513)	438.2±44.*	
22	Pump House (Bldg. 420)	461.3±46.*	364.6±13.
23	CRL Library (Bldg. 432)	656.5±33.*	

Table 4a. Carbon-14 Concentrations in Air and Vegetation, Collection Date 93/06/15 (Concluded)

Map No. Figure 3	Location	Air	Poplar Leaves
		(Bq.kg^{-1} C)	(Bq.kg^{-1} C)
25	SE Corner of Plant Site	311.2±31.*	
27	Old Logging Road	525.0±26.*	326.2±13
28	Quebec Shore, Ottawa River, East	285.0±28.*	
29	Quebec Shore, Ottawa River, West	316.4±13.	
30	Ontario Shore, Ottawa River, West	375.9±24.	
31	Stack Pipeline, Link 21	2185.2±109.*	
32	Public Information Centre	536.1±27.*	479±12
33	Bell Tower	349.2±35.*	

Errors quoted based on counting statistics only (1 σ).

* No error calculated at time of measurement; assumed to be ~ ±10% below 500 Bq.kg^{-1}; ~ ±5% above 500 Bq.kg^{-1}; ~ ± 1% above 1000 Bq.kg^{-1}.

Table 4b. Carbon-14 Concentrations in Air and Vegetation at Area "C", 1993-1994

Site No. Figure 11	Location	Air	Vegetation
		($Bq.kg^{-1}C$)	($Bq.kg^{-1}C$)
34	Duke Swamp C01	15030±113	
35	Duke Swamp C02	25651±204	
36	Duke Swamp C03	26628±190	
37	Duke Swamp C04	43380±59 63010±188	(cedar) 11290±115* (maple) 65751±658* (fern) 77840±778*
38	Duke Swamp C05	37683±219	
39	Duke Swamp C06	13590±91	
40	Duke Swamp C07	13870±62	
41	Duke Swamp C08	18483±180	
42	Duke Swamp C09	9015±100	
43	WM Area C	6547±65	
44	WM Area C	8077±81	
45	WM Area C	9682±97	
46	WM Area C Outside Fence	2053±31	

*See note at bottom of Table 4a regarding errors.

In 1982 the grass sample data gave the best coverage of the site, as well as being reasonably representative of the whole growing season, and hence were used to define the dispersion pattern for the year. The 1993 plot is based primarily on air measurements during a two-week period in June of that year, averaged over a longer time interval for sites where data were available. Contour plots of the specific activities measured in the two surveys (Tables 3a and 4a), corrected for background signal contribution, are shown in Figures 5 and 6. The highest levels measured in these surveys were in areas very close to the base of the stack, as was expected. Differences between the two surveys are sufficiently minor that they are more likely to be the result of variations in meteorological conditions than of substantial long-term changes in emissions.

Concentrations fall off rapidly with distance from the stack, most steeply in the directions perpendicular to the prevailing wind directions. The isopleths are roughly elliptical, with the

long axis oriented parallel to the river. It is obvious that wind direction plays a strong role in controlling the dispersion pattern. It is not clear what effect topography has on the pattern, but one observes that the 250 $Bq.kg^{-1}$ contour is not centred on the stack, but extends towards the lab site proper. This may be the effect of the hill upon which the stack is situated, or may have more to do with additional sources of ^{14}C within the site, such as the roof vents of the reactor buildings.

During both surveys, the meaured specific activity of ^{14}C at the main outer gate was never more than 10% above the background signal. However, at the gate on Balmer's Bay road, an equivalent distance from the stack, but in the principal wind direction, specific activities occasionally rose to 20% above background, with the signal in Deep River vegetation quite similar to that at the main outer gate. The highest ^{14}C specific activities measured on the CRL property, exclusive of areas adjacent to waste management sites or within 50 m of the base of the stack, were approximately three times background.

3.4 Calculation of Dispersion Factors

Emission data available for years 1993-1994 are listed in Table 5 (Rao and Young, in preparation.).

Table 5. Carbon-14 Stack Emission Data, 1993-1994

Sampling Period	Sampling Time (h)	^{14}C as CO_2	
		Bq/d	Bq/s
1993 May 25-26	18	1.55×10^9	1.79×10^4
1993 May 26-27	25	8.88×10^9	1.03×10^5
1993 May 27-28	23	0.62×10^9	7.18×10^3
1993 May 28-31	71	6.18×10^9	7.15×10^4
1994 May 16-17	24	7.3×10^8	8.45×10^3
1994 May 17-18	24	5.2×10^9	6.02×10^4
1994 May 18-19	24	1.1×10^9	1.27×10^4
1994 May 24-25	24	7.2×10^8	8.33×10^3
1994 May 25-26	24	6.4×10^9	7.41×10^4
1994 May 26-27	24	2.7×10^9	3.13×10^4
Av 1993 May		4.3×10^9	4.99×10^4
Av 1994 May		2.8×10^9	3.25×10^4

We have compared these measured emission rates with the contribution of ^{14}C from air in the reactor, calculated using measured ^{41}Ar emission rates for similar periods (Table 6).

Table 6. Air-derived and Measured Emission Rates of ^{14}C at CRL

Year	Measured ^{41}Ar emission (Bq day^{-1})+	Corresponding Calculated ^{14}C* emission (Bq day^{-1})	Measured ^{14}C emission (Bq day^{-1})	Measured ^{14}C/ Calculated ^{14}C	Measured HTO emission (Bq day^{-1})
1982	1.7×10^{14}	2.7×10^{9}			
1992	4.0×10^{13}	6.4×10^{8}			
1993 (5 days)	5.4×10^{13}	8.6×10^{8}	$(1.6 - 6.2) \times 10^{9}$ 4.30×10^{9} mean	5.0	$(1.9 - 19.8) \times 10^{10}$ 9.50×10^{10} mean
1994 (5 days)			$(0.7 - 6.4) \times 10^{9}$ 2.8×10^{9} mean		1.68×10^{11} mean
1994 (3 weeks)	4.6×10^{13}	7.4×10^{8}	$(3.0 \pm 0.2) \times 10^{9}$	4.1	$(2.2 \pm 0.9) \times 10^{11}$

+ ^{41}Ar measured on grab samples taken intermittently.

$$* \quad \frac{^{14}C \text{ production rate}}{^{41}Ar \text{ production rate}} = \frac{\sigma\,[^{14}N(n,p)^{14}C]}{\sigma\,[^{40}Ar(n,\gamma)^{41}Ar]} \times \frac{\%N_2 \text{ in air} \times \frac{1}{^{14}C\ t_{1/2}}}{\%\ Ar \text{ in air} \times \frac{1}{^{41}Ar\ t_{1/2}}}$$

A factor of four to five was observed between the calculated value and the five-day average measured values in both 1993 and 1994, calling into question our original assumption that the major supplier of ^{14}C emissions was air in the annular spaces of the reactors. However, a longer series of measurements in 1994 (three weeks) provided an average value much closer to that estimated for production from N_2 in air. Nevertheless, the high variability of the ^{14}C output leads us to believe that intermittent purging of cover gas containing evaporative losses of $^{14}CO_2$ from the moderator is probably the largest contributor to stack-emitted $^{14}CO_2$ at this site.

Table 7 lists atmospheric data collected weekly (passive samplers) and during 24-hour periods (active bubblers) at two sites during the same time intervals. The table includes a summary of atmospheric dispersion factors derived from concurrent 24-hour stack release and ambient air ^{14}C measurements at locations approximately 0.8 km northwest and southwest of the stack.

Dispersion factors for longer-term ambient air measurements were calculated using the average emission data listed in Table 5, which covered only five days and three weeks, respectively.

