

Fluxes of semivolatile organochlorine compounds in Bow Lake, a high-altitude, glacier-fed, subalpine lake in the Canadian Rocky Mountains

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Abstract

The fluxes of several organochlorine compounds (OCs) to and from a high-altitude lake were calculated to determine the relative contributions of rainfall, snowfall, runoff, gas absorption and volatilization, sedimentation, and outflow. Runoff inputs, particularly from a glaciated catchment, and gas absorption were major sources of many OCs. Losses by volatilization, outflow, and sedimentation were also important transport pathways for most OCs. We observed a net deposition of hexachlorocyclohexanes (HCHs) and DDT to Bow Lake by gas exchange, whereas hexachlorobenzene, chlordanes, and most polychlorinated biphenyls were seen to volatilize to the air. The insecticide lindane (γ HCH), widely applied across Canada as a seed dressing in early spring, was absorbed by Bow Lake in early summer but gradually equilibrated with water as the summer progressed. This was the result of lower concentrations of γ HCH in air and rising air temperatures in late summer. Results indicate that glacially derived tributary inputs are dominant sources of persistent organic pollutants to these glacier-fed mountain lakes and that smaller contributions may derive from air-water gas exchange. This study highlights the importance of glacial sources of OCs to mountain lakes and corroborates recent evidence of OC contamination in glacially derived aquatic systems.

Several organochlorine compounds (OCs) such as hexachlorocyclohexanes (HCHs), hexachlorobenzene (HCB), toxaphene, and polychlorinated biphenyls (PCBs) have contaminated alpine environments by atmospheric transport (e.g., Blais et al. 1998; Donald et al. 1998). Donald et al. (1998) showed that fish from alpine and subalpine lakes had concentrations of toxaphene that were two orders of magnitude higher than those in fish from nearby lakes at lower

altitudes, which indicates that there were processes acting to concentrate these semivolatile OCs at high altitudes. In previous publications (Blais et al. 1998, 2001; Donald et al. 1998), we have shown that high precipitation, low temperatures, and glacial runoff contribute to elevated concentrations of OCs at high altitudes. One of the likely reasons why OCs are concentrated in alpine and subalpine environments is that these compounds are distilled from warmer source regions and tend to accumulate in colder areas (Wania and Mackay 1993, 1996).

Atmospheric deposition and air-water gas exchange are important vectors for transport of these chemicals to surface waters. Gas absorption can be an important source of some pollutants to lakes (e.g., HCH in Green Bay; McConnell et al. 1996), and net volatilization of other compounds can be a dominant loss mechanism (e.g., HCB, many PCBs; Jeremiasson et al. 1994, 1999; Jantunen and Bidleman 1995; McConnell et al. 1996). Jeremiasson et al. (1999) showed that ~40% of PCB inputs during the ice-free season were

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lost by degassing at the air-water interfaces of two remote lakes in northwestern Ontario. Likewise, they showed that annual losses of PCBs by volatilization from Lake Superior were equal to >600% of total annual inputs by wet and dry precipitation and tributaries (Jeremiasson et al. 1994). PCB concentrations in the water column of Lake Superior decreased over 14 yr, largely as a result of their volatilization (Jeremiasson et al. 1994).

In addition to direct exchange with the atmosphere and precipitation inputs, tributary streams are also considered to be important vectors of OCs to surface waters, particularly during high flow events after heavy rainfalls (Bremle and Larsson 1997) and during snowmelt in spring (Semkin 1996; Wania et al. 1999). Jeremiasson et al. (1999) proposed that watershed contributions of PCBs were likely an important contributor to the PCB budgets of two small boreal lakes, although tributary inputs were not measured directly because of an absence of defined channelized inflows to these lakes. We have shown (Blais et al. 2001) that glaciers act as effective funnels for persistent organic pollutants, likely because glacial runoff in summer becomes channelized on the glacier surface and is routed rapidly through englacial and subglacial conduits. Our data suggested that the cold glacial meltwater has limited contact with the atmosphere and with catchment soils and sediments, leaving little opportunity for losses of OCs by volatilization and complexation to organic-poor glacial sediments (Blais et al. 2001). These circumstances, coupled with low temperatures and high precipitation, which result in heavy runoff from rocky, organic-poor catchments above the tree line, may result in alpine and subalpine lakes being more contaminated with these chemicals than previously thought.

Recent studies have shown elevated levels of several organochlorines (including toxaphene, PCBs, and DDT) in fish from glacial-fed Bow Lake in Banff National Park, Alberta (Donald 1993; Donald et al. 1998; Campbell et al. 2000). In 1998, we measured tributary inflows, losses by sedimentation, and gas exchange rates of several insecticides (HCHs, HCB, dieldrin, endosulfan, and DDT and its metabolites) and PCBs at Bow Lake to determine and quantify the prominent transport pathways for OCs in a mountain environment. Samples for air and water concentrations were collected concurrently with meteorological information to calculate rates of air-water exchange from the lake's surface. These data were compiled to construct a mass balance for Bow Lake.

Materials and methods

Study site—Bow Lake in Banff National Park, Alberta, Canada, is a subalpine lake located at the head of the Bow River Valley, near the Continental Divide, which stretches along Alberta's border with British Columbia (Fig. 1). It has a surface area of 2.8 km² and a total catchment area of 63.2 km², 18% of which is glacier covered. Although the lake supports a lodge on one side, their effective waste disposal and secondary sewage treatment under park authority make it an unlikely source for persistent organic pollutants to the lake. Biannual coliform testing by the Park Service show that fecal

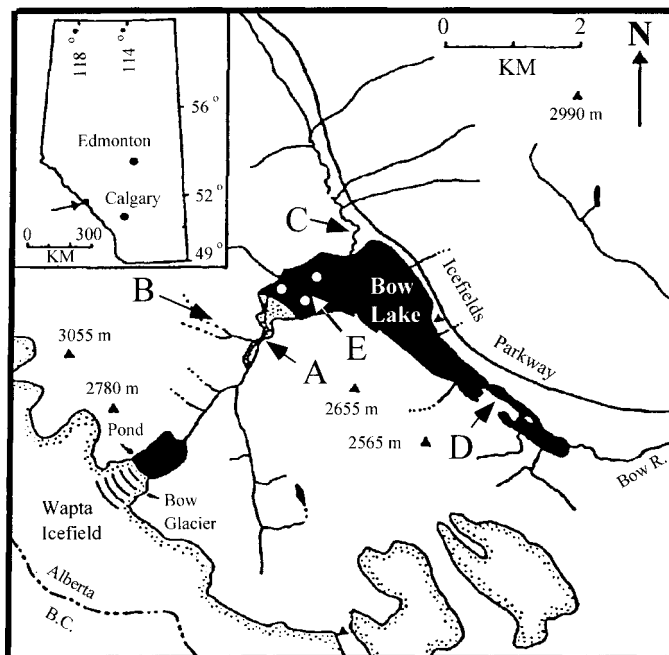


Fig. 1. The Bow Lake field study area, showing the locations of the four stream monitoring stations: (A) the glacial stream, (B) the alpine stream, (C) the subalpine valley stream, and (D) the lake's outflow. (E) The location of the three sediment traps employed in the summer of 1998.

coliforms in Bow Lake near the lodge are generally low (Charlie Pacas, Banff Warden Office, pers. comm.). The primary inflow to the lake is a glacial stream (marked "A" in Fig. 1) that is fed by meltwater from the Wapta Icefield. The stream catchment area is 27 km², ~41% of which is glaciated (11 km²). The lake is also fed by a nonglacial alpine stream (marked "B") with a catchment area of 1.6 km², and another stream that passes through a forested subalpine catchment (marked "C") with a basin area of 17.3 km². The lake's outflow ("D") was also sampled.

