

# Atmospheric transport of organochlorines to the Arctic Ocean

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## ABSTRACT

Chlorinated pesticide residues and PCB's were measured in air vapour and particulate phases, snow, ice and dissolved and particulate seawater fractions collected from the Canadian Ice Island off Axel Heiberg Island (81°N) in 1986. Concentrations of the most abundant compounds (hexachlorocyclohexanes,  $\alpha$  and  $\gamma$ -HCH) were higher in air vapour and snow during May than in August consistent with the inferred seasonal maximum of Arctic haze. Cyclodienes (dieldrin, heptachlor epoxide, *cis* and *trans*-chlordane) and PCB's (quantified as Aroclor 1254) associated with particles in snow and seawater were also present in highest concentrations in late spring. *p,p'*-DDT and *o,p'* and *p,p*-DDE were detected associated with particles in air, snow and seawater but concentrations were below levels for accurate quantification. Concentrations of dissolved HCH's in seawater in the uniformly mixed upper 50 m (4-5 ng l<sup>-1</sup>) decreased by an order-of-magnitude to 280 m depth. There were no significant differences in vertical profiles for the two sampling periods. Similarities in ratios of HCH and chlordane isomers in air, snow and seawater above 100 m imply that these compounds are transferred between environmental compartments without transformations. Lower ratios below 100 m indicate a different source or selective conversion of specific isomers.

## 1. Introduction

Atmospheric transport on regional and global scales is recognized as a major route for the transfer of contaminants to the ocean (GESA MP 1985). The exchanges from the atmosphere to the ocean are important for global budgets of many pollutants, particularly compounds such as semi-volatile organic substances with low vapour pressure like chlorinated pesticides and PCB's. They are released into the atmosphere in the vapour phase and may be transported long distances from continents or the ocean as gases, aerosols or adsorbed to particles (Duce et al., 1983). The lipophilic nature of many of these compounds leads to their accumulation in marine food webs. Woodwell et al. (1967) hypothesized that such residues would eventually accumulate in abyssal sediments. Harding (1986) reviewed

published observations and concluded that most organochlorines occur in the oceanic water column. Lipids in planktonic organisms represent a pool for concentrating and keeping the residues in suspension.

There have been no studies of airborne chlorinated hydrocarbons in offshore areas of the Canadian Arctic although chlorinated pesticides in air have been measured at a number of land-based sites around the Arctic Ocean (Oehme and Mano, 1984; Hoff and Chan, 1986). Northern surveys of organochlorines in snow (Gregor, 1987), tissues of polar bear (Norstrom et al., 1985) and Arctic marine fish (Wong, 1985) have been carried out. The present study extends this data base by providing measurements of chlorinated hydrocarbon pesticides and PCB's in air, snow, ice and seawater samples collected mid-May to early June and late August to early

September in 1986 at a site off the coast of Axel Heiberg Island.

## 2. Methods

### 2.1. Collection of samples

**2.1.1. Air.** The Ice Island used as a sampling platform was located at  $80^{\circ}57.4'N$ ,  $97^{\circ}35.5'W$  during late May and at  $81^{\circ}4.6'N$ ,  $96^{\circ}48.6'W$  in early September 1986. Throughout the summer the Island remained over the shelf in water depths of less than 300 m approximately 30 to 50 km off the north-west coast of Axel Heiberg Island, (Fig. 1). Air samples were collected at sites 3–5 km from the main camp to minimize contamination. Particulates separated from air were collected in a United Metals<sup>®</sup> high volume air sampler containing a teflon coated GFC filter ( $11 \times 8$  cm). The filter clamping gasket, a potential source of organochlorine contamination, was sealed in cleaned aluminum foil, and other parts of the sampler which would come into contact

with the filter were cleaned with hexane. Consecutive samples were collected over 30 to 52 h periods to provide multiple samples. At the end of each sampling period, filters were folded and stored frozen in prebaked aluminium foil pouches.

High volume vapour and particulate samples were collected using a 142 mm stainless steel filter unit containing a glass fiber filter (Gelman<sup>®</sup> AE, prebaked,  $1 \mu\text{m}$  pore diameter) for particles, a pair of poly-urethane foam plugs ( $15 \text{ cm} \times 7.8 \text{ cm}$ ) in a holder to trap organochlorines in the vapour phase, a calibrated flowmeter and high capacity oil-less pump driven by a brushless motor. The sampler was situated upwind of the air pump and was approached from down wind when in use. Collection periods of 30 to 52 h provided samples from air volumes of 200 to  $600 \text{ m}^3$ . After collection, filters were folded and stored in baked aluminum foil pouches. Polyurethane foam plugs were placed in screw capped sample jars stored at  $-20^{\circ}\text{C}$ .

**2.1.2. Snow.** A hexane cleaned aluminum snow

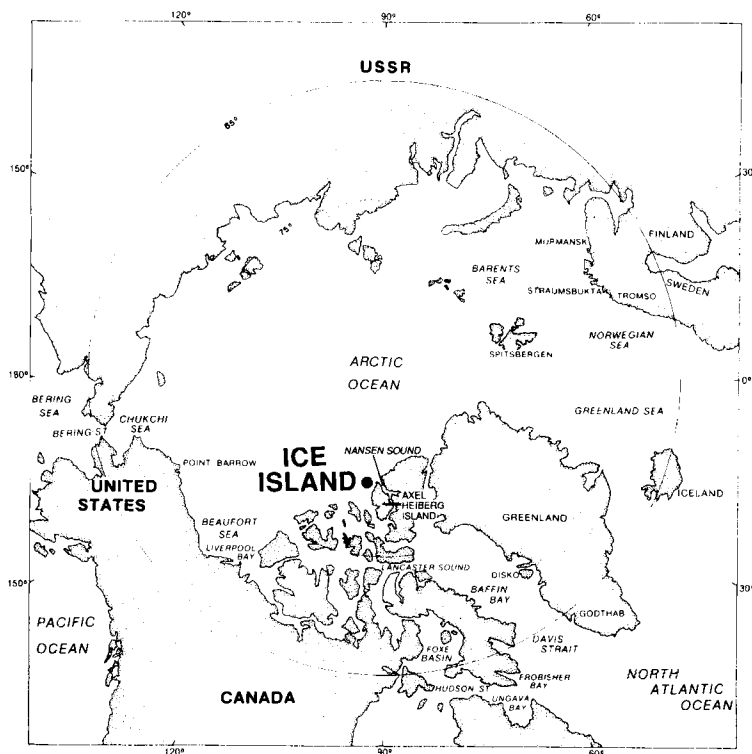


Fig. 1. Location of the Ice Island during sampling in May–June and August–September 1986.

scoop was used to fill a cleaned aluminum melting tank ( $30 \times 35 \times 95$  cm) with surface snow (upper few cm) collected from locations at least 1.5 km from the main camp away from any likely source of contamination. Snow was packed into the tank to increase sample weight. The tank was capped, fitted with a teflon in glass stopcock and the snow sample melted using two 6000 BTU/h camp stoves. To prevent volatilization of organochlorine residues, the temperature of melt water did not exceed  $5^{\circ}\text{C}$ . The procedure was repeated to provide a total liquid volume of 45–65 l for each sample. The melt water was filtered (Gelman AE, 142 mm glass fiber) and extracted by eluting through an XAD-2 resin column at  $100 \text{ ml min}^{-1}$ .