Table 7. Atmospheric Measurements at Two Environmental Sites, 1993-1994

Site	Period	Sampling Time	$Bq.kg^{-1}$ C (-Bkgd)	$Bq.m^{-3}$ Air	K
1	1993 June-Aug	3 months	196	0.035	1.4×10^6
1	1994 May-Aug	4 months	221	0.040	8.1×10^5
1	1994 May 13-20	7 days	68	0.012	2.3×10^6
1	1994 May 20-27	7 days	188	0.034	1.1×10^6
1	1994 May 27-June 7	7 days	407	0.073	4.4×10^5
1	1994 May 17	24 hours	384	0.069	4.9×10^5
1	1994 May 26	24 hours	182	0.033	1.6×10^6
2	1993 June-Aug	3 months	92	0.016	3.1×10^6
2	1994 May-Aug	4 months	60	0.011	2.9×10^6
2	1994 May 13-20	7 days	83	0.015	1.8×10^6
2	1994 May 20-27	7 days	30	0.0054	7.1×10^6
2	1994 May 25	24 hours	71	0.013	3.1×10^6
2	1994 May 26	24 hours	77	0.014	3.7×10^6

Site #1 - Public Information Centre, ≈ 0.75 km east from the base of the stack. Map No. 32, Figure 3.

Site #2 - Bell Communications Tower, ≈0.85 km west from the base of the stack. Map No. 33, Figure 3.

Atmospheric Dispersion Factor K = Q/C (m^3/s)

where C = average ^{14}C concentration in air (Bq/m^3)
Q = average ^{14}C release rate (Bq/s)

On the whole the dispersion factors measured are quite reproducible at both sites, both from year to year, and day to day, despite an order of magnitude variability in the source term. Changing winds may be the cause of some of the fluctuations in the observed dispersion. Meteorological data for May 17, 25 and 26 indicate that winds were quite high, and from the northwest on May 17, with gusty northwest winds on May 25th, and relatively calm conditions on the 26th.

3.5 Comparison of Vegetation and Air Sampling Results

It was mentioned in section 1 that it cannot be assumed that the ^{14}C in vegetation will necessarily be in equilibrium with that of the air in areas where frequent changes in emission levels are possible. Atmospheric ^{14}C measurements made in 1982 are shown graphically in Figure 7. Table 8 lists ^{14}C measurements in air, grass and poplar leaves sampled at the same 10 sites during that year.

Table 8. Levels of ^{14}C in Air and Vegetation at Same Sites in 1982 Survey

Site No.	Air $^{14}CO_2$* Bq.kg^{-1} C	Grass ^{14}C**		Poplar Leaves ^{14}C**	
		Bq.kg^{-1} C	Air/Grass	Bq.kg^{-1} C	Air/Poplar
1	346	314	1.10	314	1.10
3	398	325	1.22	348	1.14
5	536	380	1.41	362	1.48
7	728	520	1.40	486	1.50
11	454	319	1.42	364	1.25
27	545	443	1.23	484	1.13
31	294	386	0.91	321	1.09
14	350	314	0.94	285	1.03
19	398	328	1.21	344	1.16
24	303	280	1.08	---	---
Average Ratio			1.19 ± 0.19		1.21 ± 0.17

* Average concentration 1982 July 8-23.
** Grass collected at the end of August. Poplar leaves various June-August (see Table 3a).

Site nos. refer to Figure 2.

Air/vegetation ratios in both cases are seen to be approximately 20% above that for equilibrium, while corrections for isotopic fractionation are unlikely to account for more than a 3% discrepancy between these values. However, it was recognized that measurement errors were 10-15%, and that the range of values within a set of measurements for one site was large; consequently, the disagreement did not necessarily reflect a genuine disequilibrium condition.

When similar data collected at eight sites during a two-week period in 1993 June (Table 9) provided air/vegetation ratios similar to those of the 1982 survey (average ratio 1.22 ± 0.21), we decided that further investigation of the apparent disequilibrium was required.

Table 9. Levels of ^{14}C in Air and Vegetation at Same Sites in 1993 Survey

Site No. Fig. 3	Air $^{14}Co_2$ Jun 01-Jun 15	Poplar Leaves ^{14}C - June 15	
		Bq.kg^{-1} C	Air/Leaf
4	276	255	1.08
7	441	321	1.37
11	740	826	0.90
15	424	333Δ	1.27
20	381	341Δ	1.12
22	461	365	1.26
27	525	326	1.61
32	536	479	1.12

Δ denotes maple leaves

Average Ratio = 1.22 ± 0.21
Site Nos. refer to Figure 3

On the advice of Otlet and Walker, who had observed similar discrepancies in a survey conducted around the Sellafield reprocessing plant (1990), we continued these measurements at two sites every two weeks over the full season. In addition, we sampled air over both the period of photosynthesis (daylight hours) and over 24 hours. The results of these studies are shown in Figure 8 and Tables 10 and 11. The effect of all-season averaging is quite dramatic (all-season average air/vegetation ratio at one site was 0.96±0.10).

Table 10. Air-To-Leaf Ratios at the PIC* Site (#32, Fig. 3), 1993

Date of Sample Collection	Air ($Bq.kg^{-1}$ C)	Poplar Leaves ($Bq.kg^{-1}$ C)	Air/ Poplar	Maple Leaves ($Bq.kg^{-1}$ C)	Air/Maple	Alder ($Bq.kg^{-1}$ C)	Spruce Buds ($Bq.kg^{-1}$ C)	Spruce Needles ($Bq.kg^{-1}$ C)	Air/ Spruce Needles
93/15/13		625.5				609.2	718.1	504.5	
93/05/21	467.9	551.1	0.85			587.4	610.8	474.9	0.99
93/06/01	418.6	483.6	0.87					505.7	0.83
93/06/15	536	478.4	1.12						
93/06/17		518.2							
93/06/28	440	443	0.99						
93/07/09	494.6	469.5	1.05	471.3	1.05				
93/07/19	500.6	504.2	0.99						
93/07/29	704.1			482.8	1.46				
93/08/09	454.5			433.9	1.05				
93/08/19	385.6	462.5	0.83	460.6	0.84			478.7	0.81
93/08/30	461.6			486.6	0.95				
93/09/09	387.6	464.6	0.83	498	0.78				
93/09/22	420.2	474.3	0.89	468.7	0.90			475.3	0.88
93/10/06	370.9			492.3	0.75				
Mean Ratio			0.94		0.97				0.88
Seasonal Mean	464.78	484.94	0.96	474.28	0.98	598.30	664.45	487.82	0.95

* PIC: Public Information Centre

Note: Relative standard deviations 3-4%.

Input of lower specific-activity carbon from tree storage (1992 production) in the early weeks of the growing season provides a possible explanation for the discrepancies noted in May-June; a more likely cause is the large variability in the air emissions measured during short periods (Table 5). It is known that plant sugars are made and stored in a very short period; a summer project conducted by this group (Enright and Milton, in preparation) has indicated that the mean time for potential carbon replacement in a plant is approximately one week, with new material being well mixed within all growing parts of the plant in a matter of hours. Substantial variability in ^{14}C emissions toward the end of a two-week interval might be much more visible in the vegetation at the time of picking than in the integrated air sample. As anticipated, no significant variability in ^{14}C was observed between plant species at the same sites (Figure 7).

The error introduced by 24-hour sampling, rather than daylight hours only, has so far been less significant (Table 11).