Air sampling—Organochlorines in the vapor phase were measured in the air on the shore of Bow Lake twice a month on average during the summer of 1998 by use of a Graseby (GMW High-Vol) air sampler. Air sampling involved extracting contaminants from an average of 2,950 ± 1,100 m³ of air. Aerosols were collected on preashed glass-fiber filters (GF/F 0.7 μm pore size), and vapors were extracted on polyurethane foam (PUF) plugs that were pre-extracted by Soxhlet for 6 h in acetone and hexane. PUFs and GF/F filters were collected after sampling and transported to the National Laboratory for Environmental Testing in Burlington, Ontario, for chemical analysis. PUFs were transported in the sealed cartridges in which they were deployed in the air sampler, thus minimizing external handling. A weather station on the shore of Bow Lake collected hourly data for air temperature, wind speed and direction, barometric pressure, and relative humidity.

Water sampling—The three streams draining into Bow Lake and the lake's outflow were sampled twice a month

during the summer of 1998. Water was collected in four 18-liter air-tight aluminum cans that were prerinsed with acetone and hexane. A water sample for dissolved organic carbon analysis (DOC) was also collected in a polyethylene bottle and analyzed on an Ionics Corporation 1555 DOC analyzer with 100% Pt catalyst. Concurrent air sampling allowed us to calculate gas exchange rates using the two-film resistance model (Schwarzenbach et al. 1993). Water samples taken from the lake's outflow were considered to be representative of the lake's epilimnion and used in all air-water flux calculations. Mean hourly water temperatures and discharges in the streams were monitored by use of thermistors and pressure transducers feeding information to CR-10X data loggers (Campbell Scientific). Stream discharge was determined by calibrating stream water level to total discharge once every 2 weeks by use of the velocity-area technique.

Organic analysis—Stream water samples were filtered through precombusted Gelman GF/A filters (pore size 0.6 μm), and at least 65 liters per sample were extracted into dichloromethane by use of a high volume Goulden extractor (Goulden and Anthony 1985). Extraction efficiencies were determined with internal standards (aldrin, 1,3-dibromobenzene, 1,3,5-tribromobenzene, 1,2,4,5-tetrabromobenzene, δHCH , endrin ketone). Organochlorine concentrations were corrected to account for recovery efficiencies by use of δHCH and endrin ketone. Concentrations of organochlorine analytes in water were adjusted to account for binding of organochlorines to DOC colloids that passed through the glass-fiber filter. Sorption of OCs to DOC colloids is defined by a colloid-water partition coefficient (K_c), which relates to the organic carbon-water partition coefficient (K_{oc}) as follows (Landrum et al. 1984; Jeremiasson et al. 1999):

$$K_c = 0.2K_{oc}. \quad (1)$$

The K_{oc} is calculated from the octanol-water partition coefficient (K_{ow}), as follows (Karickhoff et al. 1979):

$$\log K_{oc} = 1.00 \log K_{ow} - 0.21. \quad (2)$$

The measured dissolved concentration of the organochlorine analytes ($C_{w \text{ meas}}$) relates to the actual dissolved concentration ($C_{w \text{ actual}}$), K_c , and DOC concentration (kg L^{-1}), as follows:

$$C_{w \text{ actual}} = C_{w \text{ meas}} / (1 + K_c \text{ DOC}). \quad (3)$$

$C_{w \text{ actual}}$ was used as the concentration in water (C_w) for calculating air-water gas exchange in the following section. Although this correction had a negligible impact on dissolved concentrations of OC pesticides, it reduced the $C_{w \text{ actual}}$ of penta-decachlorobiphenyls by 6%–20% in Bow Lake.

The air samples (on PUF filters) were Soxhlet extracted into 1:1 acetone:hexane. Extracts from air and water samples were concentrated by roto-evaporation and cleaned with activated silica, then solvent exchanged into iso-octane (Muir et al. 1995; Blais et al. 1998). Final extracts were evaporated with ultra-high-purity nitrogen to 200 μl final volume.

Extracts were injected into a capillary gas chromatography column (60 m \times 0.25 mm DB-5, J&W Scientific) with He carrier gas by use of splitless injection. Peak detection was with a ^{63}Ni μ -electron capture detector (HP 5890 Series

II GC). Major peaks were confirmed in selected samples by GC/MS (HP 5971 MSD) by use of a 30-m DB-5 column with He carrier gas. A total of 104 PCB congeners and 31 organochlorine pesticides and metabolites were quantified, with 50–60 PCB congeners and 17 organochlorine pesticides and metabolites regularly detected.

Air-water gas exchange—Net air-water fluxes across the lake surface were calculated by use of the two-film model (Schwarzenbach et al. 1993). Flux (F_G , $\text{ng cm}^{-2} \text{ s}^{-1}$) is calculated by use of OC concentrations in water and air and the temperature-adjusted Henry's Law constant (H) (Jantunen and Bidleman 1995) as follows:

$$F_G = K_{oi}(C_w - C_a RT/H), \quad (4)$$

where C_w and C_a are dissolved water and air concentrations (ng m^{-3}), R is the universal gas constant, T is absolute temperature, and K_{oi} is the overall mass transfer coefficient (cm s^{-1}), calculated as the sum of inverse resistance on the air and water side. (Schwarzenbach et al. 1993).

The H values were adjusted for the temperature at time of sampling by use of the estimates of enthalpy and entropy of phase change between gaseous and dissolved phases given by Tateya et al. (1988), Kucklick et al. (1991), ten Hulscher et al. (1992), and Bamford et al. (2000).

Air-side resistance (R_a , s cm^{-1}) is calculated by use of an empirical equation with air diffusivities (D_a , $\text{cm}^2 \text{ s}^{-1}$), Henry's constant (H , $\text{Pa m}^3 \text{ mol}^{-1}$), wind speed at 10 m height (U_{10} , m s^{-1}) and RT (Schwarzenbach et al. 1993):

$$R_a = 1/[(D_a/D_a^{\text{H}_2\text{O}})^{0.67} H/RT(0.2U_{10} + 0.3)]. \quad (5)$$

Wind speed was measured on site at 2 m height and converted to U_{10} with the conversion of Schwarzenbach et al. (1993). Likewise, water-side resistance under lentic conditions (no water flow) was calculated with water diffusivities and U_{10} as follows (Schwarzenbach et al. 1993):

$$R_w = 1/[(D_w/D_w^{\text{O}_2})^{0.57} (4 \times 10^{-5} U_{10}^2 + 4 \times 10^{-4})]. \quad (6)$$

Values used for $D_a^{\text{H}_2\text{O}}$ and $D_w^{\text{O}_2}$ are 0.26 $\text{cm}^2 \text{ s}^{-1}$ and $2.1 \times 10^{-5} \text{ cm}^2 \text{ s}^{-1}$, respectively. All other diffusivities were calculated by use of the compounds' molecular weights (MW) with the following equation (Schwarzenbach et al. 1993):

$$D_x^{\text{unknown}}/D_x^{\text{known}} = (\text{MW}_{\text{known}}/\text{MW}_{\text{unknown}})^{0.5}. \quad (7)$$