*2.1.3. Sea ice.* Cores of first year sea ice were taken with a hot water ice drill with a 60 cm diameter melting ring. This melter uses a 450,000 BTU oil burner to reheat recirculated water pumped through a heating chamber to a copper ring with evenly spaced holes to allow slow water release. The ice cores, approximately 1.4 m in length, were lifted out and placed on the ice surface. The upper end of each core was trimmed with a carbon steel blade to fit the same aluminum tank used to melt snow. Sections of the core (1.4 m by 0.6 m dia, 350 kg) were cut at the top to fit the tank and a lid secured during transport and melting. Melt water from ice was treated for extraction of organochlorines as described for snow.

*2.1.4. Water and suspended particulate matter.* Chlorinated hydrocarbons and other non-polar components in sea water were extracted in-situ using Seastar<sup>®</sup> water samplers onto Amberlite XAD-2 columns. This sampler manufactured by Seastar Instruments, Sidney, BC, Canada, is a microprocessor controlled battery powered pump which draws water at a preset flow rate ( $150 \text{ ml min}^{-1}$ ) through a filter unit and extraction column, and measures and displays the volume pumped. A glass fiber filter (142 mm Gelman AE,  $1 \mu\text{m}$  nominal pore size) was installed in each filter unit. The XAD-2 column, filter unit and inlet lines were precleaned with pesticide grade hexane.

Seastar samplers were suspended with clean Kevlar rope at various sampling depths. They were passed rapidly through the surface water in melted ice holes or, in August, through an open

lead off the Island. The pump was not activated during deployment to minimize contact with the surface microlayer. After a 10 s delay, samplers automatically started and they were rapidly lowered to the selected depth and allowed to pump 180 to 400 l over 20 to 44 hours. Upon recovery, samplers were rapidly (2–5 min) pulled up from the deployment depth and allowed to pump in air (approximately 1 min) to draw out most of the water present in the filter unit. Filters were removed using solvent cleaned forceps, folded to retain the collected particles, placed into a precleaned aluminum foil pouch, and stored frozen. End fittings of teflon columns containing XAD-2 resin were replaced and columns were stored at ambient temperature until analyses for organochlorines.

An Applied Microsystems CTD-12 was used to measure conductivity and temperature at depth. Conductivity was measured with an equivalent salinity accuracy of  $\pm 0.03$  PPT, temperature to  $\pm 0.02^{\circ}\text{C}$  and depth to  $\pm 0.3$  m. Salinity was calculated from conductivity, temperature and pressure. Dissolved nutrients (reactive silicate and nitrate) were measured in samples drawn in triplicate from 3 l PVC Niskin water bottles and stored frozen in Pyrex glass (nitrate) or plastic (silicate) tubes. Samples were kept frozen until analysis within 2 months. Suspended particles in the water column were collected in situ using Seastar pumps to filter 14 to 133 l through precombusted ( $600^{\circ}\text{C}$ , 3 h) and preweighed silver filters (Selas Flotronics<sup>®</sup>,  $0.8 \mu\text{m}$  pore diameter). Millipore Swinnex<sup>®</sup> holders (45 mm dia), loaded with silver filters, were connected directly to the Seastar sampler intake during the first sampling trip. Since the 4 mm aperture on the holders might not allow particle aggregates to enter, the intake was modified for the August trip. An aluminum cylinder (30 cm high, 4 cm i.d.) open at the upper end was fastened to the pump with the sampler intake at the bottom. Particles, presumably of all sizes of aggregation, entering the cylinder by water exchanges would be susceptible to collection.

## 2.2. Analytical methods

All glassware was cleaned by washing, hexane rinsing and baking ( $340^{\circ}\text{C}$ , 12 h) in a forced air oven. Glass-fiber filters were heated ( $500^{\circ}\text{C}$ , 12 h) and all pesticide grade solvents were dis-

tilled in glass. Solvent blanks were run for each new bottle of solvent before use to check for interference with known organochlorine standards and to determine blanks. Procedural blanks were determined initially and routine procedure blanks were carried through each analysis for every six samples.

**2.2.1. Air vapour-phase.** Polyurethane foam plugs were spiked with internal standard solution (decachlorobiphenyl) then serially extracted by squeezing in an all glass plunger and cylinder system with  $3 \times 150$  ml 1:1 acetone/hexane. The extracts were combined, reduced to 50 ml by rotary evaporation, dried by standing over anhydrous sodium sulphate, reduced to 2 ml and transferred to a 15 ml centrifuge tube for Florisil® column separation.

**2.2.2. Ion exchange columns.** XAD-2 columns were eluted using a system which provided vertical elution upwards from the bottom of a column. Prior to use, the glass and teflon assembly was well cleaned by a series of solvent rinses using acetone (150 ml), hexane (150 ml) and dichloromethane (250 ml). A resin column was installed and eluted at 2 to 4 ml  $\text{min}^{-1}$  with methanol (200 ml) and dichloromethane (250 ml) and the eluate was spiked with the internal standard. The dichloromethane eluate was taken down by rotary evaporation at 35°C to residual methanol, combined with the methanol eluate, to which saturated sodium chloride solution (25 l) was added. The aqueous methanol was extracted with pentane ( $3 \times 100$  ml), the pentane layer separated, taken down to approximately 50 ml by rotary evaporation, dried over anhydrous sodium sulphate for 30 minutes then rotary evaporated to 2 ml. The extract was stored in 15 ml centrifuge tube in a refrigerator before Florisil column separation.

**2.2.3. Filters.** Particulates from air and water on glass fiber filters were extracted for 10 min using a Virtis® homogeniser followed by an ultrasonic bath. Samples were placed in a fluted glass Virtis homogenisation flask or a ground glass stoppered Erlenmeyer flask (for ultrasonic extraction) to which acetonitrile (120 ml) and internal standard was added. The supernatant was decanted through a GF/A filter into 1 l separatory funnel. The filter residue was further serially extracted by the same procedure by homogenisation with 160 ml 3:1 acetonitrile-

water. The residue was rinsed with two additional 20 ml aliquots of acetonitrile. Water content was adjusted to 20% by the addition of pre-extracted water (30 ml) to the separatory funnel, then the aqueous acetonitrile was back-extracted with hexane (150 ml,  $2 \times 100$  ml). The combined extracts were taken down to 50 ml by vacuum rotary evaporation at 35°C dried over anhydrous sodium sulphate (5 g) then taken down to 2 ml. Extracts were transferred to 15 ml centrifuge tubes, sealed and stored in a refrigerator before Florisil column separation.

