

**ANNUAL CO₂ EVASION RATES
IN UNPRODUCTIVE SOFTWATER LAKES
IN CENTRAL ONTARIO**

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IN
UNPRODUCTIVE SOFTWATER LAKES IN CENTRAL ONTARIO**

Report prepared by:

Lewis A. Molot
Faculty of Environmental Studies
York University
4700 Keele Street
North York, Ontario M3J 1P3

and

Peter J. Dillon
Ontario Ministry of Environment and Energy
Dorset Research Centre
Bellwood Acres Road
P.O. Box 39
Dorset, Ontario POA 1E0

ABSTRACT

In an 8 year study of dissolved inorganic carbon in three unproductive softwater lakes in central Ontario, $\text{H}_2\text{CO}_3^\circ$ ($\text{H}_2\text{CO}_3 + \text{dissolved CO}_2$) was usually supersaturated in upper waters during the ice-free season. Net CO_2 evasion to the atmosphere occurred each year suggesting that carbon sources in the lakes were terrestrial rather than atmospheric. Differences in average evasion rates between lakes (8.4 - 25.4 g C $\text{m}^{-2} \text{yr}^{-1}$) largely disappeared when evasion rates were expressed per catchment area exclusive of lake area (2.9 - 3.8 g C $\text{m}^{-2} \text{yr}^{-1}$). Net annual carbon accumulation rates in temperate forested catchments, measured as the difference between gross photosynthetic production and autotrophic and heterotrophic respiration, may be overestimated by 2 to 8% because of stream and groundwater carbon export to lakes. Approximately 73 - 85% of exported C was discharged to the atmosphere from the lake surfaces.

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INTRODUCTION

The relative importance of CO₂ sources in temperate lakes and the role of lakes in regional carbon budgets is not well known. Norton and Henriksen (1983) stated that "oligotrophic non-dystrophic waters in North America and Europe are persistently and greatly oversaturated with CO₂ with respect to atmospheric conditions." Persistent CO₂ supersaturation rules out atmospheric invasion and implies that the catchment is a source of carbon. Hesslein *et al.* (1980) measured CO₂ evasion from softwater Lake 224 in northwestern Ontario during late July and August. Both CO₂ invasion and evasion occurred at different times of the summer in Lake 227, (a fertilized, softwater lake) in northwestern Ontario (Schindler and Fee 1973), presumably in response to changing photosynthetic consumption rates.

Rao (1978) reported that much of the dissolved inorganic carbon (DIC) in nutrient-poor Findley Lake in Washington state had been previously photosynthetically fixed and subsequently oxidized because it was dissimilar in isotopic composition from atmospheric CO₂ and concluded that nonpelagic sites were the sources of this DIC. Kling *et al.* (1991) observed CO₂ supersaturation in 27 out of 29 aquatic systems in arctic Alaska and suggested that transport of dissolved carbon from terrestrial to aquatic systems is a significant fraction of annual carbon accumulation.

If unproductive lakes are persistently CO₂ supersaturated, they offer a unique opportunity for measuring net annual carbon evasion rates and refining regional carbon fluxes. This communication presents the results of an intensive 8-year study of DIC in three unproductive, stratified lakes in central Ontario. Annual air/water CO₂ flux rates and organic carbon sediment accumulation rates were estimated and the role of the lakes in regional carbon budgets was analyzed.

STUDY LAKES

Dickie, Harp and Plastic Lakes are located in central Ontario in Haliburton County or the District of Muskoka east of Georgian Bay (Figure 1). The catchments are primarily forested and are underlain by Precambrian metamorphic silicate bedrock (Molot *et al.* 1989; Dillon and Molot 1990; Dillon *et al.* 1991). The Plastic catchment has surficial deposits of thin Pleistocene glacial till (<1 m) and small areas of peat overlying sand. The Dickie catchment has surficial deposits of thin Pleistocene glacial till (<1 m) with large areas of peat. The Harp catchment has extensive till plains (>1 m) with some areas of thin till and exposed rock ridges. Smaller deposits of sand and peat (overlying sand) occur. Mean depth, maximum depth and lake and catchment surface areas are listed in Table 1. Harp and Dickie lakes have some lakeshore cottage development, while Plastic Lake has none.