Table 11. Changes in ^{14}C Levels in Atmospheric CO_2 ($Bq.kg^{-1}$ C) with Time of Day

	Sampling Location	
	Site 33	**Site 32**
Sampling period: 93/07/29 to 93/08/09		
Days (6:00-20:30)	339	470
Average (total 24-h sample)	372	455
Nights (calculated)	423	431
Days/average	0.91	1.03
Nights/average	1.14	0.95
Sampling period: 94/06/30 to 94/07/07		
Nights (21:00-5:30)	338	409
Days (5:30-21:00)	378	522
Average (weighted)	364	482
Days/average	1.04	1.08
Nights/average	0.89	0.78
Sampling period: 94/07/07 to 94/07/14		
Nights (21:00-5:30)	485	550
Days (5:30-21:00)	388	411
Average (weighted)	421	460
Days/average	0.92	0.89
Nights/average	1.25	1.34

3.6 Correlation of ^{14}C Output with Reactor Operations

To provide a more complete history of ^{14}C emissions at this site, a study in progress is accumulating tree ring chronologies (King, Repta and Enright, in preparation). Cross-sections of trees (either cut or cored) are being separated and analyzed; the data for one such white pine growing ≈600 m east of the stack are shown in Figure 9, along with the NRX and NRU power history during the growing months over the same period. The fluctuations in these data indicate that the major increases and decreases in measured ^{14}C specific activity correlate fairly well with variability in NRU power production. NRX would appear to be a rather minor contributor, contrary to the original hypothesis that the chief source of reactor ^{14}C was the activation of nitrogen in the larger amounts of air known to be present in the free spaces of this reactor. As discussed earlier, the chief sources of these ^{14}C emissions remain unresolved. However, the fact that the specific activity of tree ring carbon did not drop to background levels during periods of low to zero power strongly supports the contention that gas purging and moderator leak releases, which undoubtedly have continued intermittently during shutdown and repair periods, are also sources for heightened levels of ^{14}C in the vicinity of this tree.

3.7 Emissions From Waste Management Areas

During the course of these surveys, some sampling was carried out around Waste Management Areas B and C (Figures 10 and 11). The data collected are listed in Tables 3b and 4b. Since the prinicipal source of ^{14}C in these areas is soil de-gassing, atmospheric dispersion patterns are quite different from those observed for stack gas emissions. For this reason, and because of the paucity of data, no detailed analysis of these data has been attempted at this time.

4. CONCLUSIONS

Although monitoring of ^{14}C has not been carried out routinely at the Chalk River Laboratories, two site surveys of air and vegetation, ten years apart, have shown that the quantities vented from the reactor stacks are very rapidly dispersed atmospherically. Specific activities at the site boundaries are never higher than 20% above natural background levels. In 1994, an average dispersion factor of $>1.2 \times 10^6$ was measured ≈0.75 km from the reactor stack and $>1.1 \times 10^7$ at the site boundary.

All-season sampling (daylight hours only in the case of air) has smoothed out any discrepancies observed in the early measurements, and air/vegetation ratios are not significantly different from unity. Such ratios are also indicators of rapid mixing of releases with the local air masses.

It has been demonstrated that in the absence of good emission records, tree ring chronologies can be utilized to take the place of monitoring data, at least for the growing season.

5. ACKNOWLEDGEMENTS

The authors are very grateful for the analytical contributions of S. Enright, K. King, S. Kramer and W. Workman to this study.

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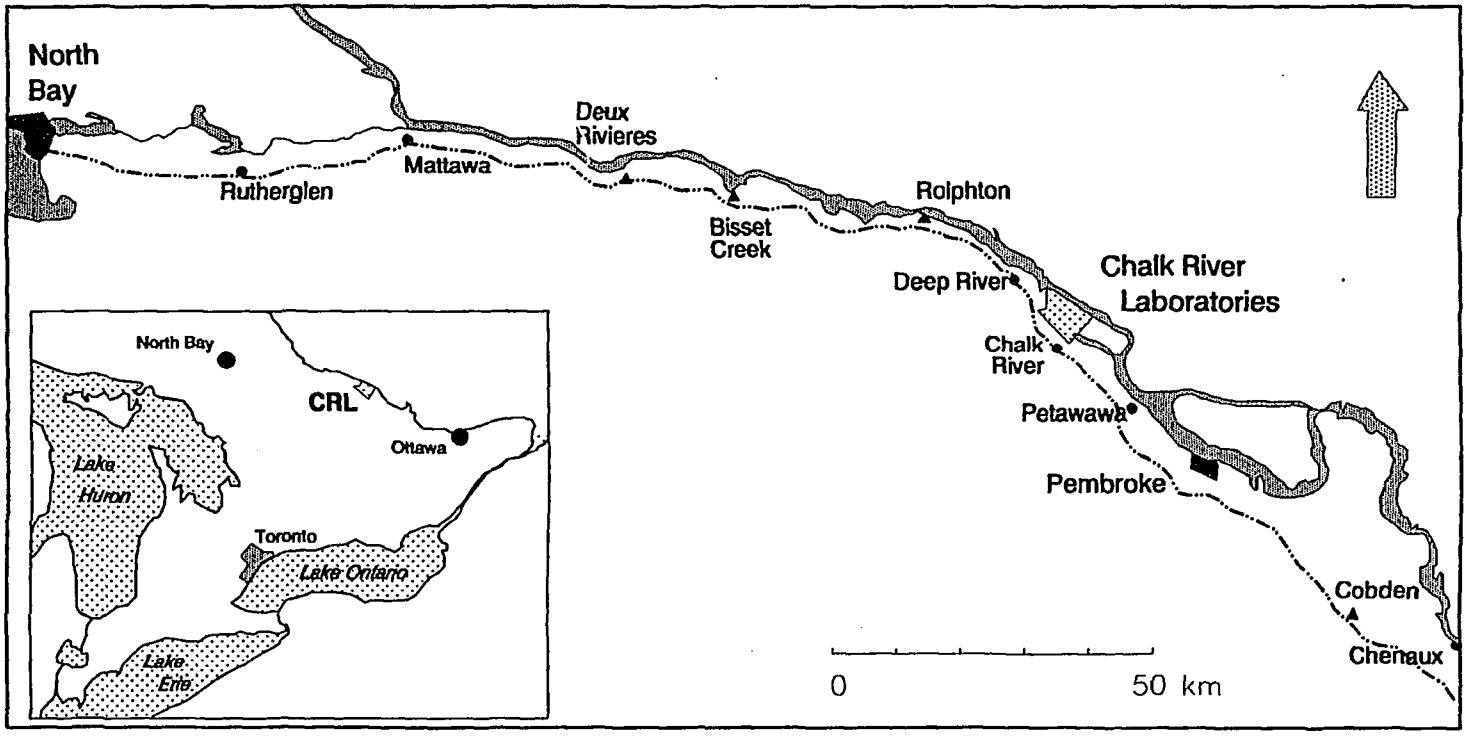


Figure 1. Map Showing the Location of the CRL Site.