The net air-water flux (F_G) described above may be further divided into gas absorption or deposition (F_D) and volatilization (F_V) to quantify the components of this two-way exchange. This is achieved by calculating fugacities (f) in air and water as follows:

$$f_w = 10^{-6} C_w / \text{MW} Z_w, \quad (8)$$

$$f_a = 10^{-9} C_a / \text{MW} Z_a, \quad (9)$$

where MW is the molecular weight of the compound (g mol^{-1}); Z_w is the fugacity capacity of water, defined as $1/H$; and Z_a is the fugacity capacity in air, defined as $1/RT$. Fluxes by volatilization (F_V) and gas absorption (F_D , $\text{ng m}^{-2} \text{ day}^{-1}$) may now be quantified as follows:

$$F_V = 10^9 \text{MW} f_w D_{aw}, \quad (10)$$

$$F_D = 10^9 \text{MW} f_a D_{aw}, \quad (11)$$

where D_{aw} is the mass transfer coefficient ($\text{mol m}^{-2} \text{day}^{-1} \text{Pa}^{-1}$), which is given by

$$D_{aw} = 864K_{ol}/RT. \quad (12)$$

Sedimentation—Sedimentation in the basin was estimated with three sediment traps in the summer of 1998. Their positions are shown in Fig. 1 (marked “E”). They were suspended at a depth of 5 m. There were a total of six collections from the three traps between 11 June 1998 and 10 August 1998. Sediment traps were deployed for 33 ± 13 d. Sedimentation rates ($\text{g m}^{-2} \text{d}^{-1}$) were calculated from the sediment trap collections. These were multiplied by sediment concentrations of OCs collected from the sediment traps to determine the sedimentation rate of OCs from the epilimnion. Sediments were measured for OCs by use of the methods in Muir et al. (1996). Organic carbon content for suspended particles collected on GF/A filters from stream waters was measured by use of a total organic carbon (TOC) analyzer. Total organic matter was determined by use of loss on ignition at 550°C for sediment trap data (Dean 1974) and converted to organic carbon by use of the equation of Håkansson and Jansson (1983):

$$C_{\text{org}} = 0.48 \cdot \text{LOI} - 0.73, \quad (13)$$

where C_{org} is organic carbon concentration (% dry weight) and LOI is organic matter as loss on ignition (% dry weight).

Precipitation—Inputs to Bow Lake by rainfall were determined by multiplying average rain concentration of OCs with rainfall quantity to the surface of Bow Lake. Rainfall quantity was determined with a tipping bucket rain gauge at the weather station on the shore of Bow Lake. Concentration of OCs in rain was estimated from data collected in 1994, prior to our sampling, with a precipitation collector that was deployed 50 km from our site (at Wapta Lake, elevation 1,890 m above sea level). This precipitation collector passed rain water immediately through a solid-phase extraction column (XAD resin) that was collected each month and later extracted into hexane by Soxhlet, followed by Florisil cleanup and analysis by gas chromatography that used the methods described above. One XAD resin sample integrated rainfall for a period of 1 month for each of the 4 summer months.

Mass balance—A mass balance was constructed for Bow Lake to account for transfer of OC compounds by runoff, precipitation, gas exchange, sedimentation, and outflow. Gas transfer (mg) for the study period spanning 18 May to 26 August 1998 was calculated as follows:

$$\text{Gas transfer} = \sum_i (10^{-9} F_{G,i} LA t_i), \quad (14)$$

where t_i (d) represents time between samplings, LA is lake area ($2.8 \times 10^6 \text{ m}^2$), and 10^{-9} converts pg to mg. Volatilization and gas absorption were calculated by substituting $F_{V,i}$ and $F_{D,i}$ for $F_{G,i}$, respectively. Likewise, tributary fluxes for each stream (mg) were calculated as

$$\text{Tributary flux} = \sum_j (10^{-6} C_{w,j} Q_j t_j), \quad (15)$$

where $C_{w,j}$ is concentration in tributary water (pg L^{-1}) at time

j , Q_j is stream discharge ($\text{m}^3 \text{d}^{-1}$) at time j , t_j represents time between samplings, and 10^{-6} converts pg L^{-1} to mg m^{-3} . Daily stream discharge was combined with stream concentrations interpolated between sampling dates to calculate tributary fluxes for the 101-d sampling period. This approach assumes linear variation in concentration over time between samplings, regardless of discharge history. This was done because correlations between stream concentration of OCs and discharge were very rare over the 2-yr interval, and variability in stream concentrations of OCs was relatively small. Inputs by precipitation (mg) to the lake surface were estimated as

$$\text{Precipitation} = \sum_k (10^{-9} C_{\text{rain},k} \phi_{\text{rain},k} LA t_k), \quad (16)$$

where $C_{\text{rain},k}$ is concentration of chemical in rain at time k , $\phi_{\text{rain},k}$ is rain flux at time k (m d^{-1}), and t_k represents time between measurements of OCs in rain. Concentrations were calculated on the basis of monthly collections of extracts, so a monthly average rain concentration was multiplied by monthly rainfall measured at Bow Lake to determine input by rain. Sedimentation fluxes (mg) were estimated with fluxes measured in sediment traps as follows:

$$\text{Sedimentation} = \sum_l (10^9 C_{p,l} \phi_{p,l} LA t_l), \quad (17)$$

where $C_{p,l}$ is concentration of chemical on particles (pg g^{-1} organic carbon) at time l , $\phi_{p,l}$ is flux of organic carbon at time l ($\text{g m}^{-2} \text{d}^{-1}$), and t_l represents time between samplings. We also determined the change in chemical storage (mg) of the Bow Lake epilimnion during our sampling period as follows:

$$\Delta \text{ storage} = 10^{-6} \Delta C_w V_{\text{epi}}, \quad (18)$$

where ΔC_w is change in epilimnetic water concentration (pg L^{-1}) over the course of the study period, V_{epi} is epilimnetic water volume (m^3), based on the known lake area and a thermocline depth of 10 m, and 10^{-6} converts pg L^{-1} to mg m^{-3} .

Error analysis of this mass balance was determined by the propagation of error analysis proposed by Hoff (1994). Fractional coefficients of variation, or error terms (ϵ), were calculated for chemical fluxes by tributaries, outflow, precipitation, sedimentation, and gas exchange and used in the mass balance calculations to derive uncertainties for loading and loss terms.

Results

Concentrations in air—Concentrations of αHCH varied little over the summer, with no pattern observed as the summer progressed from colder to warmer temperatures (Fig. 2). Concentrations of αHCH averaged $37.5 \pm 7.6 \text{ pg m}^{-3}$ SD. Mean air concentrations for γHCH were lower ($20.0 \pm 6.9 \text{ pg m}^{-3}$ SD), with values declining rapidly as the summer progressed and as temperatures rose. The spring pulse in concentrations of γHCH , followed by a decline in summer, likely reflects springtime applications of lindane as a seed dressing to canola, corn, beans, and peas across western Canada (Li, Atmospheric Environment Service, pers. comm.). Lindane was one of the top 10 insecticides used in