**2.2.4. Sample clean-up and separation.** Prior to analysis by GC/ECD, samples were passed through a Florisil column (10  $\times$  200 mm) capped with a 10 mm layer of sodium sulphate. Columns were flushed with 50 ml of hexane with a further 37 ml after sample addition. Three fractions (27, 47 and 37 ml) were collected.

**2.2.5. Instrument analysis.** Chlorinated hydrocarbons were determined using a Hewlett Packard 5830A gas chromatograph, with a  $^{63}\text{Ni}$  electron capture detector and a 15 m  $\times$  0.25 mm, 0.25  $\mu\text{m}$  film mm DB5 Durabond Fused Silica capillary column or a 60 m  $\times$  0.22 mm, 0.1  $\mu\text{m}$  film DB-5 Durabond Fused Silica Column (J&W Scientific). A baseline and injection blank (for clean solvents) and a calibration standard (two composite pesticide mixtures and a PCB standard of Aroclor 1254 and/or 1:1 Aroclor 1254/Aroclor 1260) were run at the start of each day. Peaks were assigned for a given chlorinated compound if correct retention time was  $\pm 0.02$  min of that expected by comparison with the standard, if upon co-injection the sample and standard compound co-eluate as one peak, if the compound appeared in the expected Florisil column fraction and if the GC peak shape resembled that of the standard. Assigned peaks were quantitated relative to the internal standard peak using peak heights.

A series of procedural blanks were run for each type of sample to determine detection limits. The mean blank ( $\bar{x}_{bi}$ ) and standard deviation ( $S_{bi}$ ) was calculated for each compound ( $i$ ) within the series of blanks. The mean blank was subtracted from the sample value ( $x_i$ ). If the result ( $x_i - \bar{x}_{bi}$ ) was less than three standard deviations of the blank ( $3S_{bi}$ ), the compound was reported as below detection limit. A result of  $< 10S_{bi}$  but  $> 3S_{bi}$  was reported as present but below the quantification

limit ( $Q$ ). Results were then converted to concentrations and qualified where necessary. No corrections were applied to normalize for differences in organochlorine extraction efficiency on XAD-2 resin columns. Evaluation of retention efficiencies (Green and Kowalski, 1984) showed that PCB's extracted from seawater were retained on XAD-2 columns with 62% efficiency, while values for other organochlorine pesticides such as chlordane, dieldrin and DDT were higher (70–100%).

Silver filters for suspended particulate matter analysis were kept frozen until analysis. Filters were fumed (30 min) over concentrated HCL to remove carbonates prior to drying (60°C overnight). Filters were combusted in a Perkin Elmer Elemental Analyzer (Model 240). Absolute sensitivity for carbon (twice blank value) was approximately 5  $\mu$ g. Dissolved nutrients were analyzed using a Technicon Autoanalyzer II and Technicon Industrial methods. Calibration was with Sagami standards prepared in 30.5% NaCl.

### 3. Results

#### 3.1. Organochlorine residues detected

Four classes of organochlorine compounds (hexachlorocyclohexanes, cyclodienes, isomers of DDT and DDE and congeners of PCB's) were present in our samples (Table 1). The absence of quantifiable concentrations in the solid phases of air, snow, and ice reflect the small amounts of particulate material collected for analysis. The two most common compounds,  $\alpha$  and  $\gamma$  isomers of HCH were present in air vapour and dissolved phases of snow, ice and seawater. HCB was also present in all of these samples except for the dissolved phase of ice melt water. Cyclodienes ( $\alpha$ -endosulphan, chlordane and heptachlor epoxide) were present in air and snow but were relatively minor components or not detected in other samples. Dieldrin was present in vapour in air but it could not be accurately quantified. Isomers of DDT and DDE and congeners of PCB's were measured, but only PCB's associated

Table 1. Chlorinated organic compounds quantitatively detected (+) or present in trace amounts (t) in vapour (V), particulate (P) and dissolved (D) phases of samples collected from the Ice Island during May and August 1986; t indicates traces of compounds that while present were below limits for quantification ( $\times 10$  values in blanks)

Compound	Abbr.	Air		Snow		Ice		Seawater	
		V	P	D	P	D	P	D	P
<i>Hexachlorocyclohexanes</i>									
Hexachlorocyclohexane (isomers)									
$\alpha$	HCH	+	t	+	t	+	t	+	t
$\gamma$	HCH	+	t	+	t	+	t	+	t
$\beta$	HCH	t	t	t	t	t	t	t	t
Hexachlorobenzene	HCB	+	t	+	t	t	t	+	t
<i>Cyclodienes</i>									
Alpha-endosulphan	ENDO	+	t	+	+	t	t	t	t
Chlordane	CHLD	+	t	+	t	t	t	+	t
<i>(cis, trans isomers)</i>									
Dieldrin	DIELD	t	t	+	t	+	t	+	t
Endrin	ENDR	t	t	+	t	t	t	+	t
Heptachlor	HEPTA	t	t	t	t	t	t	t	t
Heptachlor epoxide	HEX	+	t	+	t	t	t	+	t
DDT + DDE									
<i>o,p'</i> DDT, <i>p,p'</i> DDT	DDT	t	t	t	t	t	t	t	t
<i>p,p'</i> DDE	DDE	t	t	t	t	t	t	t	t
<i>Chlorinated biphenyls</i>									
Various congeners quantified as Aroclor 1254	PCB	t	t	+	+	t	t	t	t

with particulate matter in melted snow were determined quantitatively. The lack of detectable concentrations of HCB, chlordane, isomers of DDT and DDE or congeners of PCB's in ice core samples and of DDT and DDE isomers in snow and air, vapour, shows that sample collection or handling did not introduce contamination for these organochlorines.

### 3.2. Changes in concentrations of organochlorines between sampling periods

Higher concentrations of  $\alpha$  and  $\gamma$ -HCH, heptachlor epoxide and *cis* + *trans*-chlordane in air vapour, and snow melt water and PCB's in particulates from snow were observed in May-

June than in August-September (Tables 2, 3). The greatest differences occurred for chlordane and HCB in snow which decreased two orders of magnitude.  $\gamma$ -HCH in air vapour samples decreased more than  $\alpha$ -HCH between sampling periods. The  $\alpha$  isomer of HCH was present in order-of-magnitude higher concentrations than the  $\gamma$  isomer in air vapour and snow melt samples. Although not significantly different ( $p > 0.05$ ) the seasonal trend in concentrations in vapour phase and snow melt water was towards lower values during August-September. PCB's in particulate matter filtered from melted snow also decreased but variance in results was large.

