Evidence for Atmospheric Transport and Deposition of Polychlorinated Biphenyls to the Lake Tahoe Basin, California—Nevada

SEEMA DATTA,† LAURA L. MCCONNELL,‡
JOEL E. BAKER,§ JAMES LENOIR, AND
JAMES N. SEIBER*, B

Department of Environmental Toxicology, University of California, Davis, California 95616, Agricultural Research Service, United States Department of Agriculture, Beltsville, Maryland 20705, Chesapeake Biological Laboratory, University of Maryland, Solomons, Maryland 20688, and Center for Environmental Sciences and Engineering, University of Nevada, Reno, Nevada 89557

PCB congener concentrations in air, water, fish, and snow were determined in the Lake Tahoe basin and in the water and fish of an adjacent but more remote rural comparison lake, Marlette Lake, to examine whether atmospheric transport and deposition are primary sources of contamination to this alpine region of the Sierra Nevada Mountains. Profiles of 97 congeners analyzed by capillary GC-ECD from surface waters of Lake Tahoe and Marlette Lake were closely matched, with total dissolved PCB (t-PCB) concentrations of 0.37 ng/L in Lake Tahoe and 0.67 ng/L in Marlette Lake. Gas-phase t-PCB concentrations measured in air samples from the Lake Tahoe basin averaged 72 pg/m³, and t-PCB (dissolved and particulate) concentrations in snow were 4.8-5.1 ng/L. Rainbow trout from Marlette Lake and lake trout from Lake Tahoe had similar distributions of congeners, with t-PCB tissue levels varying from 3 to 14 ng/g wet weight. The finding of PCBs in all sampled compartments, particularly snow and air, and the similar pattern of congeners in surface waters and in fish from Lake Tahoe and Marlette Lake indicate an atmospheric source of contamination.

Introduction

Atmospheric deposition of organic pollutants has been reported in remote lake ecosystems including Siskiwit Lake, Isle Royale, Lake Superior (1, 2), lakes in the northeastern United States (3), high-altitude lakes in the Rocky Mountains of Colorado (4), and remote lakes in Ontario, Canada (5). Several studies have suggested that the Sierra Nevada Mountains of California—Nevada are subject to contamination from airborne pollutants (6-10); however, little information is available on the occurrence and distribution of organic pollutants in this subalpine region. Atmospheric deposition of trace organics was first suggested in a study reporting the presence of p,p'-DDE in yellow-legged frogs

(Rana boylii) collected from numerous remote high-altitude locations in the Sierra Nevada Mountains (11). More recently, organophosphate insecticides were measured at over 1900 m elevation in precipitation in the southern regions of the mountain range (10). Pesticides and other chemical pollutants are currently among the suspected causes of the relatively recent disappearance and decline of the yellow-legged frog, other ranid frogs, and other amphibians in the regions of the mountain range (12, 13).

Pesticide contamination in the mountains is assumed to be related to prevailing wind patterns from the coast and interior valleys immediately to the west of the Sierra Nevada Mountains (6, 10, 11). Winds moving eastward across the coastal cities and valleys can pick up volatile and semivolatile pollutants and subsequently deposit them through gas exchange to vegetation and surface waters or through precipitation upon reaching the Sierra Nevada Mountains, which extends in a north—south direction parallel to the Sacramento and San Joaquin valleys of California.