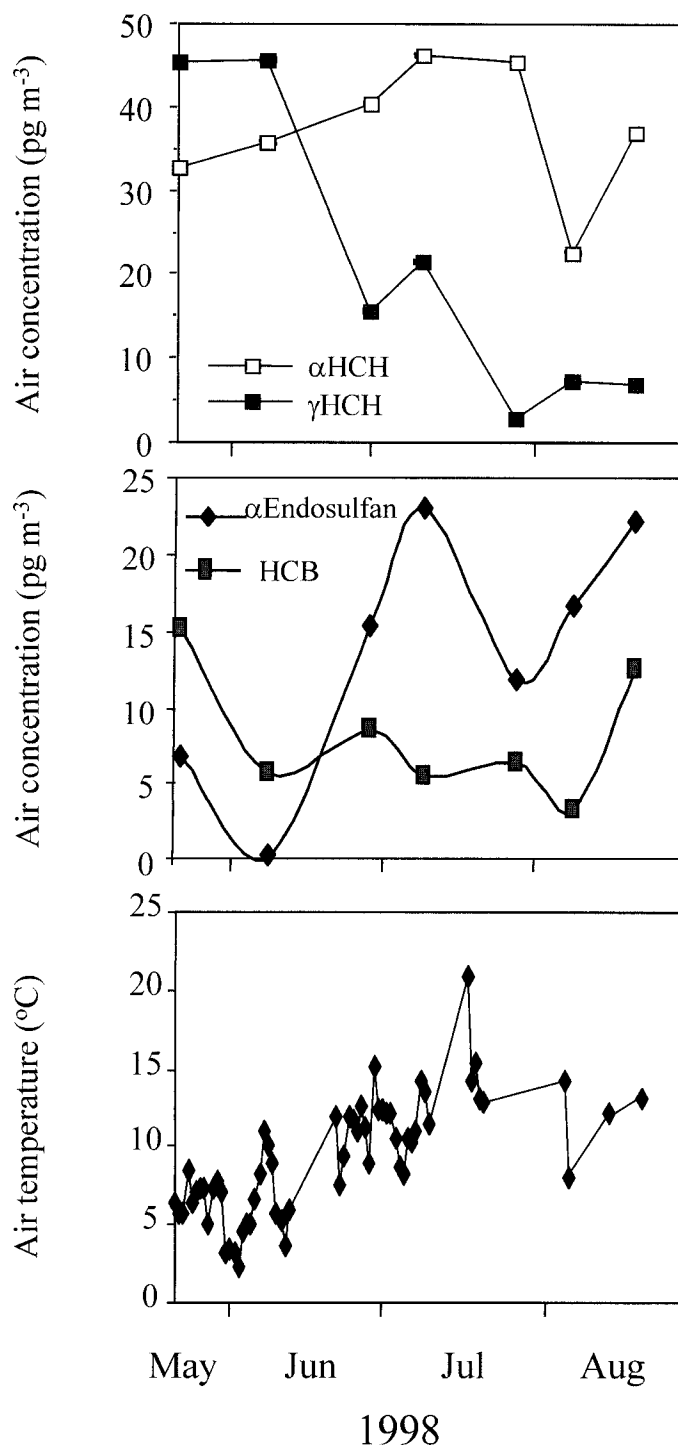


Fig. 2. Concentrations of α HCH, γ HCH, α endosulfan, and HCB in air during the summer of 1998, with concurrent air temperatures.

Canada (McConnell et al. 1993) until 1999 (Li, AES Downsview, pers. comm.). There was a progressive increase in the ratio of α/γ -HCH from May to August.

A declining air concentration of γ HCH as the summer progressed and temperatures rose coincides with our calculation of rapid gas absorption of γ HCH in early spring and

Table 1. Summary data for concentrations in air and the inflows and outflow of Bow Lake.

	α HCH	γ HCH	HCB	Dieldrin	α endo-sulfan	Σ PCB
Air at Bow Lake (pg m^{-3})						
Mean	38	20	8.3	0.48	14	14
SD	7.6	17	3.9	0.37	7.6	7.2
Max	46	45	15	1.0	23	29
Min	22	3	3	ND	0.30	7.2
N	8	8	8	8	8	8
Glacial stream (pg L^{-1})						
Mean	127	85	5.6	15	9.4	430
SD	85	60	2.4	4.9	8.7	310
Max	208	101	8.3	24	18	810
Min	ND	67	0.5	6.7	ND	124
N	8	8	8	8	8	8
Outflow (pg L^{-1})						
Mean	100	63	4.1	7.1	4.6	270
SD	36	24	1.0	1.7	1.5	230
Max	150	95	5.3	8.9	6.9	790
Min	56	35	2.8	4.3	ND	110
N	7	7	7	7	7	7
Valley stream (pg L^{-1})						
Mean	29	27	4.9	4.6	6.5	280
SD	10	24	2.4	1.6	5.1	260
Max	31	64	12	7.1	16	900
Min	3	3	5	2.8	ND	120
N	7	7	7	7	7	7
Alpine stream (pg L^{-1})						
Mean	13	25	7.3	7.1	6.4	380
SD	10	24	2.4	2.5	6.0	190
Max	31	64	12	9.6	18	720
Min	3	3	5	4.1	ND	180
N	7	7	7	7	7	7

a gradual equilibration in late summer. In contrast, α HCH maintains high air concentrations through the summer (Fig. 2). α HCH has a higher global atmospheric background concentration than other HCH isomers because more of it has been released to the environment and because it is the more persistent of the HCH isomers (Wu et al. 1997). γ HCH has been observed to isomerize to α HCH in soil and sediment studies (e.g., Wu et al. 1997). There are no recent local sources of α HCH to this area, and its presence is likely attributable to long-range transport.

There was no correlation between HCB concentrations and temperature (Fig. 2), with values averaging 8.3 ± 3.9 pg m^{-3} SD (Table 1). Other compounds that show no correlation between temperature and air concentrations include the PCBs, DDT and its metabolites, and dieldrin. A lack of correlation between OC partial pressures in air and temperature likely indicates that volatilization from local ground surfaces is not a major source of these constituents to air (Hoff et al. 1998; Wania et al. 1998). One notable exception was observed for endosulfan, for which vapor concentration increased significantly as summer progressed and temperatures rose (Fig. 2).

Table 2. Partition coefficients (K_p , L g⁻¹ ± SD) for OCs between suspended sediments and dissolved phases in the glacial and valley stream.

Compound	Glacial	Valley
αHCH	1.3 ± 1.4	39 ± 72
γHCH	3.7 ± 1.8	67 ± 77
HCB	8.8 ± 10	16 ± 7.6
Dieldrin	37 ± 24	572 ± 382
αEndosulfan	67 ± 76	788 ± 1126
βEndosulfan	7.9 ± 6.3	98 ± 97
PCB8/5	70 ± 31	150 ± 87
PCB18	237 ± 195	492 ± 562
PCB26	323 ± 328	703 ± 715
PCB44	623 ± 680	2097 ± 2152
PCB52	1175 ± 974	4954 ± 5708
PCB101	1114 ± 1193	4365 ± 4943
PCB153	570 ± 567	1821 ± 2567
PCB180	656 ± 401	1294 ± 1137

Water fluxes of OCs to Bow Lake—Sources of water to Bow Lake during the study period from 18 May to 26 August 1998 were 3.4×10^7 m³ from the glacial stream, 6.8×10^6 m³ from the valley stream, 3.4×10^5 m³ by rainfall, and 2.9×10^5 m³ from the alpine stream (Fig. 3). This added up to 4.1×10^7 m³ as total inflow by tributaries and rainfall. Total outflow volume during this time period was 4.7×10^7 m³ by comparison.

Dissolved concentrations of OCs were highest in the glacial stream (Table 1, Fig. 4), followed by the outflow stream (Table 1). Concentrations were lowest in the alpine and valley streams (Table 1). High concentrations in the glacial stream relate to a lower adsorptive capacity of suspended sediments and possibly soils in the glacial catchment. Partition coefficients (K_p) for OCs on sediments in the glacial and valley streams indicate a much lower affinity for suspended sediments in the glacial stream than for sediments in the valley stream (Table 2).

The glacial stream accounts for 99.2% of the flux of suspended sediment to Bow Lake by tributaries and 99.4% of the TOC flux from tributaries (Table 3).

Dissolved fluxes of these OCs from the glacial, valley, and alpine streams were similar in 1997 and 1998 (Table 4). This occurred in spite of higher discharge from the glacial stream in 1998, which was likely caused by low snow accumulation and a hot El Niño summer in 1998, which resulted in sustained high discharge in the glacial stream in late July and early August of that year (Fig. 3). Snowpack accumulation at Bow Summit in late March was 462 mm (water equivalent) in 1997 and 257 mm in 1998. Degree-day totals (January–August) were 838°C in 1997 and 1,148°C in 1998. Low snowfalls in 1998 over the Wapta Icefield would have resulted in more extensive exposure of glacial ice, which generally melts much faster than snow for a given energy input because of its lower albedo.

Rainfall inputs were a low proportion of the total according to our estimates. A limitation to our analysis is that we used current rainfall measurements at Bow Lake with archived rainfall data (1994) from Wapta Lake, a site 50 km distant. Although we acknowledge that this may introduce

Table 3. Suspended sediments in Bow Lake tributaries. Season (*) denotes the period between 18 May and 26 August 1998. Square brackets indicate organic carbon flux in kg season⁻¹.

