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Deposition of Atmospheric transported Polychlorinated  
Biphenyls in the Canadian Arctic

By:

D. Gregor, C. Teixeira, R. Rowsell

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## DEPOSITION OF ATMOSPHERICALLY TRANSPORTED POLYCHLORINATED BIPHENYLS IN THE CANADIAN ARCTIC

Gregor, D.<sup>1</sup>, Teixeira, C., Rowsell, R.

Lakes Research Branch, National Water Research Institute, Environment Canada, Burlington Ontario,  
Canada, L7R 4A6;

Keywords: PCBs, arctic, snow, atmospheric transport, deposition

### ABSTRACT

Snow collectors were installed in 1990 at two Canadian high arctic weather stations (Mould Bay {MB} and Eureka {EU}) in an effort to estimate annual deposition of PCBs, compare these estimates to annual snowpack measurements and to investigate the timing of the deposition. The collectors operated successfully but tended to over-collect when two snow fences were used. The daily flux of  $\Sigma$ PCBs in the snowpack for 1990/91 generally compared well to that of the snow collector at MB. The snowpack sample at EU for the same period was considered to be unrepresentative, due to low snow accumulation on the ground and high winds, with resulting low concentrations and fluxes. The congener makeup of snowpack and snow collector samples was similar for both sites. Mean  $\Sigma$ PCB fluxes for the collectors for the winter season were 2.0 and 3.8 ng  $\cdot$  m<sup>-2</sup>  $\cdot$  day<sup>-1</sup> for MB and EU respectively. Due to the tendency of the collectors to overcatch, especially late in the season, the fluxes were corrected relative to the shielded Nipher gauge used to measure snowfall at both weather stations. The corrected PCB fluxes for MB resulted in a reduction of the overall contribution of a high concentration event during the winter, in favour of the fall season, the time of greatest snow accumulation. Initial assessment of the collectors for measuring the timing and composition of PCB deposition to the arctic is encouraging. Work is continuing to improve the collectors and to provide a better estimate of deposition of organic contaminants to the arctic environment.

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<sup>1</sup> To whom reprint requests should be addressed. Current address is: Waterloo Centre for Groundwater Research  
University of Waterloo  
Waterloo, Ontario, Canada  
N2L 3G1

## INTRODUCTION

Since 1986, we have continued to quantify the atmospheric deposition of trace organic contaminants, including PCBs, to the Canadian Arctic in the belief that this is a major pathway for contaminants to the arctic freshwater aquatic ecosystem. There is an increasing concern in the arctic over the potential for ecosystem and human health effects as a result of the bioaccumulation of these contaminants. PCBs are a concern as most of the 209 congeners, especially those with 5 or more chlorines, are biomagnified <sup>1,2,3</sup>. PCBs have been used world-wide for a variety of purposes with as much as 370 tonnes estimated to have been released into the environment <sup>4</sup>.

Studies directed at measuring the deposition of PCBs to temperate regions of North America, especially the Great Lakes region, have generally utilised rainfall only precipitation collectors <sup>5,6,7,8</sup>. Rainfall collectors and collection devices have been utilised around the world including Southern California <sup>9</sup>, Hawaii <sup>10</sup>, Switzerland <sup>11,12,13</sup>, and United Kingdom <sup>14</sup>. Only one published study, that we are aware of, has specifically dealt with the collection of precipitation on a year-round basis, including falling snow <sup>15</sup>. In this case, the collectors were modified to provide a heated collection surface to melt the snow upon contact. Many of the precipitation studies in temperate and sub-polar regions have utilized grab samples of snowpack, generally late in the season, including Strachan and Huneault<sup>5</sup>, Kawamura and Kaplan <sup>9</sup>, Bevenue *et al.* <sup>10</sup>, Czuczwa *et al.* <sup>11</sup>, Lunde *et al.* <sup>16</sup>, Schrimpf *et al.* <sup>17</sup>, Schrimpf <sup>18</sup>, Bobovnikova and Dibtseva<sup>19</sup> and Haglund *et al.* <sup>20</sup>. Studies focussing on PCBs in snow of polar regions have been limited to grab samples of snow pack and include McNeely and Gummer <sup>21</sup>, Paasivirta <sup>22</sup>, and Gregor and Gummer <sup>23</sup> in the arctic and Peel <sup>24</sup> and Tanabe *et al.* <sup>25</sup> in the Antarctic.