A recent study showed the presence of organochlorine (OC) contaminants in lake trout and kokanee fish from Lake Tahoe, an alpine lake situated at an elevation of 1899 m in the central Sierra Nevada Mountains (14). Multi-residue screening, using gas chromatography-electron capture detection (GC-ECD) techniques applicable for a variety of halogenated pesticides and industrial pollutants indicated the presence of two dominant residues, polychlorinated biphenyls (PCBs) and p,p'-DDE. Total Aroclor and p,p'-DDE concentrations ranged from 5 to 430 ng/g wet weight in two species of fish tested. Confirmation of these compounds was achieved using electron impact gas chromatographymass spectrometry (GC-MS) techniques. Contribution of air transport as a potential source of pollutant input was suggested on the basis of several factors: (a) Lake Tahoe receives only minor direct contamination from industrial discharges or domestic and agricultural runoff, (b) precipitation falling on the lake's surface is a major source of water to the lake, and (c) the lake's drainage basin is relatively small compared to its surface area. Contamination of the Sierra Nevada Mountains with OC and organophosphate chemicals has been assumed to involve atmospheric deposition based upon prevailing meteorology and physical characteristics of the Lake Tahoe basin (6, 10, 11, 13), but no direct evidence has been reported to confirm that long-range aerial transport and deposition is the dominant source of these pollutants.

The primary objective of this study was to provide evidence that atmospheric transport—deposition is the significant source of OC compounds, particularly PCBs, to lakes of the Sierra Nevada Mountains. Evidence is based upon analysis of fish, water, air, and precipitation in and around Lake Tahoe and at a more remote and pristine lake (Marlette Lake) located within the Lake Tahoe basin. PCB congener profiles of fish and surface waters of Marlette Lake and Lake Tahoe were compared. A simple PCB loading inventory was constructed to estimate the input of atmospherically derived PCBs to Lake Tahoe.

Experimental Section

Study Site. Both Lake Tahoe and Marlette Lake are located in the Tahoe basin in the central Sierra Nevada Mountains of California and Nevada (Figure 1). The Lake Tahoe basin formed when faulting between the crests of the Sierra Nevada and the Carson Range created a valley. Mountains on each side of the valley rose, forming a graben or trough which later filled with water to become a deep, oligotrophic lake

^{*} Corresponding author phone: (702) 784-6460; fax: (702) 784-1142; e-mail: jseiber@med.unr.edu.

[†] University of California.

[‡] United States Department of Agriculture.

[§] University of Maryland.

[∥] University of Nevada.

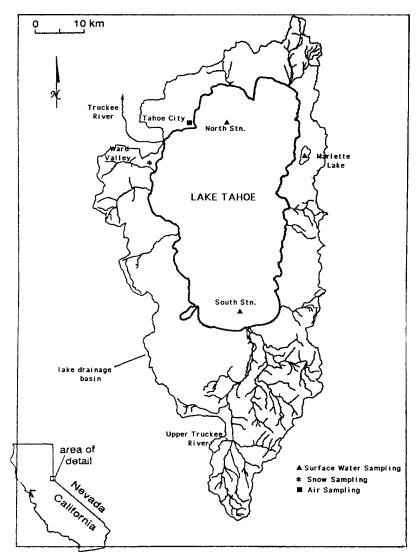


FIGURE 1. Map of Lake Tahoe basin showing sampling sites.

with a low watershed-to-lake surface area ratio. Having great depth (maximum, 505 m), volume (156 km³), and only one outlet (the Truckee River) leads to a self-purging time of the water in the lake of an estimated 700 years (15).

Topography of this region strongly affects climate. In general, marine surface winds coming from the Pacific Ocean release their moisture as they rise over the crest of the Sierra Nevada Mountains. Western Sierran slopes receive roughly double the annual precipitation compared to the drier eastern slopes. Precipitation, mainly in the form of winter and spring snow and rain falling directly on the surface of the lake, comprises approximately 40% of the lake's water inflow. The remaining inflow is from tributary streams.

In contrast to Lake Tahoe, Marlette Lake is a small shallow reservoir (13 m maximum depth) situated within the Nevada State Park System in the Carson Range of the Sierras at an elevation of 2384 m (16). Similar to Lake Tahoe, snowmelt and runoff are the only sources of water to this reservoir. Public access is via hiking trails open during nonwinter months. The Nevada Division of Fish and Game employs the lake for spawning cutthroat trout. The prohibition on other public use of Marlette Lake is strictly enforced.