Similar changes in concentrations of hexa-

Table 2. Concentrations of organochlorines mid-May to early June, 1986 in air vapour ( $\text{pg m}^{-3}$ ) dissolved in snow and ice melt water and seawater (average 10 to 60 m) ( $\text{pg l}^{-1}$ ), collected from the Ice Island; mean with standard deviation in parentheses,  $n$  = number of samples, < indicates that concentrations were not greater than the values reported. Abbreviations described in Table 1. Q indicates less than quantification limit.

Sample type	Hexachlorocyclohexanes				Cyclodienes			DDT DDE (various isomers)	PCB's (as Aroclor 1254)
	$\alpha$ -HCH	$\gamma$ -HCH	$\Sigma$ -HCH	HCB	DIELD	HEX	CHLD ( <i>cis</i> + <i>trans</i> )		
Air vapour $n = 6$	451 (116)	70 (28)	521 (143)	73 (18)	<12	4 (1)	3 (2)	2 (1)	<2
Snow $n = 4$	1503 (179)	644 (93)	2148 (256)	33 (10)	292 (104)	107 (29)	91 (42)	20 (10)	125 (58)
Ice $n = 3$	1321 (295)	186 (47)	1507 (342)	<2	6 (1)	3	<2	<2-12	Q23
Seawater $n = 6$	4399 (591)	567 (18)	4966 (477)	17 (6)	15 (2)	<3-11	3.9 (1.6)		7 (5)

Table 3. Concentrations of organochlorines late August to early September, 1986 in air vapour ( $\text{pg m}^{-3}$ ) and dissolved in snow melt water and seawater (average 10 to 60 m) ( $\text{pg l}^{-1}$ ), collected from the Ice Island; mean with standard deviation in parentheses,  $n$  = number of samples, < indicates that concentrations were not greater than values reported. Abbreviations described in Table 1. Q indicates less than quantification limit.

Sample type	Hexachlorocyclohexanes				Cyclodienes			DDT DDE (various isomers)	PCB's (as Aroclor 1254)
	$\alpha$ -HCH	$\gamma$ -HCH	$\Sigma$ -HCH	HCB	DIELD	HEX	CHLD ( <i>cis</i> + <i>trans</i> )		
Air vapour $n = 4$	253 (49)	17 (3)	270 (52)	63 (16)	<6	2 (1)	<1.4	<1	<9 (6)
Snow $n = 3$	1168 (334)	211 (45)	1379 (379)	<2	92 (11)	39 (9)	<1-10	<7	79 (21)
Seawater $n = 5$	4531 (546)	652 (55)	5183 (545)	22 (7)	14 (3)	10 (1)	4.8 (2)	<1	4-Q16

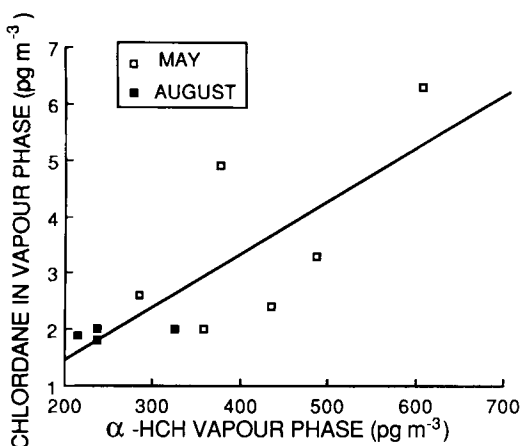


Fig. 2. Relation between concentrations of  $\alpha$ -HCH and *cis* + *trans* chlordane collected at the surface of the Ice Island combining both sampling trips. ( $y = -0.44 + 0.009x$ ,  $r = 0.78$ ).

chlorocyclohexane and cyclodiene classes of compounds in air vapour were observed between the two sampling periods. Chlordane showed a relatively small decrease to levels below the limit of quantitative detection. However, decreased concentrations of chlordane were linearly related to decreases in HCH when data was pooled from all samples (Fig. 2). Average HCB levels in the air vapour phase were constant (63–73  $\text{pg m}^{-3}$ ) during both sampling periods while values in melted snow decreased by an order-of-magnitude. Concentrations of HCH's dissolved in seawater, did not change significantly ( $p > 0.05$ ) between June and August. Average concentrations of chlordane and heptachlor epoxide dissolved in the upper 60 m were higher in August than in June, but differences were not significant due to variability in measurements.

### 3.3. Ratios between organochlorine compounds

Ratios of  $\alpha$ : $\gamma$ -HCH in air vapour and melted snow (2 to 15) were similar to those in seawater (7 to 8) (Table 4). The lowest value in snow (2.3) indicates that precipitation prior to sampling in May carried relatively fresh material (with  $\gamma$ -HCH concentrations 40% of those for  $\alpha$ -HCH) to the Ice Island. With the exception of ratios in the dissolved seawater phase, all values were higher in August indicative of photo-oxidation or conversion of  $\alpha$ -HCH over the sampling period. Concentrations of HCH's associated with particulate matter were generally below the level for reliable quantitative determination but average values for  $\alpha$ -HCH over all depths increased from 0.2  $\text{pg l}^{-1}$  in May to 0.7  $\text{pg l}^{-1}$  in August.  $\gamma$ -HCH concentrations also increased from 0.03 to 0.1  $\text{pg l}^{-1}$  to give a constant ratio of  $\alpha$ : $\gamma$  isomers of 7.4. There was no gradient over depth for either concentrations or these ratios.

The isomers of *cis*- and *trans*-chlordane were present in equal amounts in various samples with ratios between 0.9 and 1.8 in air vapour, snow melt and seawater (Table 4). Ratios of dieldrin:chlordane in these three sample phases were also similar within the range of 1.7–3.3.