The source of atmospheric sulphur at Alert, Canada, has been shown to be primarily Eurasia with the bulk of this transport occurring between October and March <sup>26</sup>. The general atmospheric circulation during the winter season, driven by the strong Siberian anticyclone, flows mainly from the Eurasian continent into the Arctic and then out over the North American continent or into major cyclonic regions in the Aleutians and off southern Greenland. In summer, the south-to north transport is replaced by a weak north-to-south transport as the Siberian high dissipates and air flow into the arctic from the north Pacific and the north Atlantic is more frequent <sup>26</sup>. As a result of the winter time north-to-south flow, PCBs emitted north of the polar front in Eurasia during the winter season, may be transported quickly to the Arctic, where both wet and dry scavenging occurs. While the cold, dry air minimizes wet scavenging as known in temperate, more humid regions, total snowfall between September and May represents fully 80% of the annual precipitation <sup>27</sup>. Further, the stability of the air masses, and the long calm periods with ice crystal deposition interspersed with snowfalls, are

likely effective scavenging mechanisms. The winter season, therefore, is an important period and possibly the dominant period of contaminant delivery to the arctic.

There are clear advantages in collecting freshly fallen snow in the arctic to measure contaminant deposition as opposed to, or in addition to, the collection of bulk snow pack samples. Of particular interest is the timing of the deposition and the relationship to specific atmospheric conditions. Additionally, the weathering of the snowpack during the course of the snow accumulation season may significantly alter the nature of contaminants in the snowpack compared to the original snowfall. On the other hand, while snow blows considerable distances in the arctic, the general absence of local sources of PCBs in the Canadian Arctic allows the use of a geographically distributed snowpack sampling network to represent deposition on a more extensive scale than is possible with collectors.

The use of snow collectors in the arctic is complicated by the extremely cold conditions. For example, a snow crystal in ambient air temperatures of  $-40^{\circ}\text{C}$  landing on a heated snow collector surface of  $4^{\circ}\text{C}$  is somewhat analagous to a raindrop landing on a hot frying pan. Clearly, revolatilization of semi-volatile compounds under circumstances involving a rapid temperature change and a phase change, must be a concern. The effect of wind on scavenging efficiency and scouring of accumulated snow are also concerns. An arctic snowfall collector, among other things, must therefore be capable of trapping sufficient quantities of snowfall for analyses, minimize revolatilization and be reasonably comparable in terms of sampling efficiency with accepted snow gauges.

This work was undertaken in the Canadian Arctic to investigate the feasibility of measuring the concentration of PCBs in fresh snow using a collector, to consider the timing of deposition, to compare total seasonal deposition between a collector and snowpack and to assess the snow scavenging effectiveness of the design.

## METHODS

In the fall of 1990, large area snow collectors ( $4.5\text{ m}^2$ ) were designed and installed at two High Arctic Weather Stations (HAWS) - Mould Bay (MB) ( $76^{\circ}15'\text{N}$ ,  $119^{\circ}16'\text{W}$ ) and Eureka (EU) ( $80^{\circ}00'\text{N}$ ,  $86^{\circ}36'\text{W}$ ) Northwest Territories (NWT) Canada (Figure 1). The collectors, sketched in Figure 2, were intended to intercept ice crystals and falling snow during calm periods but will also trap some quantity of wind blown snow due to the 50% porosity snow fencing that was installed around the perimeter of the collector. The MB collector was installed initially with two sets of fencing with the second fence set 4 m beyond the perimeter of the first and at a height of 1.5 m to

2.7 m. The outer snow fence was added at EU for the 1991/92 field season. Samples were collected approximately once a week, weather conditions permitting. The surface of the collector was aluminum and the samples were sealed into custom built aluminum cases which had been rinsed with ultrapure acetone and hexane. These samples were then stored on site until late winter when they were returned to Resolute Bay, NWT (74°42'N, 94°54'W) where the snow was melted in the sealed cases at room temperature and extracted with dichloromethane (DCM) using a Goulden Large Volume Extractor (GLVE)<sup>23</sup>. The samples were spiked with internal standards during extraction and the extracts were subsequently returned to the analytical laboratory in Burlington, Ontario.