The lakes are softwater (mean conductivity ranging from 29 to 38 μS) and oligotrophic to slightly mesotrophic with long-term mean annual epilimnetic total phosphorus (TP) concentrations ranging from 4.8 $\mu\text{g P L}^{-1}$ in Plastic Lake to 10.7 g P L^{-1} in Dickie Lake (Table 1). Long-term mean ice-free chlorophyll *a* (Chl_{ip}) in the euphotic zone (twice the Secchi depth) ranged from 2.8 $\mu\text{g L}^{-1}$ in Plastic Lake to 4.7 $\mu\text{g L}^{-1}$ in Dickie Lake (Molot and Dillon 1991). Mean Secchi depth during summer stratification ranged from 2.8 m in Dickie to 6.8 m in Plastic Lake. Dissolved organic carbon (DOC) ranged from 2.3 mg C L^{-1} in Plastic Lake to 5.0 mg C L^{-1} in Dickie Lake (Table 1).

Plastic Lake is the most acidic with measurable decreases in pH and alkalinity occurring in recent years (whole-lake, ice-free alkalinity was 15 $\mu\text{eq L}^{-1}$ in 1980 and 7 $\mu\text{eq L}^{-1}$ in 1985, and whole-lake, ice-free pH was 5.6 in 1985) (Dillon *et al.* 1987). Harp Lake is

the least acidic but is still considered acid-sensitive with a mean whole-lake alkalinity of 61 $\mu\text{eq L}^{-1}$ and pH 6.3 in 1985 (Table 1).

METHODS

One station in each lake was sampled from 1980 to 1987 at 1 to 4 week intervals. Samples were collected at 2 m intervals beginning at 1 m below the lake surface and transported to the laboratory for measurement of pH and DIC. DIC samples were acidified with H_2SO_4 and CO_2 was measured using an autoanalyzer with phenolphthalein indicator. Analytical methods are reported in Locke and Scott (1986).

$\text{H}_2\text{CO}_3^{\circ}$ concentrations ($\text{H}_2\text{CO}_3 + \text{CO}_{2\text{aq}}$) were calculated from DIC, *in situ* temperature and pH according to Stumm and Morgan (1981). Equilibrium $\text{H}_2\text{CO}_3^{\circ}$ concentration ($\text{H}_2\text{CO}_3^{\circ}_{\text{eq}}$) was estimated from $\text{H}_2\text{CO}_3^{\circ}_{\text{eq}} = K_{\text{H}} P_{\text{CO}_2\text{eq}}$ where K_{H} is Henry's Law constant which is temperature dependent and $P_{\text{CO}_2\text{eq}}$ is the partial pressure of CO_2 in lake water at atmospheric equilibrium which was taken to be $10^{-3.5}$ atm (Stumm and Morgan 1981). Ratios of $\text{H}_2\text{CO}_3^{\circ} / \text{H}_2\text{CO}_3^{\circ}_{\text{eq}}$ greater than 1 in lake water indicate supersaturation. Atmospheric fluxes of CO_2 to and from lake surfaces during ice-free periods were calculated from;

$$\text{Flux} = (\text{H}_2\text{CO}_3^{\circ} - \text{H}_2\text{CO}_3^{\circ}_{\text{eq}}) D/z$$

where D is the molecular diffusion coefficient ($1.9 \times 10^{-5} \text{ cm}^2 \text{ sec}^{-1}$) and z is the thickness of the stagnant boundary layer, taken to be 300 μm (modified from Hesslein *et al.* 1980). The $\text{H}_2\text{CO}_3^{\circ}$ concentration at 1 m was taken to be the concentration at the bottom of the stagnant boundary layer. The ice-free period was assumed to be May 1

through November 30 of each year.