### 3.4. Vertical profiles of organochlorines and water column properties

Depth profiles for dissolved  $\alpha$  and  $\gamma$ -HCH, HCB, chlordane, dieldrin and endrin were different. Concentrations of HCH's were maximum in the upper 60 m layer with decreasing values towards greater depths (Fig. 3). The depth-dependent decrease in concentration for HCB was less, with similar concentrations of total HCH. Concentrations below 175 m for both HCH's and HCB were significantly ( $p > 0.05$ ) lower than

Table 4. Ratios of  $\alpha$ HCH: $\gamma$ HCH, *trans*:*cis*-chlordane and *cis* + *trans* chlordane dieldrin in samples collected from the Ice Island in 1986. Values are averages reported in Table 3; dashes indicate no data available or concentrations not sufficient for quantitative determination

Phase	$\alpha$ : $\gamma$ HCH		<i>trans</i> : <i>cis</i> Chlordane		Dieldrin:Chlordane	
	May	August	May	August	May	August
air vapour	6.1	14.9	1.3	—	—	—
snow melt water	2.3	5.5	1.8	—	3.3	—
ice melt water	7.1	—	—	—	—	—
sea water (dissolved) (10–60 m)	7.7	6.9	1.4	0.9	2.5	1.7

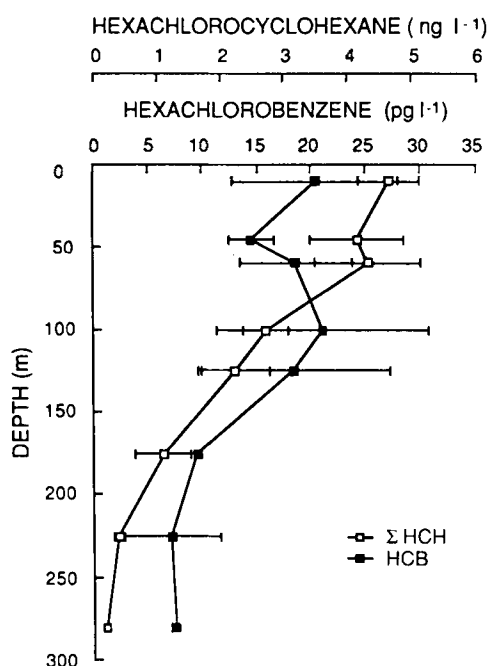


Fig. 3. Vertical profiles of dissolved hexachlorocyclohexane (sum of  $\alpha + \gamma$  isomers) and hexachlorobenzene in seawater collected from the Ice Island during both sampling trips. Horizontal lines represent  $\pm 1$  s.d. of mean values ( $n = 2$  to  $5$ ). Points without lines are single determinations.

surface values. There were no significant differences in profiles between the two sampling periods. Concentrations of both  $\alpha$  and  $\gamma$ -HCH in the upper layer increased between May and August (Tables 2, 3), but the increase was not significant given variation for replicate samples ((standard deviation:mean)  $\times 100 = 11\%$ ). Dissolved chlordane (both isomers) and dieldrin showed much weaker vertical gradients. Although highest values of chlordane occurred at 10 m depth, differences over depth were not significant. There was a general trend, however, towards decreasing concentrations below 125 m (Fig. 4). Concentrations of endrin were close to the level of detection and there was no clear vertical gradient with an average ( $\pm$ s.d.) of  $1.0 \pm 0.4$   $\text{pg l}^{-1}$  for all depths ( $n = 13$ ).

Ratios of concentrations of  $\alpha$  and  $\gamma$ -HCH did not change significantly over depth and average values for the total water column were similar in June and August ( $7 \pm 0.1$ ). Ratios for *trans/cis*-

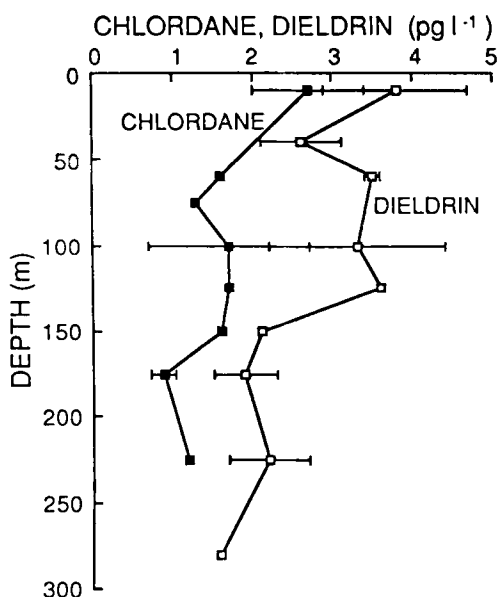


Fig. 4. Vertical profiles of dissolved chlordane (*cis + trans* isomers) and dieldrin in seawater collected from the Ice Island during two sampling trips. Symbols are described in Fig. 3.

chlordane, on the other hand, were higher in the upper 100 m than at greater depths (Fig. 5). There was also a decrease in the ratio of these isomers to lower values at all depths in August. The loss of *trans*-chlordane relative to the *cis* isomer would be expected due to metabolic transformation to oxychlordane (Suprock et al., 1980) but this residue was not monitored in either dissolved or particulate samples from seawater in 1986. Ratios of dieldrin:total chlordane (average  $2.0 \pm 1.5$ ) did not vary systematically with depth or sampling period.

Depth profiles of temperature and salinity were generally similar during both sampling intervals and only the May values are presented (Fig. 6). The upper 45 m was well-mixed with homogeneous values of temperature and salinity. In August, this upper layer was slightly fresher possibly as a result of ice melt and river input during the summer. The profiles show a well-mixed isothermal, isohaline layer over a broad thermocline and halocline separating the cold low salinity surface layer from underlying more saline and warmer Atlantic Water. Dissolved inorganic nitrate and silicate present at concentrations of



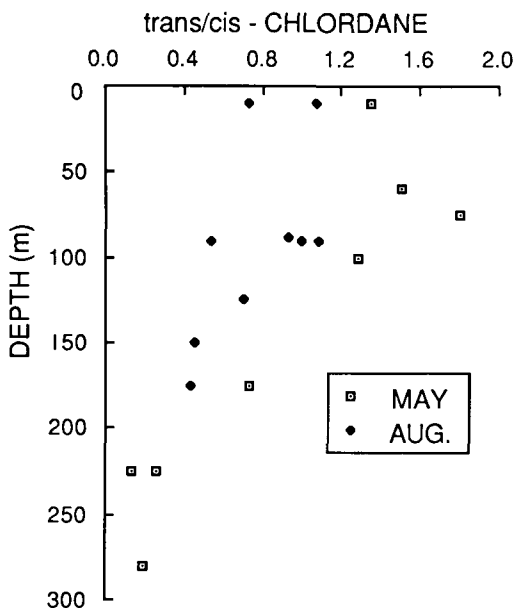


Fig. 5. Vertical profiles of the ratio of *trans/cis*-chlordane dissolved in seawater collected from the Ice Island on two sampling trips in 1986.

6  $\mu\text{M}$  and 15  $\mu\text{M}$  respectively in the upper well-mixed layer increased to a maximum at 90–100 m in water with a salinity near 33.1‰ and a temperature of  $-1.3^\circ\text{C}$ . The maximum occurred at the same depth in May and August. Both silicate and nitrate concentrations were lower in the upper 50 m during August with the greatest decrease to 1 and 6  $\mu\text{M}$  respectively in the upper 5 m. Increased concentrations of suspended particulate organic carbon in August and reduced nutrient concentrations reflect the net increase in suspended particulate organic matter due to primary production by phytoplankton and epontic algae.