Triplicate and duplicate end of winter, snowpack samples were collected in early May at MB and EU respectively, as part of a NWT snow survey and for purpose of comparison with the snow collector samples. These samples were handled in a manner identical to that used for the collector samples.

Final clean-up and fractionation of these samples followed procedures developed in Environment Canada's National Water Quality Laboratory<sup>28</sup>. In summary, these procedures included the transfer of the sample, after field extraction in the GLVE, to clean glass separatory funnels (1 l) for final separation of the solvent. The DCM was passed through a 5 cm layer of fired Na<sub>2</sub>SO<sub>4</sub> in an Allihn suction funnel. The Na<sub>2</sub>SO<sub>4</sub> was pre-washed with 50 ml DCM. The water remaining in the separatory funnel was twice rinsed with 25 ml of DCM, which was also passed through the Na<sub>2</sub>SO<sub>4</sub> without suction. The Na<sub>2</sub>SO<sub>4</sub> was finally rinsed with 20 ml of iso-octane, which was drained using gentle suction. This mixture was concentrated at 30°C by rotary evaporation to 3-5 ml, and then evaporated to 1 ml under a nitrogen stream. Column chromatography on 3% deactivated silica gel was used to fractionate the extract, with 40 ml of hexane followed by 60 ml of DCM/hexane (1:1 by volume). Iso-octane (10 ml) was added to each fraction, which was again concentrated to a final volume of 1 ml.

A total of 51 PCB congeners were analysed with quantification by a single point calibration using an external standard mix of the target compounds. Analyses were performed on an HP 5890 Gas Chromatograph with dual electron capture detectors (ECD). The two columns used were SE-54 (SPB5) and OV-1 (SPB1). Injection in the splitless mode was at an initial oven temperature of 80°C, held for 2 minutes and then increasing at 4°C per minute to 270°C and held for 12 minutes. The carrier gas was helium at 1.0 ml·min<sup>-1</sup> and the makeup gas was nitrogen at 30 ml·min<sup>-1</sup>. Compounds had to be confirmed on both columns with a retention time tolerance of ±0.02 minutes to be quantified.

## RESULTS

Due to exceptionally low snowfall at EU and some small weekly samples at MB, a number of smaller samples were pooled to provide a larger sample volume. This yielded only four samples for the season for EU and a total of nine samples for MB for the 1990/91 winter season. This experience has shown that the concentrations for most of the compounds of interest were higher in this fresh snow than expected and thus for subsequent work, sample pooling will be minimized.

While the design of these snow collectors followed an internationally accepted design for snow fall measurements<sup>29</sup>, it was observed that at MB where a double snow fence was used, that snow accumulated beyond the outer collector fence on the lee side relative to the prevailing winds. This snow accumulation eventually migrated toward the collector such that the collector was incorporating excessive quantities of blowing snow. The efficiency of the large area snow collectors, for each sampling period, are compared to the measured snowfalls determined by the shielded Nipher Gauge utilized by the weather stations in Figure 3 for both the 1990/91 and 1991/92 seasons. The weather stations report snowfalls of less than 0.1 cm (0.1 mm equivalent precipitation) as "trace" quantities. As our collector was designed to include these light snowfalls, usually resulting from the deposition of snow crystals, we included the trace quantities in the estimation of total precipitation by the Nipher Gauge by assigning a value of 0.05 mm. Previous studies<sup>27,30</sup> have demonstrated that actual snowfall in the arctic may be as much as 130 to 300% greater than that estimated by the weather stations using the Nipher Gauge. EU collected slightly less snow than the Nipher Gauge during the first season in all but one sampling period (Nov 20-26, 1990). In contrast, with the addition of an outer snow fence at EU in 1991, the collector frequently overcaught. The MB collector, having double snow fences in both years, generally overcaught relative to the Nipher gauge, especially during the latter part of the collection period (February and early March) when the snow drift was migrating upon the collector. There was no obvious relationship between the number of days with blowing snow and the amount of over catch. The MB site is more exposed than EU, but EU is typified by winds of higher velocity.

There was considerable variability in the concentration of individual congeners among the snow collector samples. Consequently, the discussion will be restricted to results summarized by congener groups with the congeners comprising each group listed in Table 1. Field method blanks using pre-tested ultra-pure water revealed no interferences with the PCB congeners analyzed.