Annual sediment C accumulation rates were estimated for the top 1 cm of sediment cores by multiplying C/P sediment ratios by the amount of TP retained annually measured with mass balance studies of stream inflows, lake outflow and precipitation (Dillon and Evans 1992). The difference between TP inputs and outflows is assumed equivalent to sediment storage (Evans and Rigler 1980; Cross and Rigler 1983; Dillon and Evans 1992). Details of the P mass balance study will be reported elsewhere (P.J. Dillon *et al.* 1992. unpub. data). Briefly, stream samples were collected approximately one to four times per week and water level or stage was recorded continuously at weirs or flumes installed on the study streams. Stage-discharge relationships were constructed for each stream.

Five to eight cores were taken from each lake at depths ranging from 8.3 m to the lake maximum depths using a KB gravity corer with a 4.8 cm inside diameter core tube. One to three cores were obtained at each station and sectioned using an extruder and plastic collar. Replicate slices were combined, dried at 100°C to a constant mass for a minimum of 24 hrs and ground to a fine powder. Loss on ignition (LOI) was determined gravimetrically after igniting samples at 600°C for 1 hr. Sediment LOI was converted to carbon by assuming that organic matter was 51% carbon by dry weight. Subsamples for TP determination were digested in hot sulfuric acid with the addition of potassium persulfate. TP was measured colourimetrically using an ammonium molybdate-stannous chloride method (Ontario Ministry of the Environment 1983) after eliminating Fe interference by precipitation and filtration.

Mean annual precipitation (m yr^{-1}) was estimated for 1980-1987 using 4 to 12 sampling stations. The catchments are sufficiently close such that precipitation measurements were considered representative of the three study catchments. Annual baseflow, quickflow and runoff (m yr^{-1}) for the same period as precipitation were estimated for each stream subcatchment (Dillon *et al.* 1991) and area-weighted to produce total estimates for each catchment. Baseflow was defined as the minimum observed stream discharge within each month. runoff was the measured stream discharge to the lakes and quickflow was defined as runoff less baseflow.

RESULTS

The study lakes were frequently supersaturated with $\text{H}_2\text{CO}_3^{\circ}$ during the ice-free seasons at all depths, with Harp Lake the most supersaturated and Plastic Lake the least supersaturated. Undersaturation in the upper waters, an indication of high photosynthetic activity relative to supply, occurred occasionally but never below 5 m in Dickie and Harp Lakes and never below 7 m in Plastic Lake which was more transparent (Table 1). Between 1980 and 1987 at 1 m depth, Dickie Lake was undersaturated on 15 out of 104 ice-free sample dates, Harp Lake was undersaturated on only 3 out of 112 sample dates and Plastic Lake was undersaturated on 27 out of 121 sample dates.

Supersaturation typically increased with depth (Figure 2). On occasion, $\text{H}_2\text{CO}_3^{\circ}$ supersaturation occurred at 1 m with a zone of undersaturation occurring immediately below these waters. Since $\text{H}_2\text{CO}_3^{\circ}$ originating from microbial respiration in deep sediments can not diffuse from deep, supersaturated waters against a concentration

gradient to reach supersaturated surface waters, supersaturation at 1 m probably resulted from shallow sources of H_2CO_3^* such as stream flow, groundwaters or sediment respiration with rapid lateral mixing.

Net annual CO_2 gas evasion rates from the lakes varied from year to year (Figure 3). On an annual basis, there was no net atmospheric invasion. The evasion rates ranged from a minimum of $4.9 \text{ g C m}^{-2} \text{ yr}^{-1}$ in 1981 in Plastic Lake to $33.7 \text{ g C m}^{-2} \text{ yr}^{-1}$ in 1980 in Harp Lake. The mean annual CO_2 evasion rates were 15.2, 25.4 and $8.4 \text{ g C m}^{-2} \text{ yr}^{-1}$ from Dickie, Harp Lakes and Plastic Lakes, respectively (Table 2).

Sediment carbon accumulation rates expressed per lake area were 5.6, 4.3 and $2.1 \text{ g C m}^{-2} \text{ yr}^{-1}$ in Dickie, Harp Lakes and Plastic Lakes, respectively (Table 2). Total net evasion and sediment accumulation rates expressed per lake area were 20.8, 29.7 and $10.5 \text{ g C m}^{-2} \text{ yr}^{-1}$ in Dickie, Harp and Plastic Lakes, respectively. Approximately 73 - 85% of exported C was discharged to the atmosphere from the lake surfaces.