Tributary	Suspended sediment conc. (mg L ⁻¹) ± SE, and number of observations	Seasonal flux (kg season ⁻¹ *)
Glacial	120 ± 50.1, n = 40	1.09×10^7 [1.7×10^6]
Valley	12.7 ± 5.6, n = 32	8.6×10^4 [1.0×10^4]
Alpine	0.38 ± 0.20, n = 32	1.10×10^2 [14]
Outflow	1.21 ± 0.48, n = 32	4.48×10^4 [5.8×10^3]

error to the model, we point out that our estimates of rainfall inputs of OCs to Bow Lake are relatively small compared with other sources. This is seen in spite of evidence that rainfall and snowfall inputs of these compounds have been either holding steady or declining over the past decade in Alberta (Crosley et al. 1998; Donald et al. 1999). Therefore, the relatively small inputs by rainfall that we report are likely an inflated representation of actual inputs.

Sedimentation—Sedimentation of organic carbon in Bow Lake was 9.3 ± 4 g m⁻² d⁻¹ in 1998, on the basis of five observations in each of three sediment traps deployed in Bow Lake between 11 June and 27 September (Table 5). The dominant source for sediments to Bow Lake is the glacial stream, where 120 + 50 (SE) mg L⁻¹ of suspended sediments were observed on the basis of 40 observations made between 8 May and 24 August 1998. This translates to 1.09×10^7 kg of suspended sediment transported to Bow Lake from the glacial stream during that time interval. Suspended sediment

Table 4. Comparison of fluxes (ϕ_w , mg season⁻¹) among the 1997 and 1998 stream sampling periods.

	ϕ_w (1997)	ϕ_w (1998)
Glacial		
αHCH	5200	5800
γHCH	2600	3760
αendosulfan	420	260
Dieldrin	530	500
HCB	140	160
Water (m ³)	2.5×10^7	3.4×10^7
Valley		
αHCH	184	170
γHCH	157	140
αendosulfan	46	25
Dieldrin	57	30
HCB	40	32
Water (m ³)	8.0×10^6	6.8×10^6
Alpine		
αHCH	13	5
γHCH	10	8
αendosulfan	4	2
Dieldrin	4	2
HCB	2	3
Water (m ³)	3.6×10^5	2.9×10^5

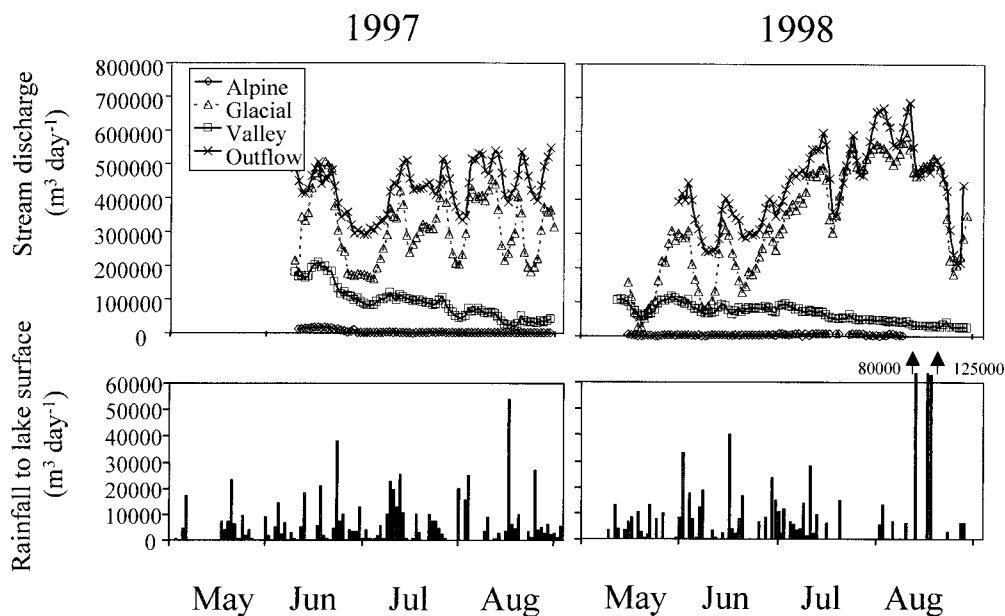


Fig. 3. Stream discharges and rainfall inputs to Bow Lake in 1997 and 1998. These data were used to construct a water budget for Bow Lake.

losses from the lake's outflow were estimated to be only 4.48×10^4 kg of sediment over the same time interval.

Gas exchange—The two HCH isomers (α and γ) showed a net absorption to water from air (Fig. 5). Lake water was undersaturated in HCH relative to its partial pressure in air; thus, there was a net absorption of HCHs for much of the ice-free season. This may partly explain why the HCH concentration in lake water is so much higher than that of other OCs, although high delivery of HCHs by runoff is another important factor. The highest rates of absorption of HCHs by water were observed in the early summer, when temperatures were lowest. As the summer progressed and temperatures increased, the rate of gas absorption declined for HCHs, particularly for the γ isomer (Fig. 5). This is the combined result of higher temperatures and a drop in air concentration of γ HCH (Fig. 2) after seed applications in early spring. Thus, atmospheric delivery of γ HCH is a pulse in early summer, with air concentrations dropping and waters gradually equilibrating with air by late July.

Most other constituents showed a net volatilization from the lake surface, particularly dieldrin, p,p'DDD, p,p'DDT, and most PCB congeners (Figs. 5, 6). We observed the highest rates of volatilization for many PCB congeners in late spring and early summer (Fig. 6). Although temperatures were lowest during this period, favoring decreased volatilization, water concentrations of PCBs were highest after the snowmelt pulse, which caused higher volatilization losses of PCBs during this period. The volatilization of OCs like PCBs, DDD, and dieldrin to the air throughout the summer months indicated that stream inflows were the dominant sources of these chemicals to Bow Lake.

Whole-lake mass fluxes—The glacial tributary is clearly the dominant source of most OCs to Bow Lake (Figs. 7, 8). Dissolved fractions in the glacial stream dominated for HCHs, and suspended sediment loads from the glacial stream were the dominant source of most other constituents to Bow Lake, including dieldrin, endosulfan, and the PCBs. These results help to explain why glacially fed lakes have

Table 5. Sediment accumulation measured in sediment traps. C_{org} sedimentation rate ($\text{g m}^{-2} \text{d}^{-1}$) denotes sedimentation of organic carbon.

Date deployed	Date collected	No. days deployed	Total sediment collected (g)	Sedimentation rate ($\text{g m}^{-2} \text{d}^{-1}$)	C_{org} sedimentation rate ($\text{g m}^{-2} \text{d}^{-1}$)
11 Jun 98	21 Jul 98	40	47.1	150	19
21 Jul 98	10 Aug 98	20	0	0	0
10 Aug 98	27 Sep 98	48	49.8	130	3.5
21 Jul 98	10 Aug 98	20	54.5	350	15
10 Aug 98	27 Sep 98	48	7.5	20	7.9
Mean				110	9.3
SE				42	4.0

Over the 101-d season, and a 2.8 km^2 surface area, we estimate 2.6×10^6 kg of organic carbon deposited in Bow Lake.