Mean concentrations by congener group for each sample are presented for MB in Figure 4. EPCB concentrations throughout the sampling season ranged from approximately  $7 \text{ ng} \cdot \text{l}^{-1}$  to more than  $20 \text{ ng} \cdot \text{l}^{-1}$  with a mean concentration of  $14 \text{ ng} \cdot \text{l}^{-1}$ . EPCB concentrations at EU averaged  $23.7$

ng·l<sup>-1</sup>, ranging from 17.6 to 32.5 ng·l<sup>-1</sup> for the four aggregated samples. Quite in contrast are the snowpack samples which averaged 5.7 and 3.1 ng·l<sup>-1</sup> for ΣPCB at MB and EU, respectively.

## DISCUSSION

The median winter concentration of total PCBs (based on an arochlor mixture) in snow at sites in the Canadian Arctic sampled in 1986 and 1987, from approximately 63°N to 81°N, was 0.75 ng·l<sup>-1</sup> with a range of 0.02 to 1.76 ng·l<sup>-1</sup><sup>23</sup>. The mean ΣPCB concentrations measured in three snowpack samples at each of MB and EU in 1990 were from 5 to 8 times higher than the median concentration from the earlier study and at least twice the highest concentration of the 1986/87 study. Median winter total PCB yields for the arctic snowpack were 70 and 100 ng·m<sup>-2</sup> for 1986 and 1987, respectively<sup>26</sup>. The seasonal yields for the MB snowpack in 1990/91 was 967 ng·m<sup>-2</sup>, much higher than the earlier measurements. In contrast, the seasonal yield for the EU snowpack was only 25 ng·m<sup>-2</sup>. These differences must be, in part, attributable to the switch to congener specific analyses in the current study as the timing of the sampling was approximately the same in all years. Annual depositional variability will also be a factor. Neither factor has been investigated but the very low yield measured at EU in 1990/91 is likely due to limited snow accumulation that year.

Fluxes, determined on the basis of the quantity of water comprising each sample from the collector, provide a more appropriate comparison of the snow collector and snowpack than do concentrations. The calculated fluxes for MB for each congener group for the period of October 26, 1990 to March 1, 1991 are presented in Figure 5 for MB. Daily deposition rates of ΣPCB were more uniform than concentrations at about 2 ng·m<sup>-2</sup>·day<sup>-1</sup> except for a period in the middle of January, 1991 when deposition of ΣPCBs increased to nearly 11 ng·m<sup>-2</sup>·day<sup>-1</sup>. This increase was primarily made up of the two, three and four chlorinated congeners. This sample represents the period of January 3 to January 8, 1991 during which time there was a strong wind for several days followed by a period of calm to light winds. Trace quantities of snow were reported for January 3 to 6 and a total of 1.4 cm of snow fell on January 7 and 8. Assuming 100% efficiency for the collector, this snowfall would account for only about 8 l of water as opposed to the 25 l collected. This over-sampling of snow likely indicates collection of wind-blown snow during the high wind event plus possible over-collection of the fresh snow. The flux of ΣPCB for the collector at EU averaged 3.8 ng·m<sup>-2</sup>·day<sup>-1</sup>.

Concentrations in the snowpack were less than 50% of the mean concentrations of the weekly collector samples. However, the calculated daily flux to the MB snowpack exceeded that of the collector (Table 2). Apparently, during the months of March and April when the collector did not

operate, PCB deposition continued but in a less concentrated form, thereby diluting the snowpack. This will have to be confirmed in subsequent investigations. The situation at EU is quite different with both low concentrations and very low flux measured in the snowpack. This is likely more indicative of an unrepresentative snowpack sample in the spring of 1991 due to the exceptionally low annual snowfall and very strong winds which removed most of the snowpack in the area.