Sample Collection. High-volume surface water samples were collected from Lake Tahoe aboard the Research Vessel John Le Conte between June 20 and 22, 1995. Total volumes of 180 L were collected in 18 L stainless steel cans at two sites, North Station (39°05.63′ N120°09.00′ W) and South

Station (38°59.25′ N120° 01.76′ W) (Figure 1). Water collecting cans were secured with lines and dropped from the bow of the ship as it slowly moved forward. In December 1995, a 50 L volume of surface water sample was collected at the shore of Marlette Lake in a combination of cleaned 18 L cans and in 4 L glass amber jars. In total, three high-volume water samples were collected, two from Lake Tahoe and one from Marlette Lake.

A high-pressure stainless steel filter head assembly (Millipore) and pump were used to filter water through previously baked (450 °C for 24 h) glass fiber filters (Whatman, 9.0 cm diameter Glass Microfiber Filters). Hydrophobic organic residues contained in filtered surface water were isolated by pumping water at approximately 200 mL/min through two Amberlite XAD-2 resin (Sigma) custom-made glass columns (Ace Glass) 21 mm × 150 mm, connected in series to ensure no breakthrough of organic residues. This type of extraction method has been used previously for PCB extraction from surface waters (17, 18). Column fittings and tubing were of Teflon construction. All water and snow samples were processed through only one resin column, since no evidence of breakthrough was noted in initial examination of Lake Tahoe water. XAD-2 columns were stored sealed with endcapped fittings and refrigerated until analysis. The procedure for purification of XAD resin is reported elsewhere (14).

TABLE 1. Method Recovery of PCB Surrogates in Blanks and Environmental Samples

	mean percent recovery and range of surrogates			
	PCB 14	PCB 65	PCB 166	
surface water and snow				
XAD field blanks, $n = 4$	$82 \pm 14 \ (67 - 101)$	$83 \pm 11 \ (70 - 99)$	$91 \pm 20 \ (71 - 123)$	
XAD water samples, $n = 3$	$103 \pm 17 (87 - 124)$	$103 \pm 6 \ (96 - 109)$	$115 \pm 19 (99 - 139)$	
XAD snow samples, $n = 2$	$132 \pm 4 \ (129 - 135)$	$123 \pm 25 \ (105 - 123)$	$130 \pm 9 \ (123 - 136)$	
snow filters, $n=2$	$113 \pm 9 (112 - 140)$	$127 \pm 5 \ (123 - 131)^{2}$	$132 \pm 3 (130 - 134)$	
air, PUFs, and filters	,	,	,	
field blank PUFs, $n = 3$	$83 \pm 4 (79 - 83)$	$88 \pm 18 \ (76 - 101)$	$114 \pm 9 \ (103 - 119)$	
air samples, 2 PUFs/sample, $n = 2$	$100 \pm 5 (95 - 107)$	$75 \pm 8 (66 - 83)$	$107 \pm 8 (98 - 114)^{'}$	
filter blank, $n=1$	112 ` ´	92 ` ´	87 ` ´	
air sample, filters, $n=2$	111 (nd ^a – 111)	5 (nd - 51)	67 (63-71)	
fish	,	,	,	
fish, $n=3$	108 \pm 2 (84 $-$ 125)	114 \pm 17 (95 $-$ 122)	126 \pm 22 (104 $-$ 148)	
and = represents not detected				

Two snow samples of 56 and 26 L (after melting) were taken in clean 75 L gallon galvanized aluminum cans on December 18 and 28, 1995, near a subalpine tributary stream, in Ward Valley, to the west of Lake Tahoe (Figure 1). Clean cans were partially filled with snow, which accumulated in an adjacent open field, using a cleaned aluminum shovel. Covered containers of snow were slowly thawed overnight indoors, and melted snow water was periodically transferred to 18 L stainless steel cans using a stainless steel funnel. Identical procedures were employed for extracting snowmelt water as were used for surface water.