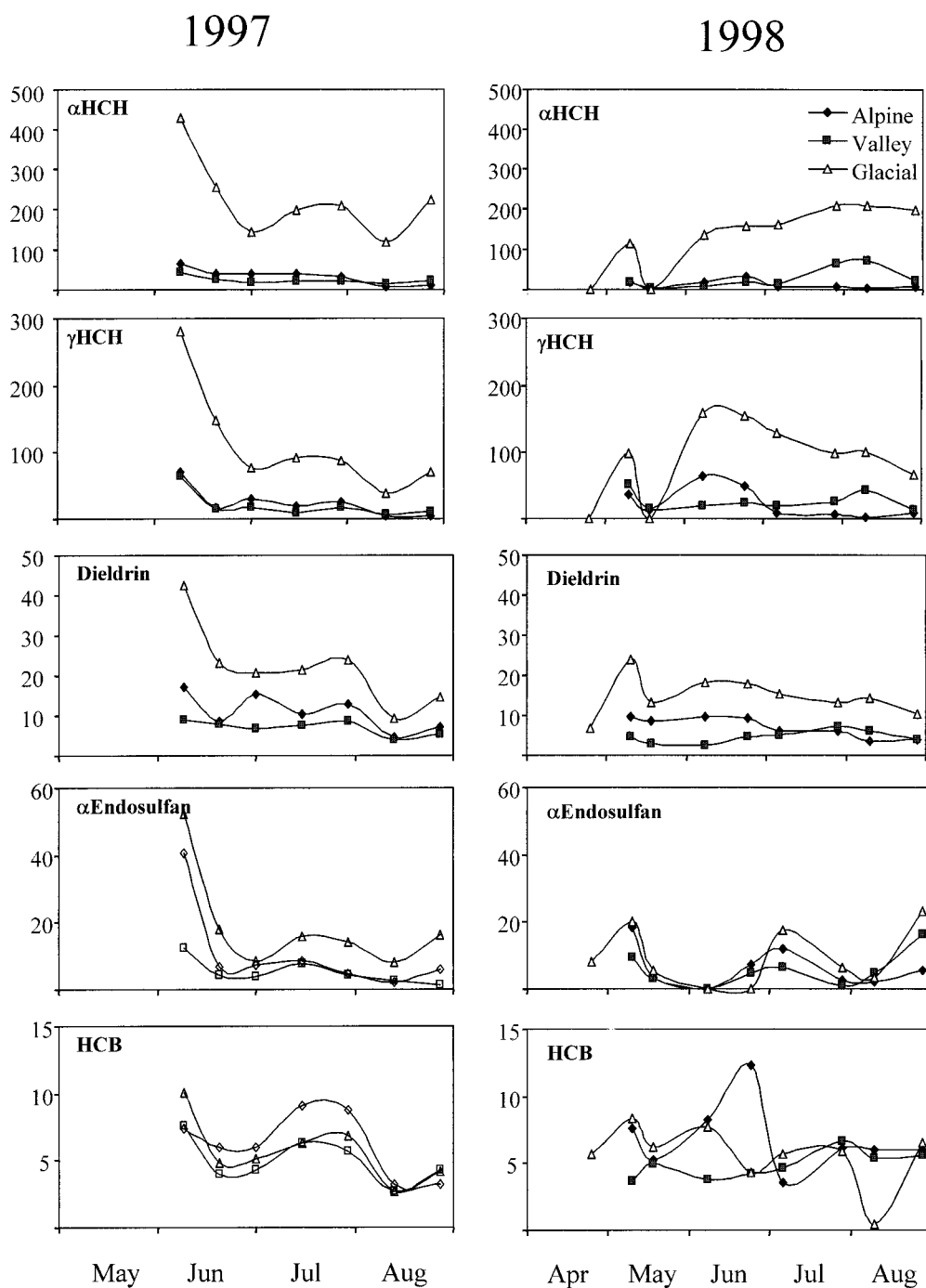


Fig. 4. Concentrations of several OCs in the three tributaries draining into Bow Lake.

the highest contaminant burdens in their foodwebs (Donald 1993). Wet deposition contributed significantly to loading of HCHs but was a minor source of other constituents such as dieldrin and PCBs. With net volatilization predominating for all constituents except the HCHs and precipitation accounting for such a small proportion of total inputs, tributary inputs, mainly from glacial sources, were clearly the dominant sources of OCs to Bow Lake. Tributary inputs accounted for 79% of dieldrin inputs and 92% of Σ PCB inputs, for example (Figs. 7, 8).

Error analysis—Error terms (or fractional coefficients of variation) were calculated for input and loss terms ($\varepsilon_x = \sigma / \langle x \rangle$, where σ is the SD of variable x and $\langle x \rangle$ is the mean value of x) (Table 6). These ε values tended to be highest for gas absorption and volatilization estimates, because of the dependence of their derivation on sensitive parameters (e.g., air and water OC concentration and mass transfer coefficients), and sedimentation losses because of a high temporal variability in sediment fluxes (Table 5). Tributary inputs, on the other hand, have relatively small temporal

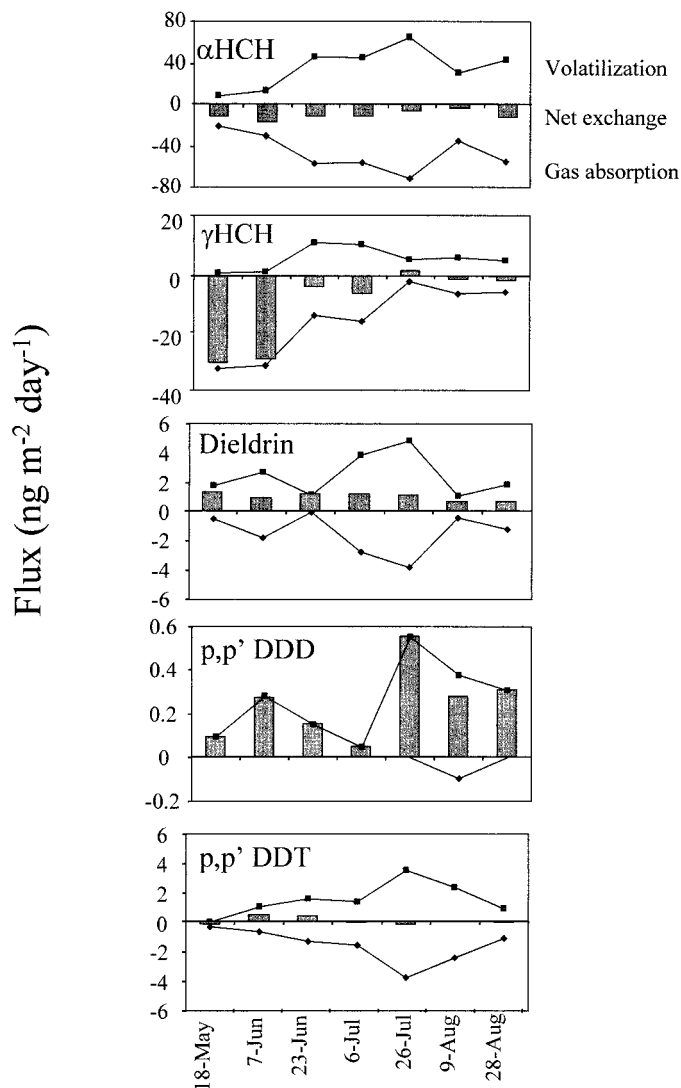


Fig. 5. Net air-water fluxes of OCs by gas exchange into and out of Bow Lake during the summer of 1998. Positive fluxes indicate a net volatilization to air, and negative fluxes indicate a net absorption by the lake.

variability, and there is little error associated with these values, which, in many cases, constitute a major source of inputs to Bow Lake. This analysis of error propagation aids in explaining imbalances observed in the Bow Lake budget. For example, inputs exceed losses for both α - and γ HCH, but the imbalance (inputs – outputs) does not exceed the coefficients of variation for the loss terms multiplied by the mean flux values. Thus, although an imbalance is measured, our data do not allow speculation as to why such an imbalance may exist because the uncertainties in the combined mass budget exceed the imbalances.