PCBs have been shown to be between 80 and 100% in the vapor phase in urban atmospheres and in the area of the Laurentian Great Lakes <sup>31,32,33</sup>. Duinker and Bouchertall <sup>34</sup> investigated the partitioning of several PCB congeners and concluded that while only a very small fraction of total PCB was found in aerosols, this compartment accounted for more than 99% of the PCBs in rain. More highly chlorinated congeners were particle-associated while the lower chlorinated congeners remained essentially in the vapor phase. The partitioning of PCBs between vapor and particles in the atmosphere is inversely related to temperature <sup>35,36</sup>. Consequently, at ambient arctic temperatures, a greater proportion of lower chlorinated PCBs will likely be particle associated than in the temperate source regions. Further, the atmospheric concentrations of higher chlorinated PCBs in the arctic will be less than the lower chlorinated PCBs as the former will be preferentially precipitated during transport <sup>16</sup>. Thus, it is speculated that during the arctic winter, ice crystals, fresh snowfall and dry deposition effectively scavenge the full range of PCB congeners delivered to the arctic.

There are differences between the collector and snowpack samples for concentrations and flux as noted above. A comparison of the relative contribution to  $\Sigma$ PCB by each congener group clearly indicates that the composition of the means of the MB and EU snow collector samples are similar to the snowpack samples for both sites (Figure 6). This suggests that the key to estimating total loadings of PCBs to the arctic is largely a function of obtaining a representative snowpack sample near the end of the accumulation season. Ongoing work at both sites, with sampling representative of comparable deposition periods, will allow the confirmation of this.

The timing of PCB deposition during the winter season is also of interest. As noted above (Figure 4) daily flux at Mould Bay tended to be about  $2 \text{ ng} \cdot \text{m}^{-2} \cdot \text{day}^{-1}$  except for the one event in early January, 1991. However, as the collector generally overcaught, relative to the Nipher gauge, the flux must be corrected for efficiency. The corrected PCB fluxes by congener group are illustrated in Figure 7. This results in a general decrease in the daily flux overall, especially for the January event, but greatly increases the flux during the early part of the season when precipitation is higher due to the greater abundance of open water and the relatively warmer temperatures. The decreased deposition in February and March supports the conclusion reached above that while deposition continued to the snowpack during March and April, the net effect was a dilution of the snowpack concentrations relative to mean concentrations for the snow collector.

## CONCLUSIONS

The simple, large area snow collectors developed for measurements of organic contaminant deposition in the arctic operated effectively during the first two years from a physical stand point. All collectors are now being installed with only a single fence to avoid the excessive accumulation of snow on the lee side and to minimize the frequency of over-catch. This will likely result in more frequent undercatch which can be corrected by using a collector efficiency factor based on the standard Nipher snow gauge. Further evaluation of the relative collection efficiency of these collectors at a variety of sites in Canada's Arctic, characterisitic of wind-swept tundra, lightly treed and mountainous regions, is continuing.

These data represent the first time that a detailed seasonal deposition record has been obtained for PCBs in snow in the arctic. The PCBs are dominated by the lower chlorinated congeners although the higher chlorinated compounds make up a larger portion of  $\Sigma$ PCB as the season progresses. The flux is generally uniform at about  $2 \text{ ng} \cdot \text{m}^{-2} \cdot \text{day}^{-1}$  although some events may have higher fluxes. Daily deposition of PCBs, corrected for overcatch relative to the standard Nipher Gauges, indicates that most of the deposition occurred early in the season when greater snowfall occurred resulting in increased scavenging.

## ACKNOWLEDGEMENTS

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**Table 1** Identification of PCB congener measured in this study comprising each congener group\*

Number of Chlorines	Congeners Analyzed
1	nil
2 & 3	15,18,31
4	40,44,49,52,54,60,
5	86,87,101,103,105,114,118,121
6	128,129,137,138,141,143,151,153 154,156,159
7	170,171,173,180,182,183,185 187,189,191
8, 9 & 10	194,195,196,199,200,201,202,203 205,206,207,208,209

\*Numbering of congeners is based on IUPAC

**Table 2** Comparison between snow collectors and snowpacks for Mould Bay and Eureka of seasonal flux of PCB congener groups ( $\text{ng} \cdot \text{m}^{-2} \cdot \text{day}^{-1}$ )

congener group (number of chlorines)	Mould Bay		Eureka	
	collector	snowpack	collector	snowpack
2 & 3	1.26	1.92	1.55	0.04
4	0.93	1.31	0.63	0.04
5	0.52	0.86	0.58	0.02
6	0.37	0.44	0.51	0.02
7	0.31	0.27	0.25	0.02
8,9 & 10	0.34	0.34	0.26	0.01
$\Sigma$ PCB	3.73	5.14	3.77	0.14