## 4. Discussion

### 4.1. Hexachlorocyclohexanes

Hexachlorocyclohexanes quantified in samples collected from the Ice Island are commonly observed atmospheric pollutants in Arctic air masses (Oehme and Stray, 1982; Oehme and Mano, 1984; Oehme and Ottar, 1984), snow (Gregor, 1987) and tissues of Arctic marine mammals and fish (Norstrom et al., 1985; Wong,

1985). Concentrations of  $\alpha$  and  $\gamma$ -HCH measured seasonally in air vapour phase off the northern coast of Norway varied from 160 to 1300 and 10 to 50  $\text{pg m}^{-3}$  respectively with highest levels during summer (Oehme and Mano, 1984). Values over the Ice Island are comparable to the lowest of these concentrations and similar to levels reported by Bidleman et al. (1987) over southern Sweden during winter. However, they are lower by a factor of two than air vapour-phase concentrations measured on the Ice Island simultaneously with our study (Patton et al., 1989) (119–223  $\text{pg m}^{-3}$  and 283–731  $\text{pg m}^{-3}$  for HCB and  $\alpha$ -HCH respectively). Since interlaboratory comparisons of sample extracts showed acceptable agreement the difference is attributed to differences in sampling and flow measurements. In a subsequent intercalibration in June 1987 using a modified air sampling system, differences were less (110–146  $\text{pg m}^{-3}$  and 190–340  $\text{pg m}^{-3}$  for HCB and  $\alpha$ -HCH, respectively). In contrast, Tanabe and Tatsukawa (1980) reported concentrations of total HCH to be approximately an order-of-magnitude greater in air samples from the Northwest Pacific and Bering Sea.

Ratios of  $\alpha$ : $\gamma$ -HCH measured in air off the coast of Norway (12–28) and in southern Sweden (2–6) in these earlier studies span the range of values measured from the Ice Island in May (6.1) and August (14.9) (Table 4). Lower values, due to enrichment of  $\gamma$ -HCH, may indicate supply through long range atmospheric transport to high latitudes during winter when the cold high-pressure center is located over Asia (Oehme and Mano, 1984; Barrie, 1986). We observed lower ratios in snow (2 to 5) which are comparable to values measured by Gregor (1987) for sites throughout the Northwest Territories, including the Ice Island in May 1986. Tanabe and Tatsukawa (1983) described latitudinal differences in proportions of HCH isomers due to an historic tendency for use of purified  $\gamma$ -HCH (lindane) in the southern hemisphere. However, there has been an increasing use of purified lindane in the US since 1978 (Bidleman et al., 1987). Technical mixtures used previously in the northern hemisphere contained approximately 50 to 80%  $\alpha$ -HCH (the inactive and more stable isomer) with 8 to 15% lindane giving a ratio of  $\alpha$ : $\gamma$  5 to 6. The proportions are similar to the ratio (7.4) on suspended particles in our study.

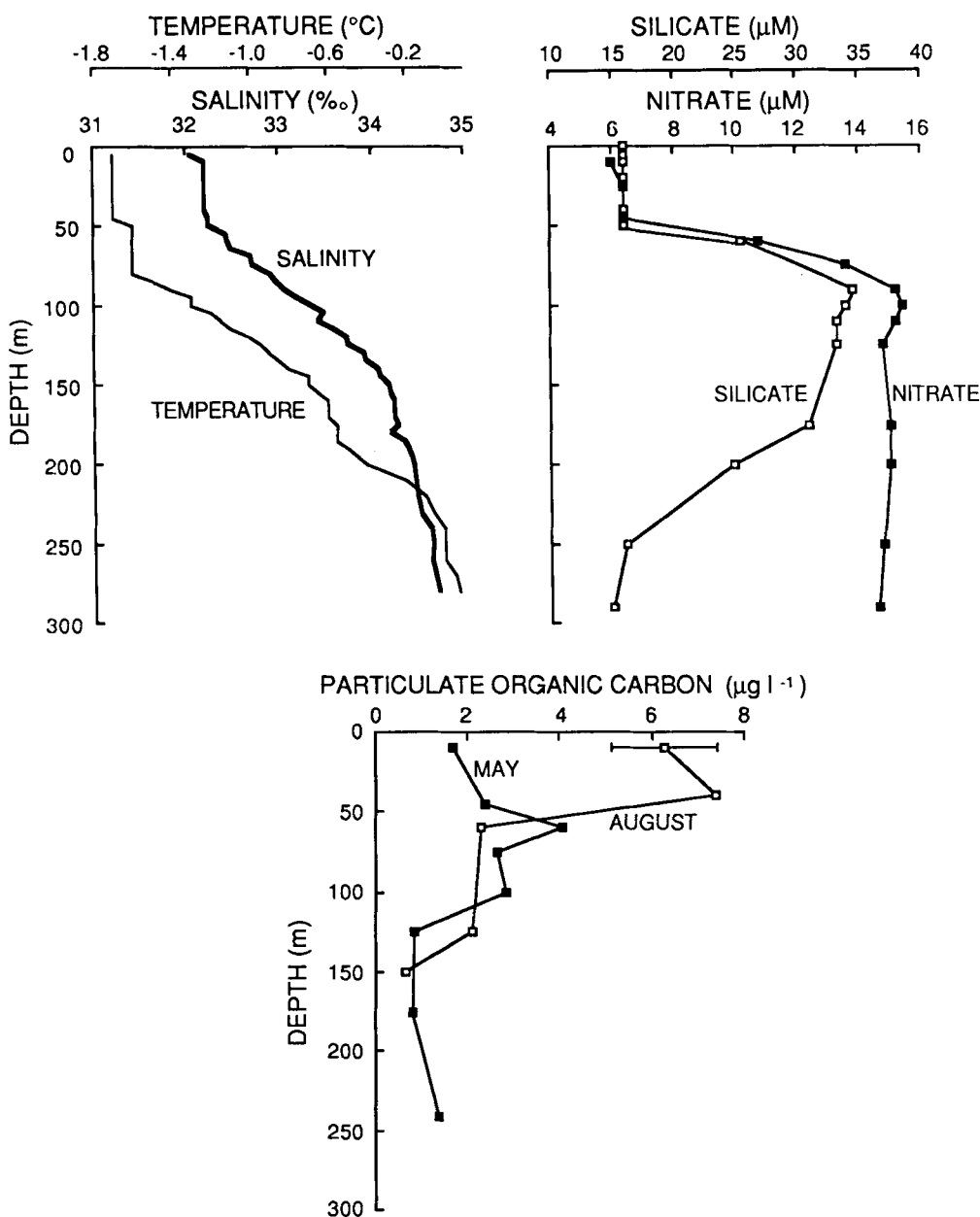


Fig. 6. Vertical profiles of temperature, salinity, dissolved silicate and nitrate during May and particulate organic carbon in seawater collected in May and August 1986 from the Ice Island.