Discussion

These results show the importance of tributaries as a source of OCs to Bow Lake. Atmospheric transport is the most likely source of OCs to these environments because

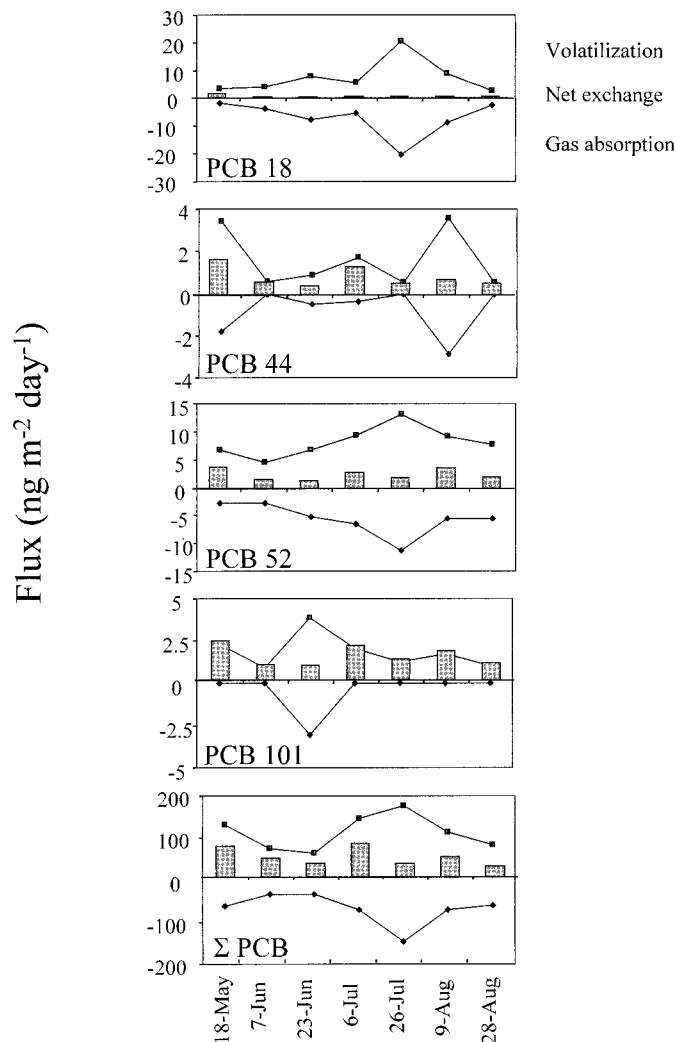
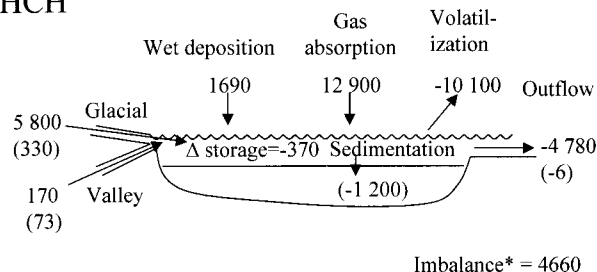


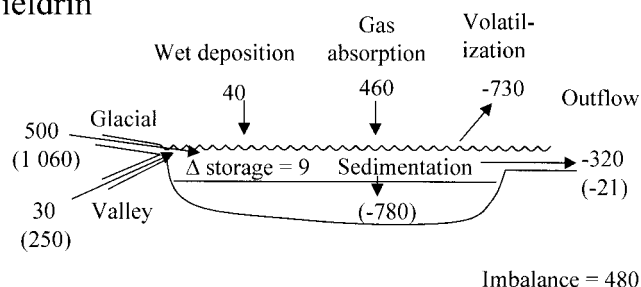
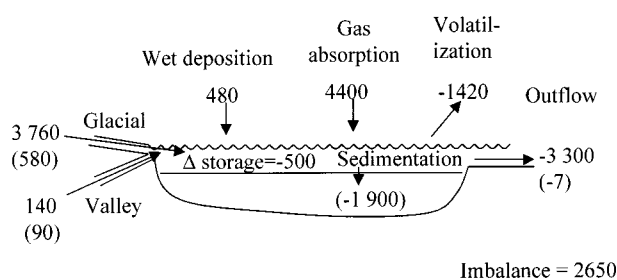
Fig. 6. Net air-water fluxes of PCBs by gas exchange into Bow Lake during the summer of 1998. The positive fluxes all indicate a net volatilization to air.

applications of these chemicals in the Park were negligible. However, after deposition, surface runoff becomes a major route of transport to Bow Lake. Many OCs in runoff peaked in concentration in early June 1997 during the snowmelt pulse; however, a similar peak was not observed in 1998 (Fig. 4). The snowmelt pulse in 1998 was not as pronounced and began earlier than in 1997 because of earlier warming and a thin overwinter snowpack; thus, if a spring pulse occurred in 1998, it preceded our sampling.

Other stream sampling has shown spring pulses of α HCH, γ HCH, and endosulfans coincident with snowmelt pulses (Wania et al. 1999; MacDonald et al. 2000). These more polar compounds tend to elute readily through the snowpack, whereas more volatile substances (chlorobenzenes) tend to volatilize to the air, and more hydrophobic substances (DDT and PCBs) tend to be retained by organic detritus in snowpacks (Wania 1997). Bremle and Larsson (1997) recorded an inverse relationship between discharge and dissolved PCB concentration, which suggests that desorption of PCBs from sediment is an important determinant of PCB concen-

α HCH

Dieldrin

 γ HCH

*Imbalance = inputs-outputs

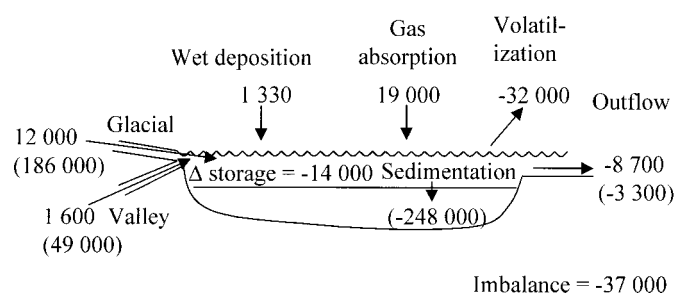
 Σ PCB

Fig. 7. Mass balance of α HCH and γ HCH for Bow Lake. Numbers are total fluxes of chemical (mg) over the 101-d study period. Brackets show chemical fluxes on particles. Positive numbers are inputs to the Bow Lake epilimnion, and negative numbers are losses. Δ storage is the mass change of chemical (mg) in Bow Lake's epilimnion over the same time interval. *Imbalance = inputs - outputs.

Fig. 8. Mass balance of Dieldrin and Σ PCB for Bow Lake. Numbers mean the same as described in the caption for Fig. 7.

trations in stream waters. We observed no correlation between PCBs and stream discharge in any of our streams.

The high values of HCHs in glacially derived streams may reflect the effective elution of these chemicals from snow and firn, as described by Wania (1997). Another contributor to these high concentrations may be that the glacial stream derives runoff partly from older glacial strata that are known to contain high concentrations of these chemicals (Donald et al. 1998; Blais et al. 2001). Studies have shown much higher concentrations of HCHs in stream water during the 1960s and 1970s (e.g., Donald et al. 1998). Stream monitoring of HCHs in Alberta rivers since the 1970s has shown a four- to sevenfold decrease between 1975 and 1994 (Crosley et al. 1998). We have observed increased tritium levels in the Bow glacial stream in late summer indicative of waters deposited during accelerated atmospheric nuclear weapons testing (1950s to mid-1970s; Blais et al. 2001). This provides evidence that a proportion of these glacial stream waters derive from decades-old glacial ice. We know from analysis of a glacial ice stratigraphy at Snow Dome, which is situated within 50 km of our study site, that HCH deposition to glaciers was 100 times higher during the mid-1940s than during the mid-1990s (Donald et al. 1999). Likewise, Jantunen and Bidleman (1995) have recorded declines in air concentra-

tions of HCHs since the 1970s over arctic stations near the Bering and Chukchi Seas. However, not all OCs in these studies were seen to decline over this time interval. HCB increased steadily toward the surface of Snow Dome glacier, for example, and dieldrin, endosulfan, chlordanes, and heptachlor epoxide were all higher in horizons corresponding to the 1980s and 1990s than they were in ice layers deposited during the 1950s-1970s (Donald et al. 1999).