## **FIGURE CAPTIONS**

1. Circumpolar map showing the locations of Mould Bay and Eureka weather stations, Northwest Territories, Canada.
2. Cross section and plan view sketch of the snow samplers used at Mould Bay and Eureka. The samplers are shown here with only the single snow fence.
3. Quantity of snow collected in large area snow collector compared to the standard shielded Nipher snow gauge used by Atmospheric Environment Service, Environment Canada for the the Mould Bay and Eureka weather stations during the 1990/91 and 1991/92 sampling seasons.
4. Concentrations of PCB congener groups compared for the nine samples analyzed from Mould Bay during the 1990/91 season.
5. Flux of PCB congener groups for the nine samples analyzed from Mould Bay for the 1990/91 season.
6. Comparison by PCB congener group for the mean of all samples analyzed for the snow collectors at Mould Bay and Eureka during the 1990/91 season and the mean of the bulk snow pack samples collected at each site in May, 1991.
7. Flux of PCB congener groups for the nine samples analyzed from Mould Bay for the 1990/91 season corrected for snow collection efficiency relative to the Nipher gauge.

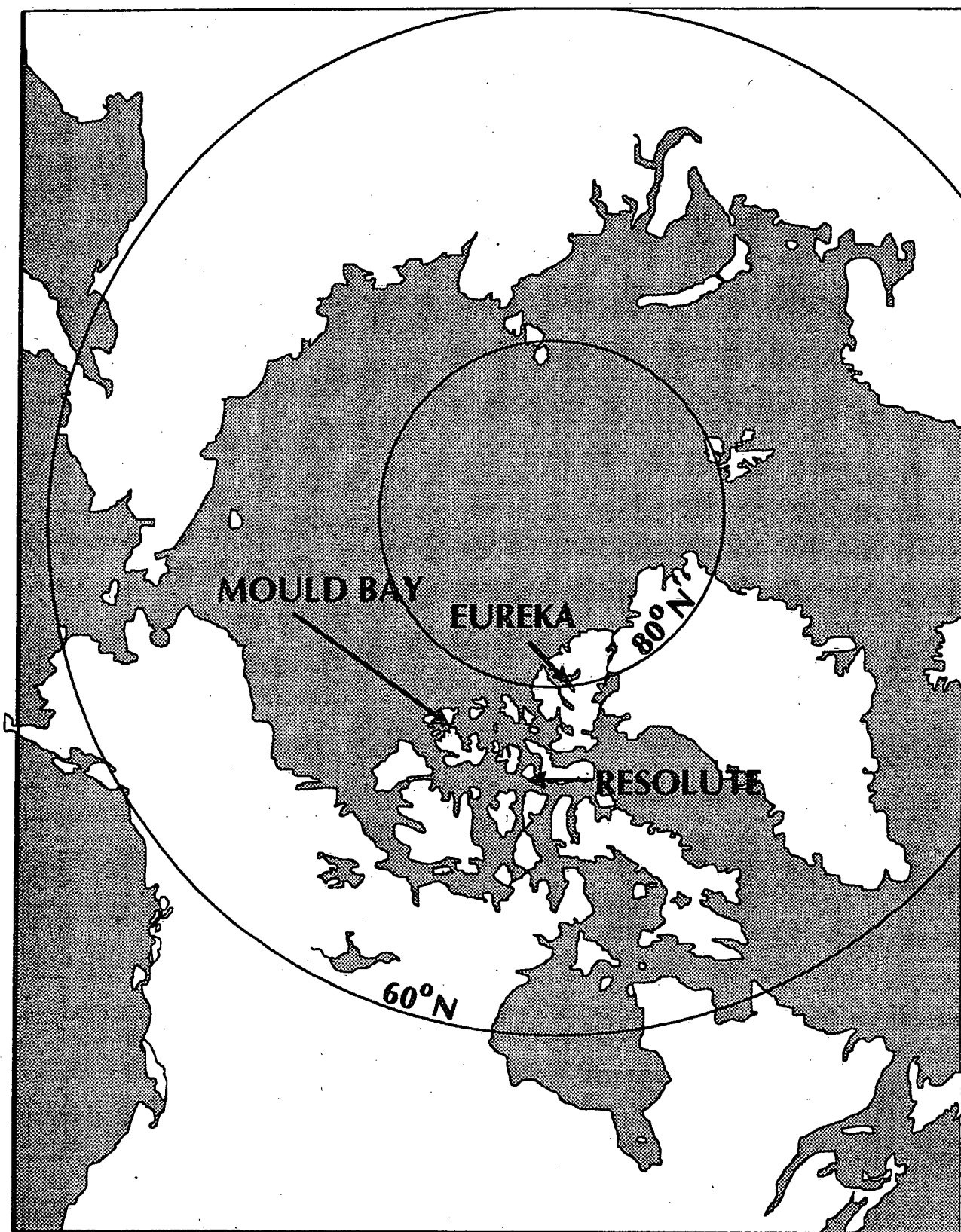


Figure 1

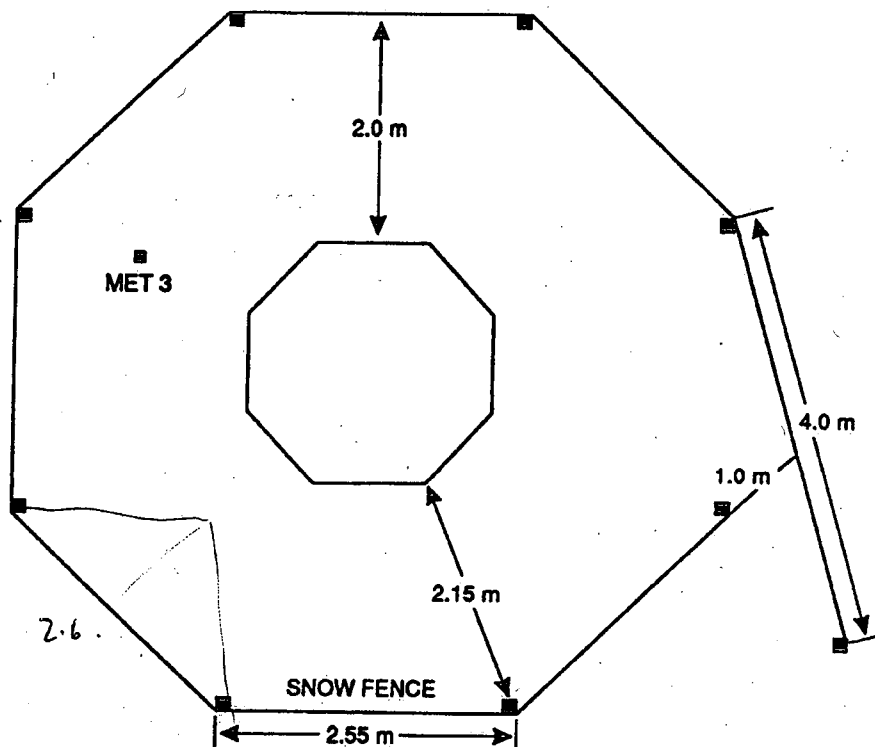
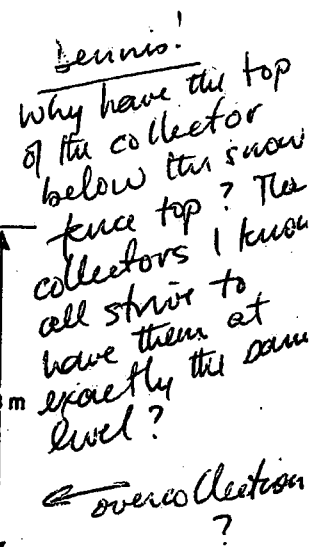


Fig 2.