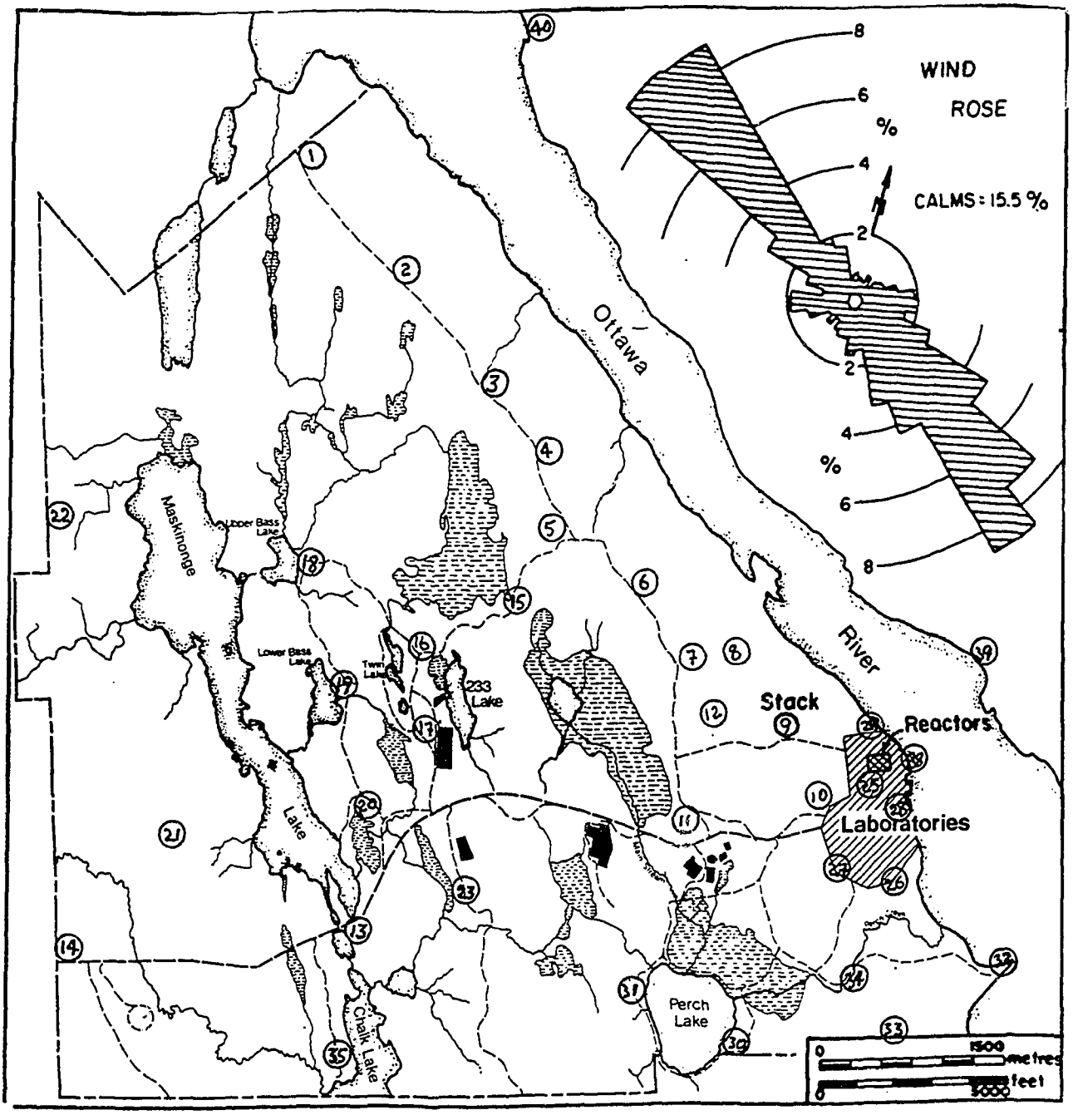
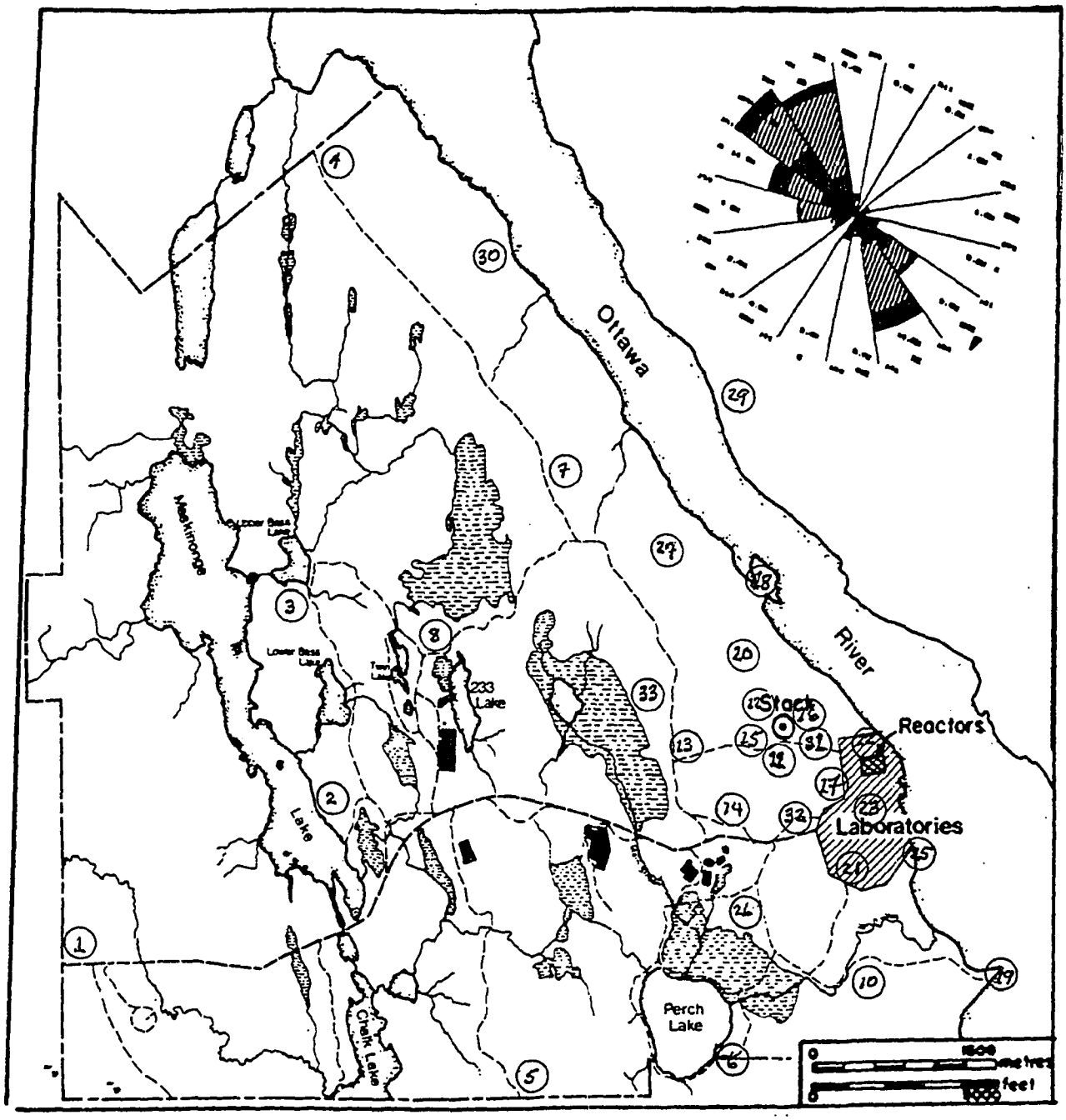


Figure 2. 1982 Collection Sites.

Sites Not Shown:
- 24: Deep River, 12 km NW of Stack (9)
- 36, 37: Sites on Quebec Shore SE of CRL
- 41: McSourley Lake, 30 km NW of CRL
- 44, 45, 46: Sites close to (17) outside Area C fence
- 48: Close to (20)
- 49, 50, 51, 52: Sites close to Stack (9)
- 60: Eganville, 84 km SE of CRL



Site not shown: 28: Québec shore, Ottawa River, across from (19)