Net retention of total carbon (DOC + DIC) during 1979 to 1988 as determined by mass balance studies was 24.4, 13.7 and $10.8 \text{ g C m}^{-2} \text{ yr}^{-1}$ expressed per lake area in Dickie, Harp and Plastic Lakes, respectively (P.J. Dillon unpublished data). Retained carbon consisted of 81%, 93% and 100% DOC in Dickie, Harp and Plastic Lakes, respectively.

Differences in CO_2 lake evasion rates expressed per lake area could be due to differences in carbon supply rates to surface waters, carbon export rates via outflows, photosynthetic demand or undetected carbon-rich groundwater. Differences in areal chlorophyll densities are too small to account for the large differences in flux rates (Table 2). For example, the evasion rate from Harp is three times that of Plastic but

the chlorophyll areal density in Plastic Lake is only 1.1 times that of Harp. A high input of carbon in groundwater to Harp Lake, a possibility because of its relatively thick tills, would account for the evasion plus accumulation rates being much higher than the mass balance retention estimate which assumes negligible groundwater input.

Differences between lakes decreased when evasion rates were expressed per catchment area (exclusive of lake area) rather than lake area. Normalized for catchment areas, the evasion rates were 3.5, 3.8 and 2.9 g C m⁻² yr⁻¹ in Dickie, Harp and Plastic Lakes, respectively (Table 2). Total net evasion and accumulation rates expressed per catchment area were 4.8, 4.4 and 3.6 g C m⁻² yr⁻¹ in Dickie, Harp and Plastic Lakes, respectively.

DISCUSSION

Persistent net annual evasion rates from the three study lakes strongly suggests that carbon from the terrestrial portion of the catchment was the major carbon source for primary producers and that atmospheric invasion was negligible. Since most of the C load was organic (P.J. Dillon, unpublished data), we infer that mineralization of organic carbon in shallow waters was sufficient to produce persistent epilimnetic CO₂ supersaturation and net annual CO₂ evasion. Agreement between estimates of net retention from mass balance calculations and evasion plus sediment accumulation rates lends confidence to the accuracy of both methods. The relatively high evasion rate in Harp may also be due to a significant groundwater source of carbon.

Terrestrial carbon export may also be a significant source of CO₂ in more productive lakes even though high photosynthetic demand produces persistent CO₂ under-saturation and, hence, atmospheric invasion.

If persistent supersaturation and evasion is typical of unproductive lakes in temperate, boreal and arctic regions (Rao 1978; Hesslein *et al.* 1980; Kling *et al.* 1991), atmospheric invasion of CO₂ into northern freshwaters is insignificant; hence, elevated atmospheric CO₂ levels will likely not result in higher dissolved CO₂ levels.

The average net CO₂ evasion rate ranged from lowest in the most acidic lake (Plastic) to highest in the least acidic lake (Harp) which is consistent with laboratory studies which have shown that decomposition rates of newly sedimented material decrease with decreasing pH (Kelly *et al.* 1984). However, between-lake differences in evasion plus sedimentation rates are greatly reduced when rates are expressed per catchment area, suggesting that carbon export rates are similar between catchments. Hence, between-lake differences may be accounted for more by catchment and lake morphologies than by degree of acidification.

Assessing the role of these lakes in regional carbon budgets depends on estimates of net annual carbon accumulation in biomass and soil. Net annual carbon accumulation rates over short periods of time can be measured as net ecosystem production (NEP) which is the difference between gross photosynthetic production and autotrophic and heterotrophic respiration. The uncertainty associated with NEP is large because small changes in gross photosynthetic production or respiration can result in large changes in NEP. NEP ranged from 56 g C m⁻² yr⁻¹ in a mixed deciduous forest in Tennessee, USA to 265 g C m⁻² yr⁻¹ in an oak-pine forest in New York, USA (Edwards *et al.* 1981;

Whittaker *et al.* 1979). Lake evasion rates expressed per catchment area averaged $4.3 \text{ g C m}^{-2} \text{ yr}^{-1}$ in this study; hence, if this NEP range is typical of northern temperate ecosystems, net annual carbon accumulation may be overestimated by 2 to 8% because of stream and groundwater carbon export to lakes. Stream export of C was approximately 1% of NEP in Hubbard Brook (Whittaker *et al.* 1979).