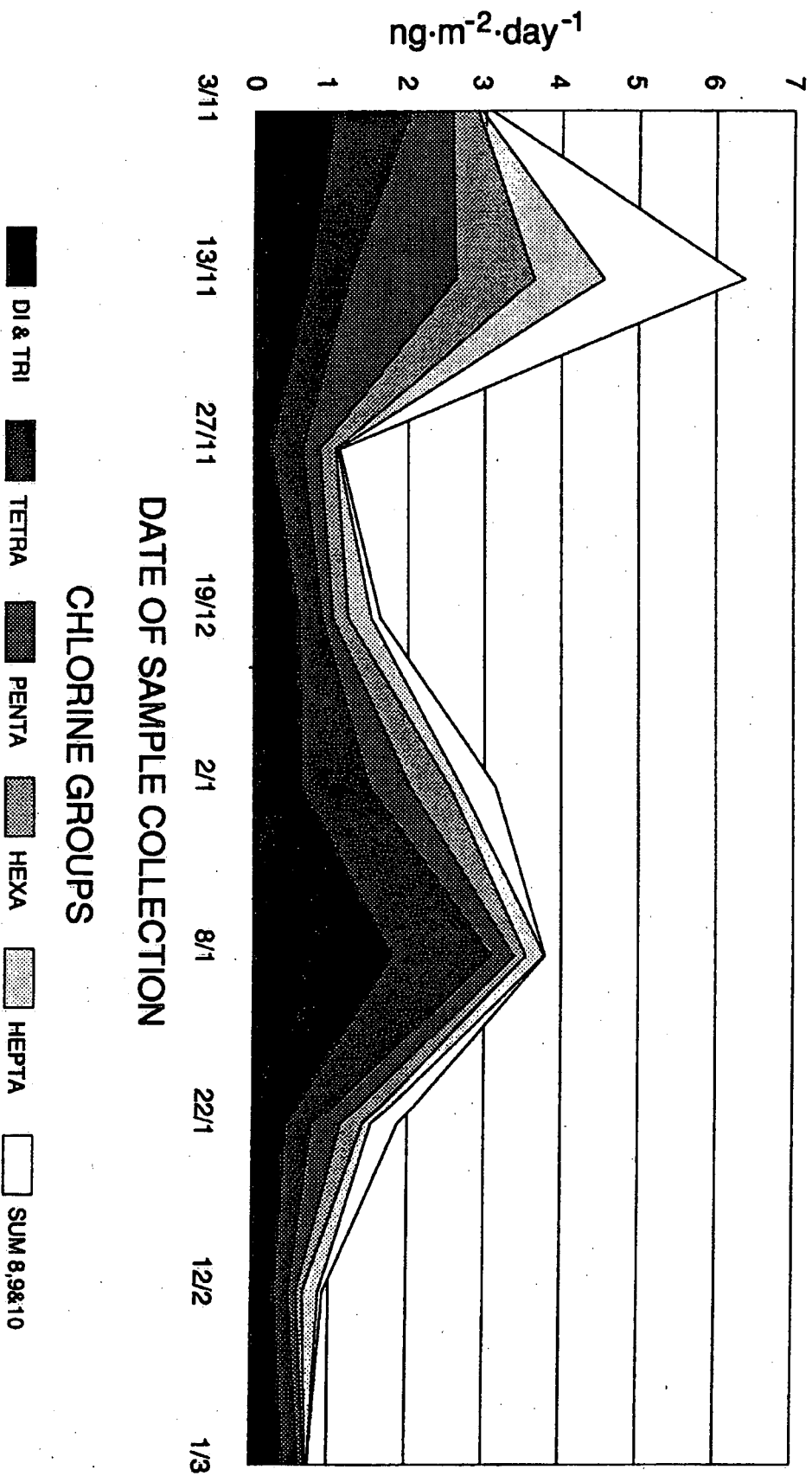


Fig 7

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Burlington, Ontario  
L7R 4A6 Canada

**National Hydrology Research Centre**

11 Innovation Boulevard  
Saskatoon, Saskatchewan  
S7N 3H5 Canada

**St. Lawrence Centre**

105 McGill Street  
Montreal, Quebec  
H2Y 2E7 Canada

**Place Vincent Massey**

351 St. Joseph Boulevard  
Gatineau, Quebec  
K1A 0H3 Canada

**Centre canadien des eaux intérieures**

Case postale 5050  
867, chemin Lakeshore  
Burlington (Ontario)  
L7R 4A6 Canada

**Centre national de recherche en hydrologie**

11, boul. Innovation  
Saskatoon (Saskatchewan)  
S7N 3H5 Canada

**Centre Saint-Laurent**

105, rue McGill  
Montréal (Québec)  
H2Y 2E7 Canada

**Place Vincent-Massey**

351, boul. St-Joseph  
Gatineau (Québec)  
K1A 0H3 Canada