Figure 3: 1993-94 Collection Sites

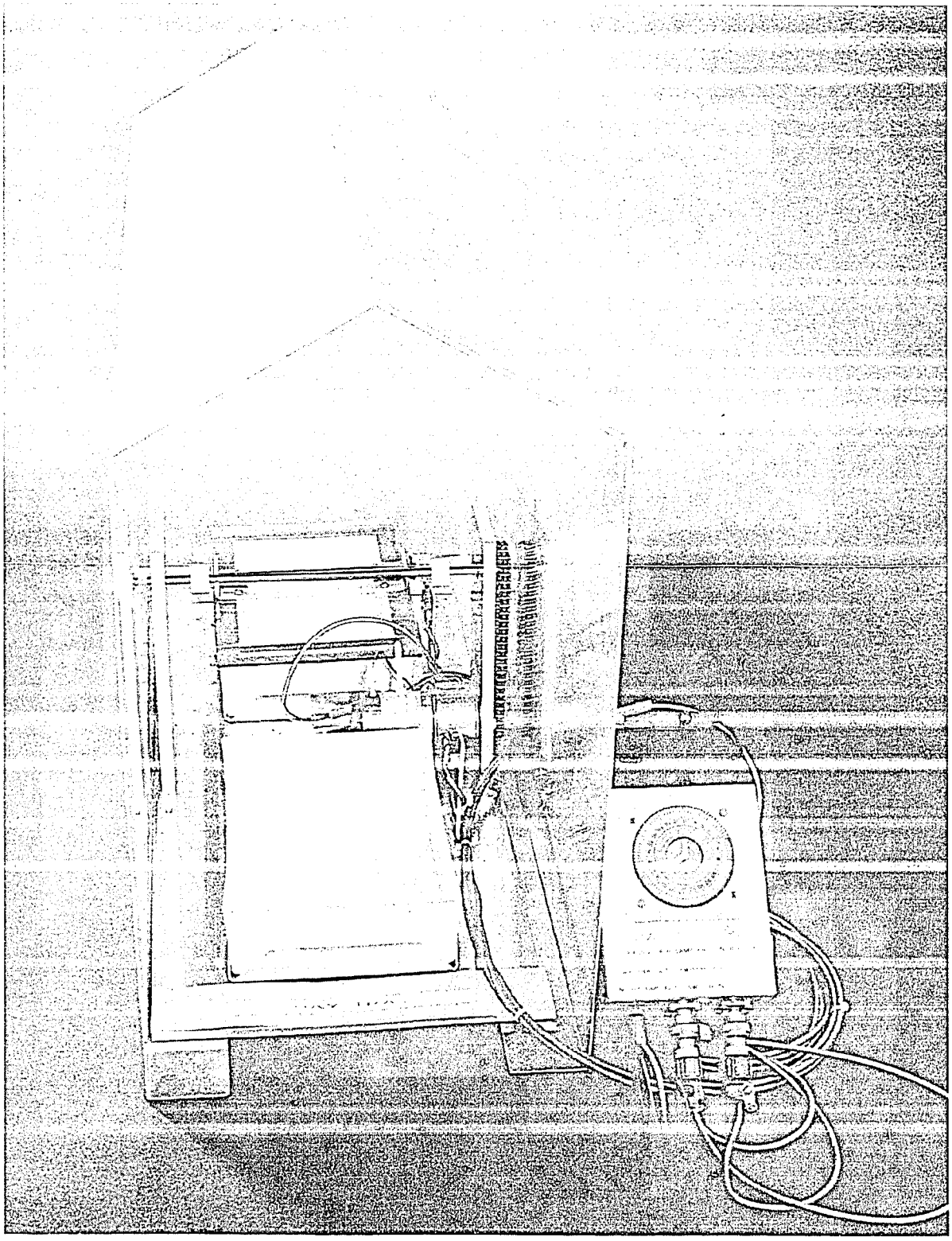


Figure 4: Photo of Day-Night Air Sampler

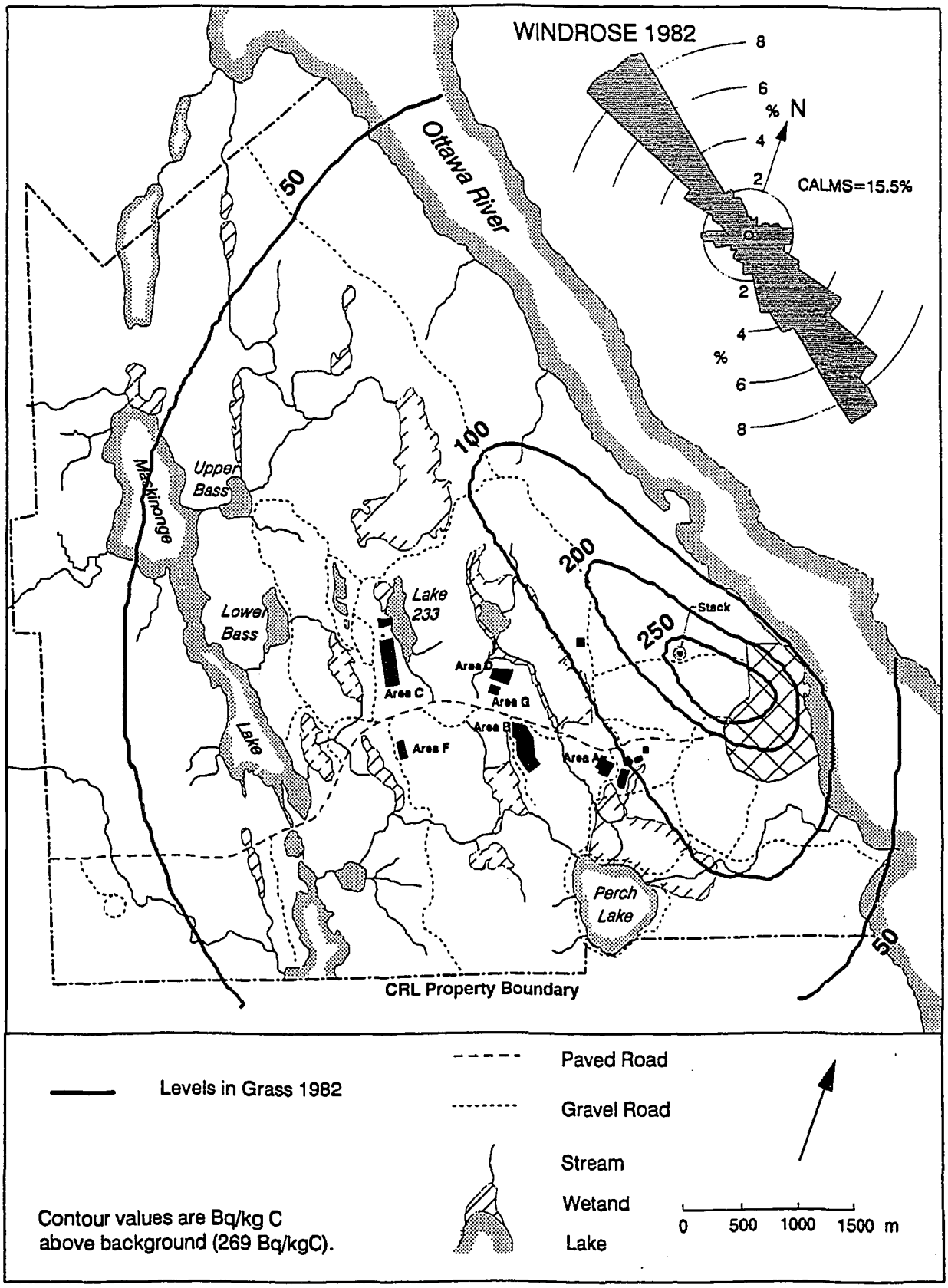


Figure 5: Carbon-14 Contours Drawn from Grass Survey, 1982