Two high-volume air samplers (Grasby) borrowed from the California Air Resources Board were set up near the University of California, Davis, Tahoe Research Group laboratory in Tahoe City, approximately 100 m from the northern shore of Lake Tahoe (Figure 1) on July 6–7, 1995. Air samplers, A and B, with flows of 1.29 and 1.37 m³/min, respectively, were operated for 24 h. Glass fiber filters (GFF, 20.3 cm \times 25.4 cm, Gelman Type A) baked at 400 °C for 24 h, were used to collect aerosol particles. Cleaned polyure-thane foam (PUF) plugs were used to trap vapor components from the air stream. For cleaning, PUFs were washed twice in warm tap water and distilled water. After being rinsed in acetone (GC Resolve, Fisher Scientific), PUFs were further cleaned by 24 h Soxhlet extractions sequentially in acetone and hexane, then dried in a vacuum oven for 24 h (19).

Juvenile lake trout (*Salvelinus namaycush*) (n=2) from Lake Tahoe (November 18, 1995) and rainbow trout (*Oncorhynchus mykiss* ssp.) (n=6) from Marlette Lake (December 4, 1995) were caught by hook and line from near the shore. Samples were wrapped in aluminum foil and transported on ice until storage in the laboratory at $-30~^{\circ}\text{C}$.

All XAD resin and filter extraction, cleanup, and fractionation were carried out at the Chesapeake Biological Laboratory in Solomons, MD, following the methods of Leister and Baker (20) and Baker and Eisenreich (21). A detailed description can be found in Datta (14). Internal standards of congeners that are not present in commercial PCBs, 2,4,6-trichlorobiphenyl (IUPAC No. 30) and 2,2,3,4,4′,5,6,6′-octachlorobiphenyl (IUPAC No. 204) (Ultra Scientific), 50 μ L of 100 ng/mL in hexane, were added to each sample vial prior to chromatographic analysis.

Extraction. The method of fish extraction was patterned after that of Ribick et al. (22). Briefly, the procedure involved grinding 10 g of fish tissue with 40 g of Na₂SO₄, then extracting the packed mixture in a glass column with dichloromethane (DCM) to obtain fish lipids. Fat content of 10 g of fish tissue ranged from 0.1 to 1.3 g (0.5 \pm 0.5 g). Chlorinated organic components were separated from fish lipids using gel permeation chromatography. Extracts were cleaned up on Florisil and silica gel columns, which separated PCBs from

to xaphene. Internal standards (IUPAC Nos. 30 and 204) were added before ${\rm GC-ECD}$ analysis.

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Analysis. All PCB congener analyses were performed using a Hewlett-Packard 5890A gas chromatograph equipped with a 63 Ni electron capture detector. A 60 m DB-5 capillary column of 0.25 mm i.d. and 0.25 μm film thickness (J&W Scientific) was used. Splitless injections of 2 mL were made with a Hewlett-Packard (7673) autosampler. Carrier and makeup gases were ultrahigh purity hydrogen (1 mL/min) and nitrogen (30 mL/min), respectively, and the column head pressure was 100 kPa. The oven temperature program was 100 °C for 2 min; 4 °C/min to 170 °C; 3 °C/min to 280 °C; 280 °C for 5 min. A Hewlett-Packard Chemstation 3365 was used for integration and quantitation of standards and samples.

PCB congener identification and quantitation procedures were based on the methods of Mullin et al. (23) and Mullin (24). A PCB standard of 610 ng/mL was prepared by mixing together three Aroclors: 250 ng/mL Aroclor 1232, 180 ng/mL Aroclor 1248, and 180 ng/mL Aroclor 1262. In this single-point calibration, a retention time window of \pm 0.05 min was used to identify congeners. Relative response factors were calculated for each congener in the calibration standard. Masses of congeners were calculated from known concentrations of congeners in the calibration standard given in the method of Mullin (24). The sum of 41 resolved congeners and 56 unresolved congeners groups gave the mass of total PCB (t-PCB) in the blanks, spikes, and samples. Dividing the mass of t-PCBs by the total volume or mass of the matrix gave the concentration of t-PCBs in the samples.