Net annual carbon accumulation in the Alaskan arctic based on differences between gross ecosystem photosynthesis and respiration rates may also be overestimated because of significant terrestrial export of dissolved carbon to aquatic systems (Kling *et al.* 1991). Hence, net global annual carbon accumulation at temperate and arctic latitudes may be less than previously thought.

Carbon-14 dating of peat deposits can be used to estimate net carbon accumulation rates over much longer time intervals. Estimates of carbon accumulation rates in eastern Canada peatlands ranged from 11 to $59 \text{ g C m}^{-2} \text{ yr}^{-1}$ over periods ranging from 1320 to 7620 years (Bowville *et al.* 1983). These estimates fall mostly below the NEP range. The different ranges can be reconciled if boreal NEP is at the low end of the NEP range and/or much of the accumulated biomass is periodically burned and returned to the atmosphere. The average lake evasion plus sediment accumulation rates expressed per catchment area in this study, 4.3 g C yr^{-1} , is 7 to 39% of the long term peatland carbon accumulation rate.

However, DOC appears to contain relatively recent carbon (post-bomb) in Harp Lake (Schiff *et al.* 1990), suggesting that most peat remains resistant to weathering and mineralization and that the exported C is likely derived from recently viable communities. Since the C export rate is a linear function of the extent of peatlands

(P.J. Dillon, unpublished data), we can conclude further that exported C is derived from recently viable terrestrial communities associated in large part with peatlands.

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Table 1. Mean and maximum depths (m), residence time (yr), lake surface area (ha), catchment area (ha) (exclusive of lake area), mean annual epilimnetic TP ($\mu\text{g L}^{-1}$), mean ice-free chlorophyll a in the euphotic zone, mean Secchi depth (m) during summer stratification, pH, alkalinity ($\mu\text{eq L}^{-1}$) and mean annual whole-lake DOC (mg C L^{-1}). TP, chlorophyll, Secchi and DOC are means for 1976-1987 for Dickie and Harp Lakes and 1979-1987 for Plastic Lake. Alkalinity and pH are whole-lake means for 1985.

	Dickie	Harp	Plastic
Mean depth	5.0	13.3	7.9
Maximum depth	12.0	37.5	16.3
Residence time	1.7	3.2	3.8
Surface area	93.6	71.4	32.1
Catchment area	406.4	470.7	92.5
TP	10.7	6.7	4.8
Chlorophyll $\mu\text{g L}^{-1}$	4.7	32	2.8
Chlorophyll mg m^{-2}	1.9	2.0	2.2
Secchi	2.8	3.8	6.8
pH	5.8	6.3	5.6
Alkalinity	18	61	7
DOC	5.0	3.9	2.3

Table 2. Mean (\pm standard deviation) CO₂ evasion rates, sediment carbon accumulation rate in surficial sediments (top 1 cm) per lake surface area and per catchment area exclusive of lake area and mass balance estimates of net retention of total C (Dillon, unpublished data). CO₂ evasion rates are means for 1980-1987. Units are g C m⁻² yr⁻¹ and mg TP m⁻² yr⁻¹.