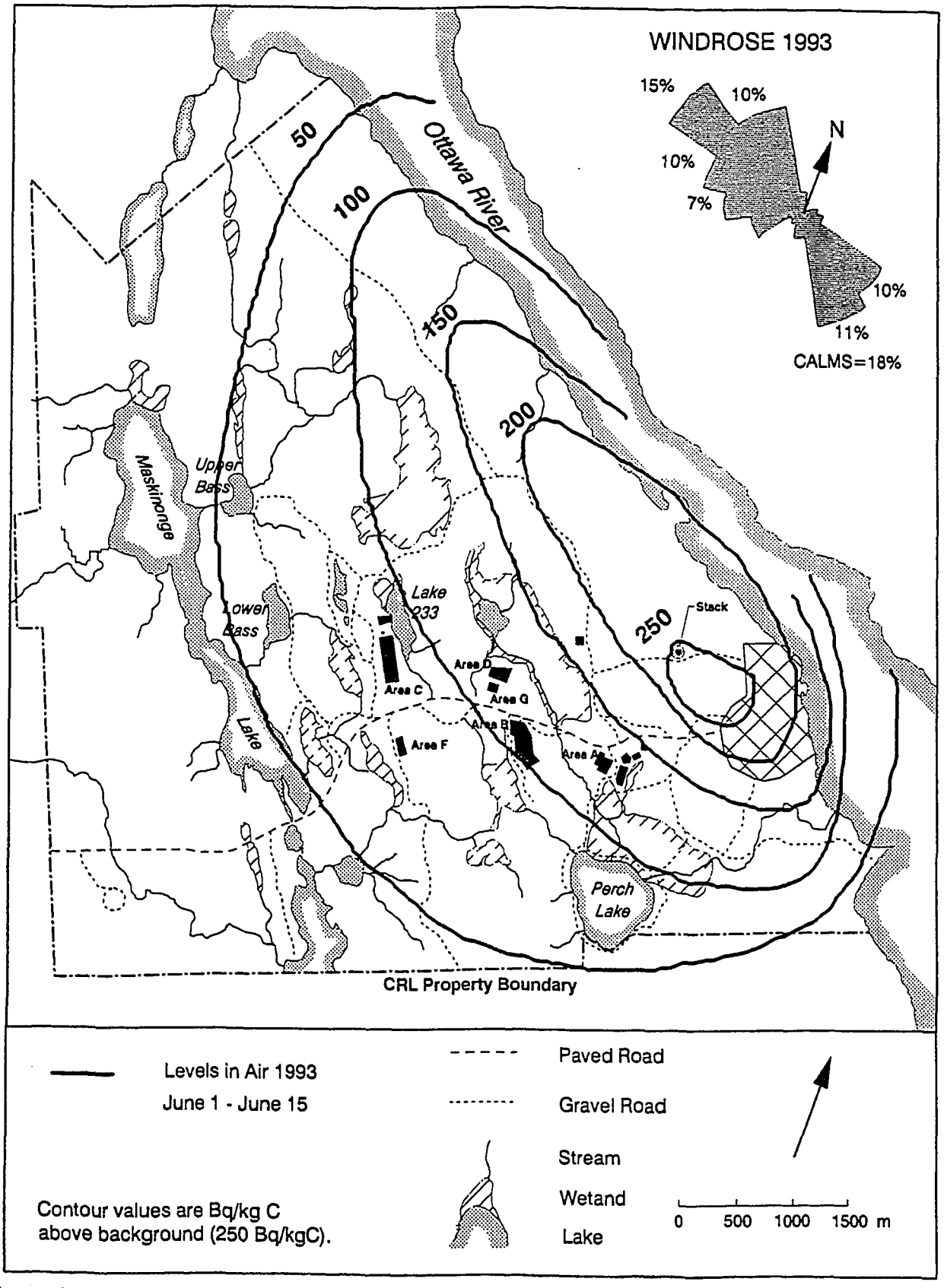


Figure 6: Carbon-14 Contours Drawn from Air Survey, 1993 June

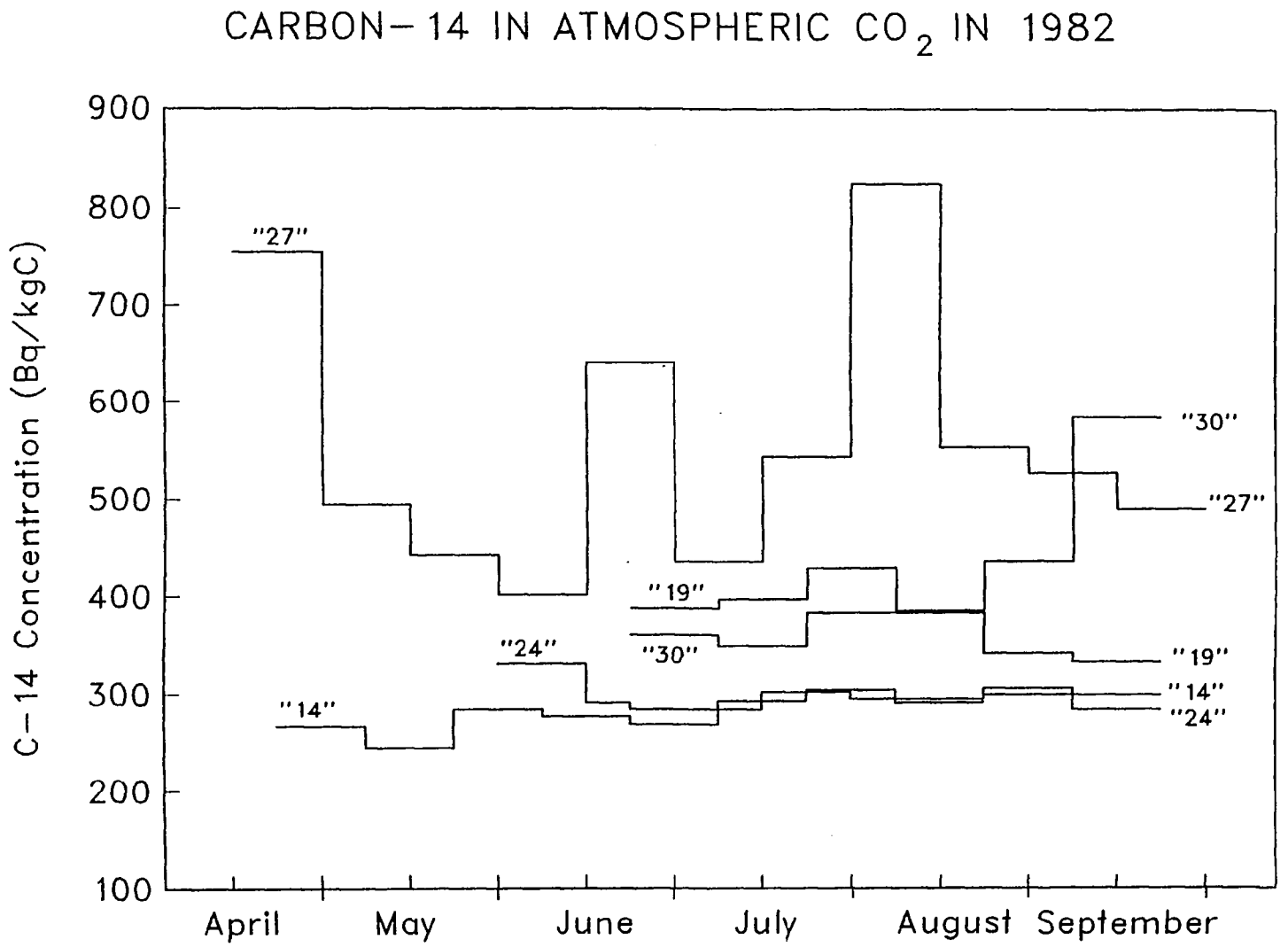


Figure 7: Carbon-14 in Atmospheric CO_2 in 1982
Numbers (e.g., "27") refer to Map Nos. of Fig. 2 and Table 3a