Transformation of lindane to the more stable  $\alpha$  isomer may occur on exposure to UV radiation and in aerated water (Oehme and Mano, 1984). Low ratios (<7) in air vapour in May and snow

during both sampling periods could reflect the transformation of the  $\gamma$  isomer. The ratios for the HCH isomers in seawater did not vary significantly between May and August (Table 4)

perhaps reflecting that the large reservoir of HCH's dissolved in the seawater is relatively stable.

HCb concentrations in air vapour measured over the Ice Island ( $70 \text{ pg m}^{-3}$ ) (Tables 2, 3) are similar to the lowest values reported during summer off the Norwegian Arctic coast (Oehme and Stray, 1982; Oehme and Ottar, 1984) but they are an order-of-magnitude less than concentrations measured in air over the continental US and North Atlantic (Bidleman et al., 1987). Levels measured in air and seawater in our study were similar for the two sampling periods but values in snow decreased by an order-of-magnitude. Although HCB has a vapour pressure which is low compared to many other organochlorines (Bidleman et al., 1986), it is a semi-volatile organic compound. Decreases in concentrations dissolved in snow could arise from volatilization after deposition in the early spring. In general, our values for organochlorine residues in snow are about a factor of two lower than those observed by Gregor (1987) who sampled on the Ice Island three weeks earlier in May 1986. Patton et al. (1989) collected freshly fallen snow on the Ice Island in June 1987 and showed that levels of  $\alpha$ -HCH and  $\gamma$ -HCH were two to six times higher than concentrations present in "old" subsurface snow. Sublimation of previously deposited hexachlorocyclohexanes could have depleted concentrations in snow, especially during summer when air temperatures rose above  $0^\circ\text{C}$ .

The lower water solubility of HCB relative to HCH's (Oehme and Mano, 1984) is reflected in differences in concentrations of these residues in seawater. Vertical profiles of HCH's, with highest concentrations near the surface, are similar to observations reported by Tanabe and Tatsukawa (1983). Levels of HCB dissolved in the upper 60 m were two orders-of-magnitude less than those for the HCH's (Fig. 3). Increased levels of both HCH's and HCB in the surface layer could be due to water exchange from the North Atlantic and Bering Sea and to river input. Concentrations of all hexachlorocyclohexanes measured in ice melt water were lower than those in seawater by up to an order of magnitude (Table 2). Thus some increase in surface layer seawater concentrations could also arise by extrusion on ice formation. On the other hand, dieldrin concentrations in ice were three-fold

higher than in seawater. There is no obvious explanation for this reverse gradient.

Concentrations of HCH measured in coastal and oceanic waters have been reviewed by Harding (1986). Levels of  $\alpha$  and  $\gamma$ -HCH  $> 10 \text{ ng l}^{-1}$  occur in some coastal waters near urban areas but values for oceanic surface waters are an order-of-magnitude lower. Concentrations for  $\Sigma$ -HCH in the NW Pacific and Bering Sea ( $1.3$  and  $7.3 \text{ ng l}^{-1}$ ) span the range of values observed at the Ice Island ( $5.1 \pm 0.5 \text{ ng l}^{-1}$ ) (Tables 2, 3). Concentrations of  $\alpha$ -HCH and  $\gamma$ -HCH of  $7.1$  and  $0.8 \text{ ng l}^{-1}$  reported from 10 m depth in samples collected from the Ice Island in June 1987 (Patton et al., 1989) are about 40% higher than average values for the mixed layer we measured in June 1986 (Table 2). Differences in extraction methods used in each study must account for this variance. Reported values for isomers of DDT and congeners of PCB's in oceanic waters are highly variable but levels in recent studies, where the possibilities for contamination were minimized, are generally between  $0.1$  and  $1.5 \text{ ng l}^{-1}$ . Lowest concentrations occur in the Bering and Antarctic Seas ( $< 0.06 \text{ ng l}^{-1}$  for  $\Sigma$  DDT and PCB's). Concentrations of these compounds associated with particles separated from seawater under the Ice Island were an order-of-magnitude lower (Tables 2, 3).

#### 4.2. Cyclodienes

The principal cyclodiene insecticides measured in air over the Ice Island were present at a few  $\text{pg m}^{-3}$  typical of atmospheric samples from oceanic areas (Tanabe and Tatsukawa, 1980; Kawano et al., 1985). *Trans* and *cis*-chlordane in air vapour during summer at  $76^\circ\text{N}$  at a land-based sampling site (Mould Bay) on Melville Island (Hoff and Chan, 1986) were present in concentrations comparable to our values. Oehme and Mano (1984) measured similar amounts of *cis*-chlordane in air vapour during late summer off the Arctic coast of Norway, but concentrations at more southern latitudes in both hemispheres summarized by Kawano et al. (1985) are an order-of-magnitude higher. Bidleman et al. (1987) reported high chlordane concentrations ( $140 \text{ pg m}^{-3}$ ) over the North Atlantic at  $32^\circ\text{N}$  with lower values ( $31 \text{ pg m}^{-3}$ ) at  $45^\circ\text{N}$  and in Barbados ( $8 \text{ pg m}^{-3}$ ) implying that the US is a major source for chlordane to the atmosphere over the North Atlantic. As noted for hexachlorocyclohexanes in

air vapour, Patton et al. (1989) measured average concentrations of *cis* + *trans* chlordane in August 1986 that were approximately twice levels we reported ( $3.9$  versus  $1.4$   $\text{pg m}^{-3}$ ) (Table 3). In a subsequent intercalibration in June 1987, mean concentrations were comparable ( $5.1$   $\text{pg m}^{-3}$  in our study versus  $6.3$   $\text{pg m}^{-3}$  by Patton et al., 1989) and higher than in 1986.

Hoff and Chan (1986) reported that the ratio of *trans*:*cis*-chlordane for technical chlordane is 1.26. Ratios of 1.3 and 1.8 in air vapour and snow collected in our study and also observed in snow from the Ice Island (1.0 to 1.6) by Gregor (1987) are similar to ratios derived from air samples over southern Sweden (Bidleman et al., 1987). Lower ratios (0.3–0.7) measured by Hoff and Chan (1986) at Mould Bay and by Patton et al. (1989) on the Ice Island in August 1986 and June 1987 are in the range of values (0.2–0.7) we observed in the water column below 100 m depth (Fig. 5). The chlordane isomers dissolved in seawater below 100 m could have a different origin from those in the air, snow and upper water column or the composition of isomers could reflect the metabolic transformation of the *trans* isomer to oxychlordane described by Suprock et al. (1980). Low ratios below 100 m could also reflect residual chlordane not adsorbed by sinking particles or accumulated by planktonic organisms. Zitko (1978) and Kawano et al. (1985) found that fish and seal tissues were depleted of *trans*-chlordane relative to the *cis* isomer.