	Dickie	Harp	Plastic
CO ₂ evasion rate:			
per lake area	15.2 \pm 3.6	25.4 \pm 7.6	8.4 \pm 1.9
per catchment	3.5 \pm 0.8	3.8 \pm 1.1	2.9 \pm 0.6
Surficial sediment accumulation:			
C/P (weight)	107 \pm 14	83 \pm 5	104 \pm 27
P per lake area	52.7	51.7	20.2
C per lake area	5.6	4.3	2.1
C per catchment area	1.3	0.6	0.7
Total evasion + accumulation:			
per lake area	20.8	29.7	10.5
per catchment	4.8	4.4	3.6
Mass balance retention:			
per lake area	24.4	13.7	10.8

Figure 1

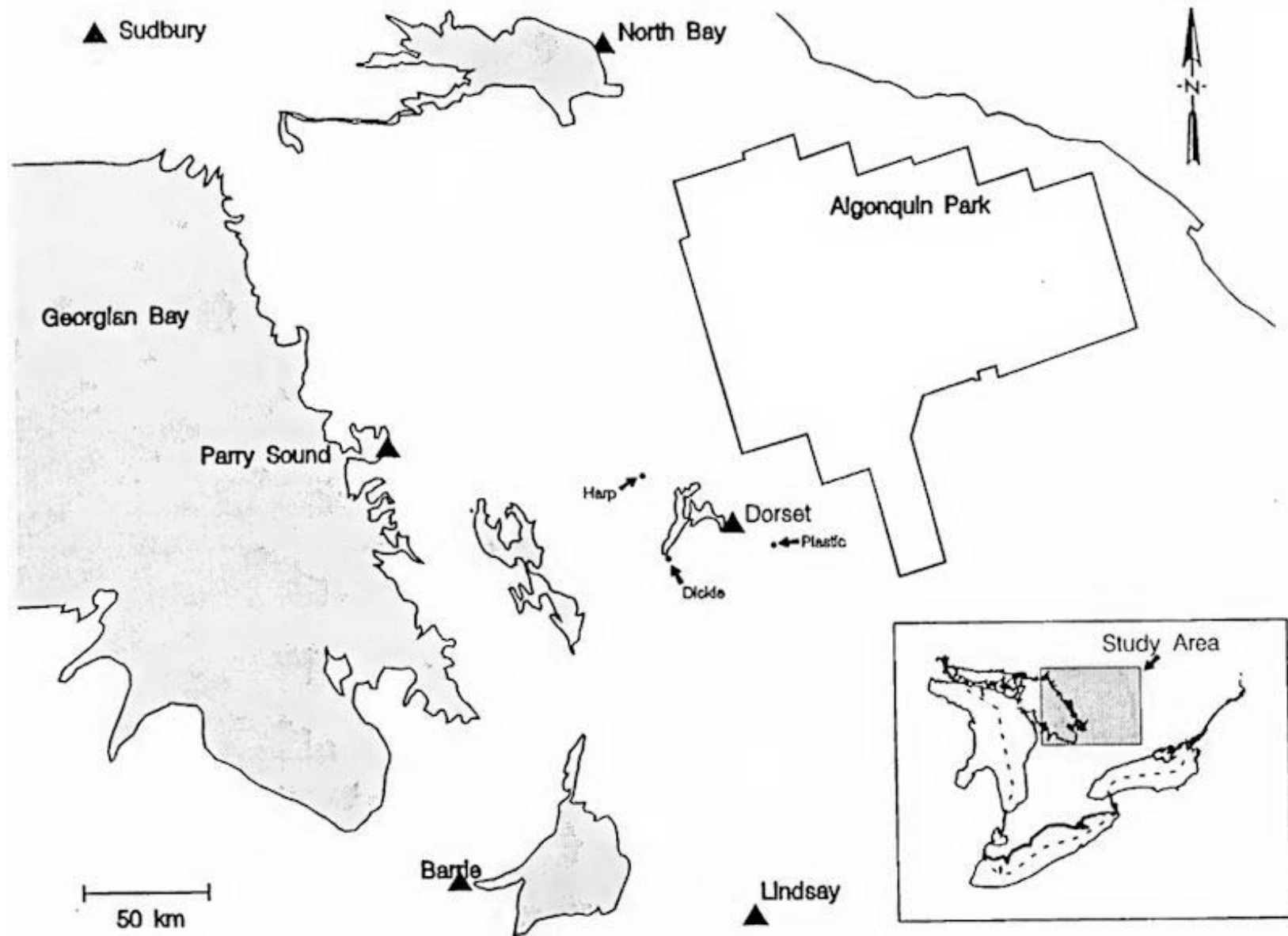


Figure 2

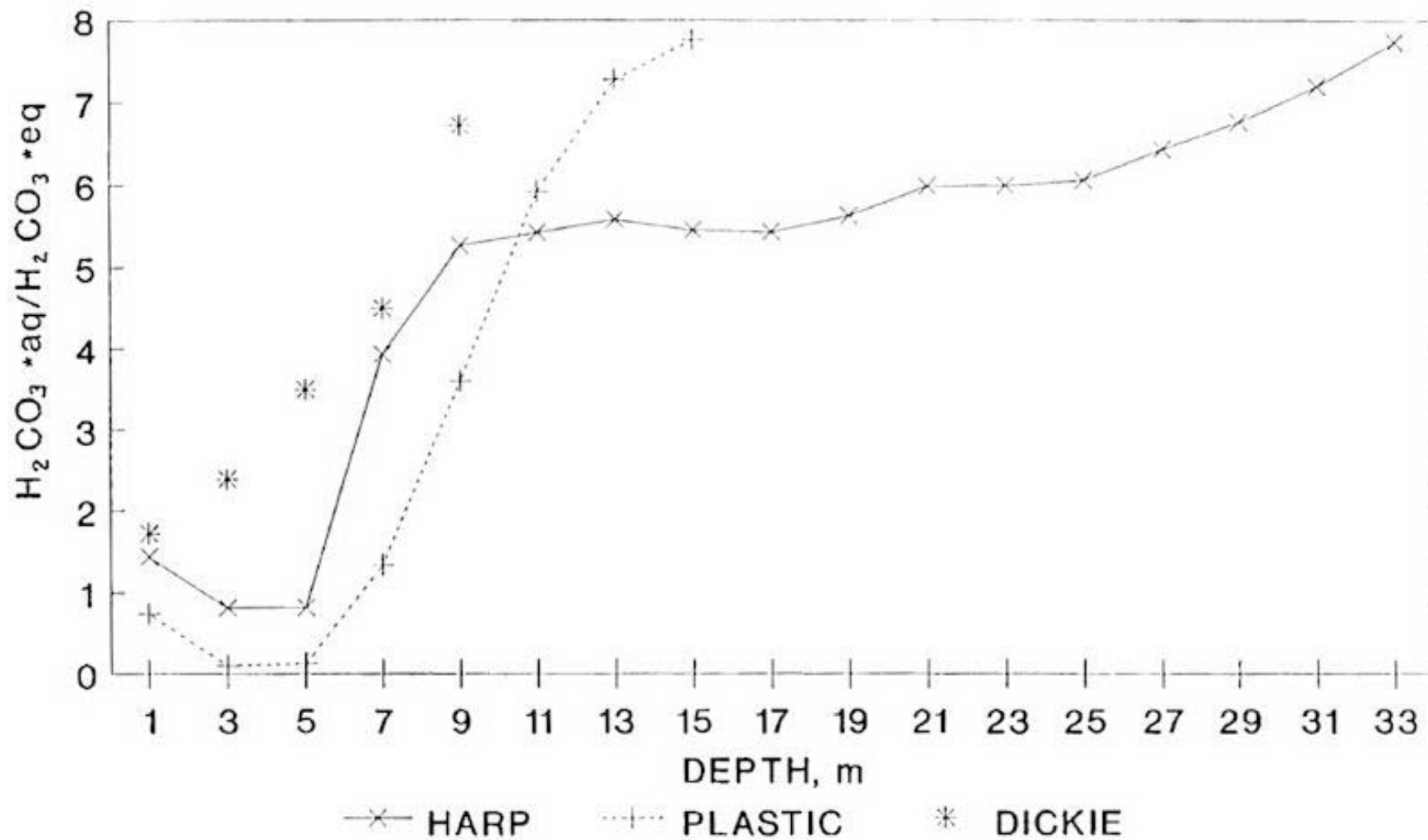


Figure 3