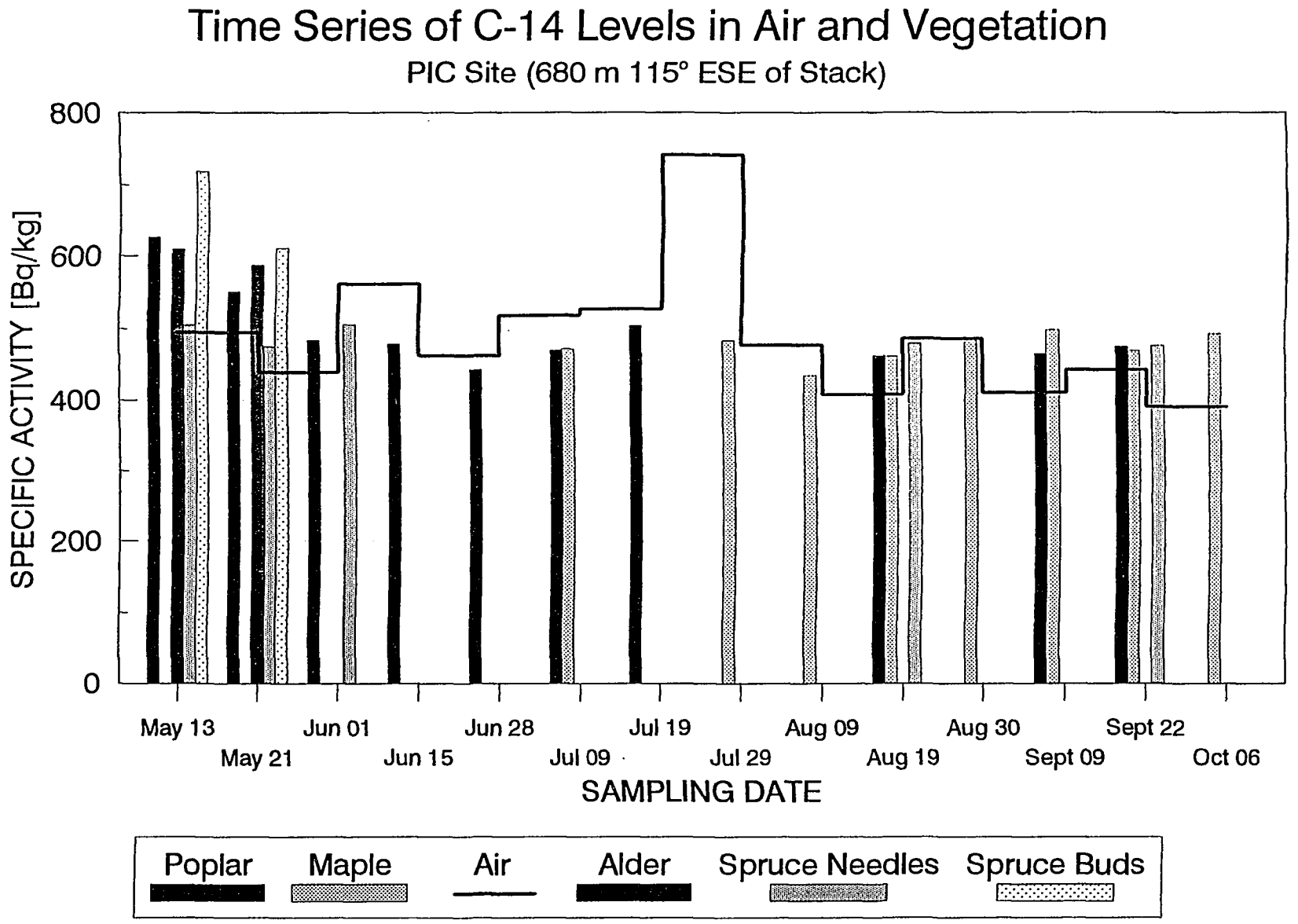


Figure 8: Carbon-14 in Air and Several Species of Vegetation at One Site, Throughout the 1993 Growing Season