Air concentrations of α HCH averaged 37.5 ± 7.6 $\mu\text{g m}^{-3}$ SD, which is somewhat lower than values measured over the Pacific Ocean (Iwata et al. 1993; Jantunen and Bidleman 1995; Barrie et al. 1997). Air concentrations of α HCH over the Bering Sea ranged from 44 to 120 $\mu\text{g m}^{-3}$ (Barrie et al. 1997), but these may be influenced by inputs from Asia, which is likely an important source for this isomer (Iwata et al. 1993).

The seasonal decrease in γ/α HCH ratios that we observed in both air and water after the spring pulse suggests that a transient, regional source of γ HCH was active over this time interval. An early spring pulse of γ HCH in air followed the annual lindane application on seed planting, and this interpretation of high lindane in spring air has also been made in other studies in Canada (e.g., Poissant and Koprivnjak 1996).

Gas exchange across the surface of Bow Lake favored volatilization for most compounds except the HCHs and p,p'DDT. This study describes a direct impact of spring γ HCH (Lindane) application on both air concentration of that chemical and gas absorption to a distant water body.

Table 6. Calculated fluxes and propagated error estimates to Bow Lake for 1998. Positive values represent inputs, and negative values represent losses.

Source	Loading (mg season ⁻¹)	Propagated error ϵ
αHCH		
Tributaries	6,370	0.62
Outflow	-4,790	0.33
Gas absorption	12,900	2.71
Volatilization	-10,100	2.65
Precipitation	1,690	0.79
Sedimentation	-1,200	0.80
Δ storage	-370	0.33
Total Inputs	20,960	0.65
Total Outputs	-16,300	1.64
γHCH		
Tributaries	4,570	0.65
Outflow	-3,300	0.35
Gas absorption	4,400	2.72
Volatilization	-1,420	2.77
Precipitation	480	0.49
Sedimentation	-1,900	0.85
Δ storage	-500	0.35
Total Inputs	9,450	0.56
Total Outputs	-6,800	0.64
Dieldrin		
Tributaries	1,840	0.34
Outflow	-340	3.15
Gas absorption	460	2.66
Volatilization	-730	2.75
Precipitation	40	1.46
Sedimentation	-780	0.77
Δ storage	9	0.23
Total Inputs	2,340	0.33
Total Outputs	-1,860	1.29
ΣPCB		
Tributaries	248,600	0.72
Outflow	-12,000	0.12
Gas absorption	19,000	6.04
Volatilization	-32,000	6.02
Precipitation	1,330	0.70
Sedimentation	-248,000	0.99
Δ storage	-14,000	0.84
Total Inputs	269,000	0.67
Total Outputs	306,000	1.03

Σ PCB consisted of the following congeners, with dashes representing co-eluting PCBs: 1, 3, 4-10, 9-7, 6, 8-5, 19, 12-13, 18, 15-17, 24-27, 16-32, 54-29, 26, 25, 31-28, 33, 20, 53, 51, 22, 45, 52, 49, 43, 48-47, 44, 59, 42, 64-41-71, 100, 63, 40, 74, 70-76-98, 66, 95, 91, 55, 56-60, 92, 84, 101, 99, 119, 83, 97, 87-81, 85, 136, 110, 82, 151, 135-144, 147, 107, 149, 118, 133, 114, 134-131, 146, 153, 132, 141, 179, 105, 137, 176, 130, 138-163, 158, 129, 178, 175, 187-182, 183, 128, 167, 185, 174, 177, 202-171, 156, 173, 157-200, 172, 197, 180, 193, 191, 199, 170-190, 198, 201, 203-196, 189, 206-195, 207, 194, 205, 208, 209.

From this pulse of γ HCH in air over an \sim 40-d period, we estimate gas absorption of 4 g of γ HCH to Bow Lake directly as a result of this seed application. Although net volatilization of HCHs is largely reported in marine environments such as the Bering and Chukchi Seas (Jantunen and Bidleman 1995), net gas absorption of HCHs is often re-

ported in freshwaters because there are generally lower water concentrations of HCHs in freshwaters compared with ocean waters. For example, a net gas absorption of HCHs was shown in Lake Baikal in Russia (McConnell et al. 1996), and Ridal et al. (1996) observed net absorption of HCHs by Lake Ontario in early spring but net volatilization by mid-summer. This was a result of increasing temperatures and a high storage of HCHs in Lake Ontario waters, which was significantly enhanced by runoff from local sources. Bow Lake does not receive agricultural effluents, and HCH concentrations in water never became high enough to promote net volatilization to the air. PCBs, on the other hand, showed a net volatilization from the lake throughout the ice-free season, particularly from May to early July (Fig. 6). This occurred in spite of lower temperatures during these early months (Fig. 2), which would not favor volatilization. Rates of PCB volatilization were sensitive to changes in measured PCB concentration in water, which were high during snow-melt in May and June and gradually declined as the summer progressed.

Whole-lake mass fluxes—There are very few mass flux estimates of these OC compounds over a whole-lake scale, and most studies of this nature were for the Great Lakes (e.g., Jeremiasson et al. 1994). Recently, PCB fluxes in two small boreal lakes at the Experimental Lakes Area in northwestern Ontario were compared, and it was concluded that tributaries were likely important PCB sources (Jeremiasson et al. 1999). Although tributary inputs of PCBs were not measured in that study, the large imbalance between inputs and outputs was taken as evidence that tributaries must be a major source of PCBs to the lakes (Jeremiasson et al. 1999). Our study supports this conclusion, showing that tributaries to Bow Lake accounted for an estimated 92% of PCB inputs. This proportion is made especially large by inputs from glacially derived waters.

Although we measured the tributary inputs to Bow Lake, imbalances in OC inputs and outputs still persisted. One term that we did not measure in this mass balance was loss by reaction in the water column. OCs are susceptible to numerous hydrolysis, photolysis, and dehydrochlorination reactions in the water column (e.g., Mackay et al. 1997), which we were unable to measure. One possible reason for the imbalances that we observed for OCs in Bow Lake might be reaction losses, particularly for the HCHs that have relatively short (30–300 d) reaction half-lives in water (Mackay et al. 1997). Given these half-lives, our reported HCH concentrations in Bow Lake, and an estimated lake volume of 5.6×10^7 m³, we estimate reaction losses of α HCH over the 101-d sampling to be between 1,300 and 13,000 mg, compared with our calculated imbalance (inputs-outputs) of +4,660 mg. For γ HCH, we used these rough estimates to calculate reaction losses between 820 and 8,200 mg, compared with our calculated imbalance of +2,650 mg. Thus, although uncertainties in inputs and outputs restrict our speculation of causes for imbalances, reaction losses are likely to be responsible for a noticeable contribution to the balance.

Conclusions—This study showed the importance of glacial runoff and gas exchange of OCs as primary sources of

contaminants in mountain lakes. Inputs by runoff, particularly by glacial melt and snowmelt in late spring, and gas absorption across the lake's surface constitute the bulk of inputs. However, net gas absorption to Bow Lake is only seen to a significant extent for the α and γ isomers of the HCHs. Glacier-fed lakes may be contaminated with OCs for years in the future because of the high storage of these substances in glaciers and their propensity to be released to surface waters. Previous surveys that have shown contaminated proglacial lakes in Banff National Park (Donald 1993) should be extended to other glaciated parts of the world, including northern Canada and Greenland, where PCB and other OC problems persist (e.g., Barrie et al. 1997).

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