Significant decreases in concentrations of all cyclodienes occurred in snow collected from the Ice Island between May and August with the greatest difference observed for *cis* + *trans*-chlordane ( $91 \pm 42$   $\text{pg l}^{-1}$  to  $<1$   $\text{pg l}^{-1}$ ). Decreases also occurred in air vapour concentrations but these were much reduced in comparison. Average concentrations of chlordane dissolved in seawater in the upper 60 m increased in August but differences were not significant because of variability in measurements.

The positive linear relation between  $\alpha$ -HCH and chlordane in the vapour phase (Fig. 2) could indicate that changes in atmospheric supply during summer affected both compounds similarly. However, the estimated vapour pressures ( $20^\circ\text{C}$ ) for chlordane isomers ( $2.9$ – $3.0 \times 10^{-3}$  Pa) are an order-of-magnitude lower than values for  $\alpha$ -HCH ( $8.4 \times 10^{-2}$  Pa) Bidleman et al., 1986). Other cyclodienes such as dieldrin have vapour pres-

ures similar to chlordane (T. Bidleman, personal communication). Since these compounds are all less volatile than  $\alpha$ -HCH, they might be expected to show less seasonal variation. The linear correlation observed in Fig. 2 could also only reflect seasonal changes in supply to the Arctic air mass if removal rates from the atmosphere were similar for both compounds.

The lower concentrations of cyclodienes relative to hexachlorocyclohexanes in the vapour phase is reflected in their reduced concentrations in seawater (three orders-of-magnitude less than those for  $\Sigma$ -HCH) (Table 2, 3). Profiles of cyclodienes dissolved in seawater also showed small vertical gradients (Fig. 4). Dissolved concentrations of chlordane and dieldrin between 50 and 100 m were not significantly different despite sharp gradients in salinity, temperature and dissolved nutrients (Fig. 6). On the other hand, despite variability in concentration profiles, the ratios of *trans*-chlordane relative to the *cis* isomer were increased at depths near 100 m (Fig. 5). Recent studies have suggested that the widespread occurrence of the nutrient maximum near this depth in the Arctic Ocean is not due to intrusion of Bering Sea water, as previously thought, but due to brine formation and drainage from the large continental shelves into the central basin (Melling and Lewis, 1982; Jones and Anderson, 1986; Macdonald et al., 1987). Water in contact with the continental shelves in the Arctic Ocean could increase in dissolved nutrient concentrations through regeneration from deposited particulate matter. Organochlorines in this water mass could also reflect input from coastal drainage and atmospheric exchange during the summer when coastal waters are ice free.

#### 4.3. DDT, DDE and PCB's

Organochlorines such as isomers of DDT and DDE and congeners of PCB's have volatilization transfer coefficients from water to air that are two to three times greater than  $\alpha$  and  $\gamma$ -HCH (Oehme and Mano, 1984). These organochlorines may be released back into the atmosphere following precipitation in snow as described for HCB. This would explain the concentrations of PCB's found in snow in the Ice Island during August that were one-third of values in May.

High concentrations ( $1.7$   $\text{ng l}^{-1}$ ) of total PCB reported for snow collected from the Ice Island in

May 1986 by Gregor (1987) are inconsistent with our values ( $17\text{--}54 \mu\text{g l}^{-1}$ ) (Tables 2, 3). Although differences in extraction techniques preclude direct comparison, the discrepancy could arise from contamination. Gregor collected snow three weeks earlier at a site on the Ice Island close to the base camp and landing strip. Teflon bags were used to contain water from melted snow prior to solvent extraction without separation of particulate and dissolved phases as in our study. This extraction method may be more efficient than we achieved using XAD-2 resin columns. However, blanks from the teflon bags showed that PCB's were present (Gregor, 1987, Table 9) and sample values were not corrected for this contamination. There was no contamination in our samples since concentrations for many organochlorines were below detection levels (Table 1). Other studies of organochlorine distribution in oceanic environments have commented on problems of contamination. McNeeley and Gummer (1984) concluded that PCB's measured in snow from sites on Ellesmere Island were unreliable due to contamination. Harding (1986) pointed out that reported values for PCB's and isomers of DDT dissolved in seawater may be vastly overestimated due to their hydrophobic nature.

The presence of detectable concentrations of organochlorines such as DDT, DDE and PCB's in seawater is greatly affected by the availability of suspended particles. All isomers and congeners of these compounds have estimated water solubilities of  $10^{-8}$  to  $10^{-10} \text{ g l}^{-1}$ , two to three orders-of-magnitude lower than values for HCH isomers of  $10^{-5}$  to  $10^{-7}$  (Tanabe and Tatsukawa, 1983). These authors observed that a high percentage of DDT and PCB's in seawater was adsorbed to suspended particles at high latitudes. Concentrations in vertical profiles were highest where concentrations of suspended particles were also greatest.

Concentrations of suspended particulate matter under the Ice Island are as low as those observed in the deep sea. An average (upper 100 m) value for suspended particulate organic carbon ( $9 \mu\text{g C l}^{-1}$ ) measured by Gordon and Cranford (1985) in samples collected by 12 l Niskin bottles at a station over the Alpha Ridge ( $85^\circ\text{N}$ ) was higher than concentrations in our study (average values of  $2.7$  and  $5.7 \mu\text{g C l}^{-1}$  in

the upper 60 m layer in May and August) (Fig. 5). We used the same type of silver filter and procedure for elemental analysis as was used in this earlier study. Mil'nikow and Pavlov (1978) showed that seasonal maxima ( $25\text{--}45 \mu\text{g C l}^{-1}$ ) occurred in the upper 50 m at  $84^\circ\text{N}$  in the Arctic Ocean between June and August. Values during the remainder of the year and at deeper depths ( $5\text{--}10 \mu\text{g C l}^{-1}$ ) were similar to data reported by Gordon and Cranford (1985).

Low concentrations of suspended particulate matter at all depths, but especially below the upper mixed layer, in the Arctic Ocean should lead to low rates of sedimentation and increased residence time of particle-reactive organochlorines in the water column. The relatively uniform distribution of DDT isomers and congeners of PCB's over depth in oligotrophic oceans (Tanabe and Tatsukawa, 1983) have been attributed to the low solubility of these compounds and removal from the water column by settling particles. These authors estimated a much shorter residence time for DDT and PCB's (0.2–0.4 y) in the mixed layer than for the more soluble HCH's (5–10 y). Organochlorines adsorbed to suspended particles are available for consumption by grazing zooplankton, and rapid removal from the water column can occur as aggregated particles sediment (Osterroht and Smetacek, 1980; Iseki et al., 1981). The impact of these biological processes for transfer of organochlorines through the water column may be enhanced by the low concentration of suspended particles and low sedimentation rates in the Arctic Ocean.

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