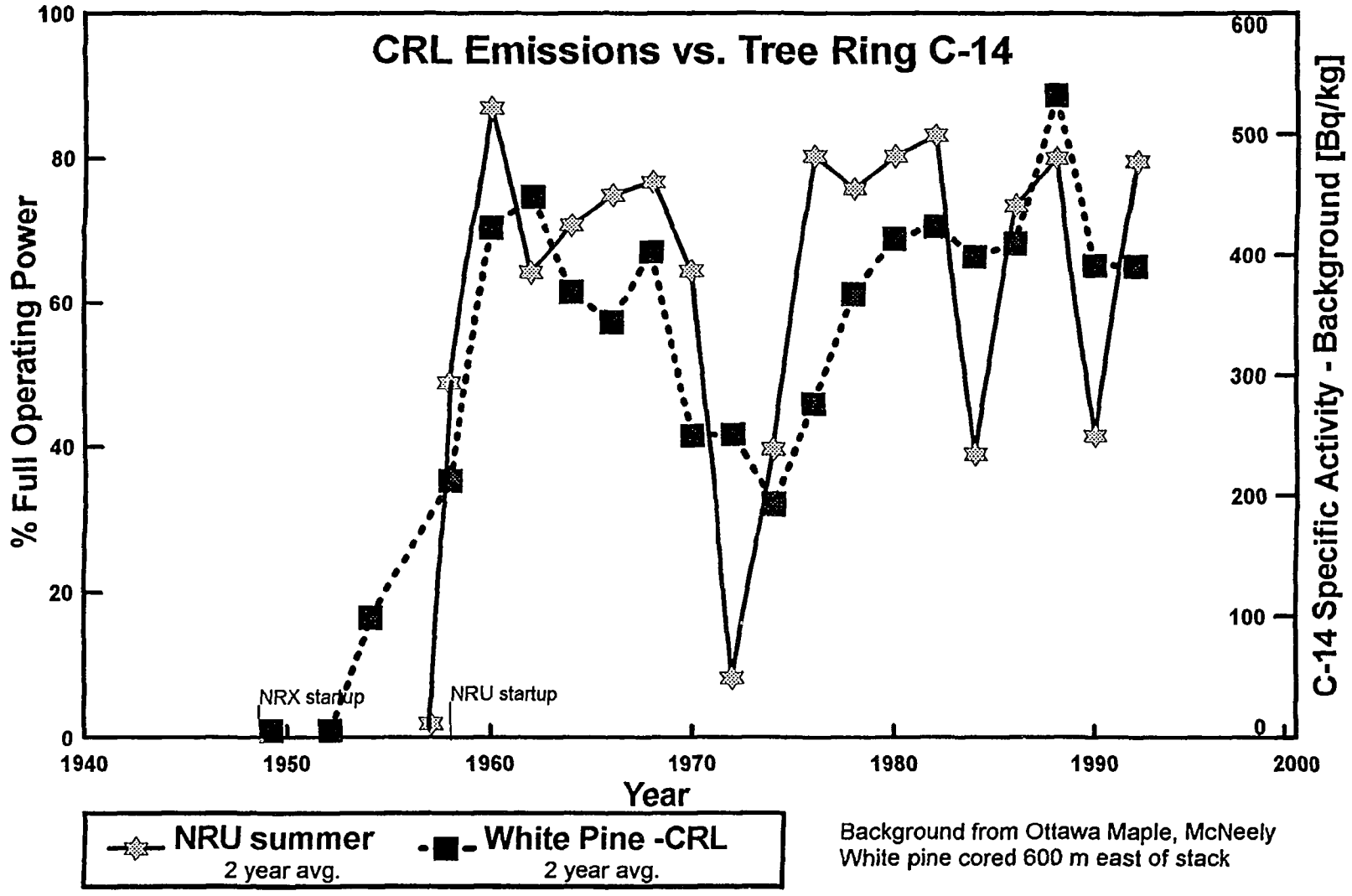


Figure 9: CRL Reactor Power Production vs Carbon-14 in Tree Rings

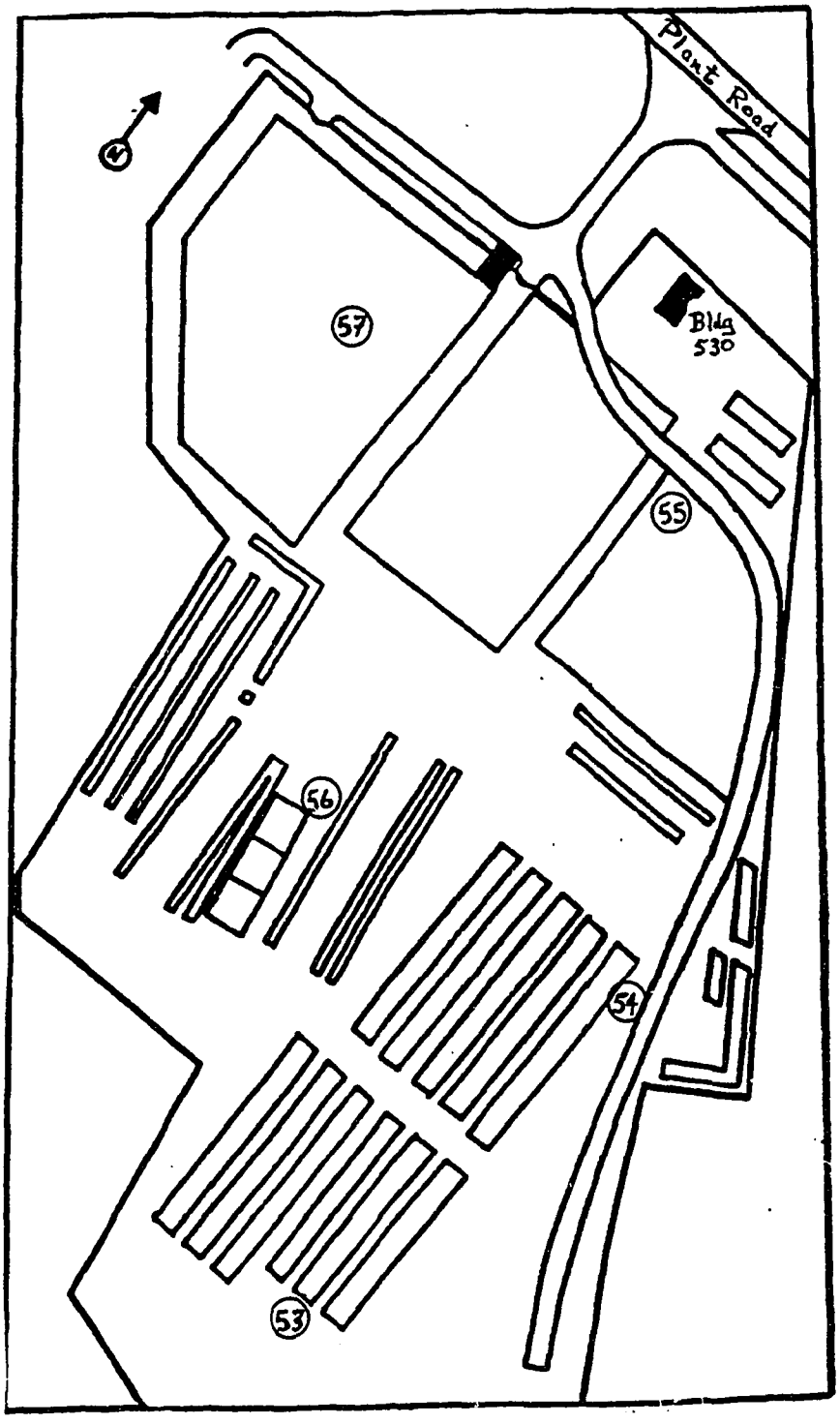


Figure 10: Site Map, Waste Management Area "B". Grass samples taken at edge of subterranium bunkers which extend about 50 cm above ground level. Sampled 1983 June 2.

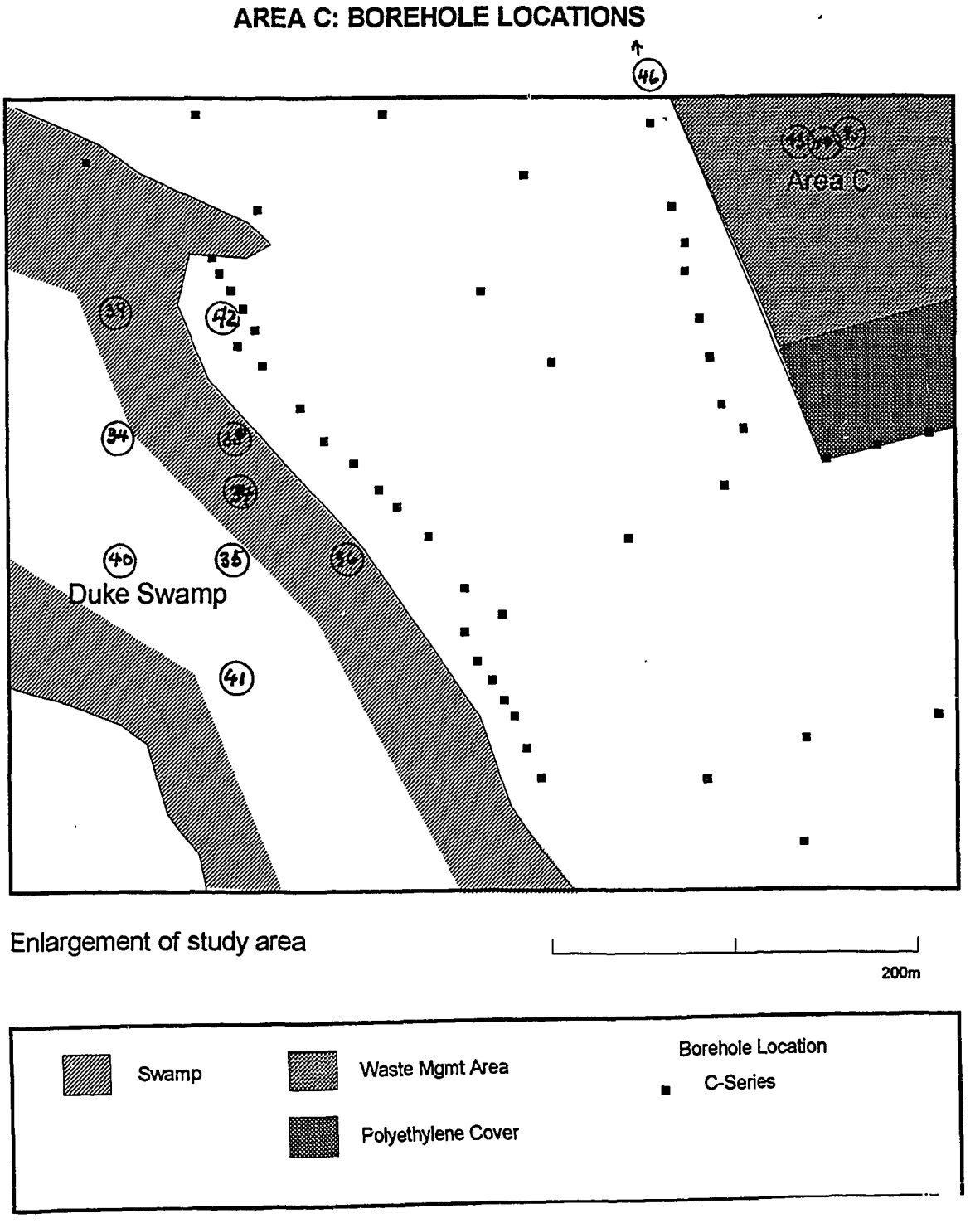


Figure 11: Site Map, Waste Management Area "C". Sample 1993, 1994.

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ISSN 0067-0367

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