Quality Control. XAD resin field blank t-PCB mass ranged from 5.5 to 8.1 ng (average 6.6 ± 1.0 ng, n=4), while masses of t-PCBs in samples varied between 33.7 and 121.4 ng. For PUF field blanks, the range of t-PCB mass was 2.0 to 8.3 ng (average 5.2 ± 3.7 ng, n=3) compared to sample masses of 116.7 to 163.3 ng. The filter field blank contained 6.0 ng, and the filter sample contained 40.4 to 165.9 ng. Two solvent blank masses for the fish analyses method averaged 1.5 ng, and the samples varied from 23.2 to 146.1 ng of t-PCBs. The recovery of three PCB congeners not present in environmental samples (IUPAC Nos. 14, 65, and 166) that were added to each sample and matrix blank prior to extraction are summarized in Table 1. Data were not surrogate-corrected.

Detection limits were based on the method of Leister and Baker (20) employed for atmospheric deposition studies. Sample data were reported only if they were three times greater than the blank value of the matrix. All compounds reported exceeded analytical detection limits.

Results and Discussion

Surface Water. Analysis of surface water samples from Lake Tahoe showed similar levels of dissolved t-PCBs at the North

TABLE 2. Summary of t-PCBs in All Compartments of the Lake Tahoe Basin

	date of collection	concentration	concentration
Lake Tahoe water	June 20-22, 1995		
North Station (dissolved)			0.37 ng/L
South Station (dissolved			0.38 ng/L
Marlette Lake water	Dec 18, 1995		
dissolved			0.68 ng/L
particulate			0.60 ng/L
Ward Valley snow	Dec 18, 1995		
dissolved			2.17 ng/L
particle-bound			2.96 ng/L
Ward Valley snow	Dec 28, 1995		"
dissolved			3.3 ng/L
particle-bound_	7 4005		1.56 ng/L
Northshore Lake Tahoe air	July 6-7, 1995		12 1 2
pump A, gaseous			62 pg/m ³
pump B, gaseous	D E 100E		83 pg/m ³
Marlette Lake fish	Dec 5, 1995	ng/g wet wt	ng/g lipid wt
range; mean ± SD	No. 10 1005	$3-11; 6 \pm 3$	$34-231$; 110 ± 44
Lake Tahoe fish	Nov 18, 1995	0 14.12	E/ 100: 70
range; mean		9-14; 12	56-100; 78

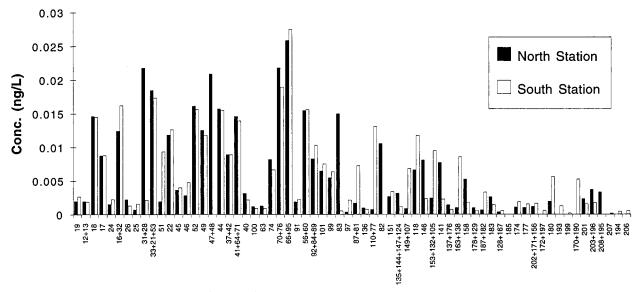


FIGURE 2. Concentration of PCB congeners (dissolved) in surface water from Lake Tahoe at North Station and South Station, June 21–22, 1995.

Station and the South Station (0.37 and 0.38 ng/L, respectively) (Table 2). Particle-phase determinations are not available for these samples. The congener distribution profiles of dissolved PCBs were also nearly identical and dominated by tri- and tetrachlorobiphenyls with congeners 66 + 95, 70 + 76, 52, 44, and 18 being most abundant (Figure 2). Similarity of the levels and distribution of dissolved PCBs at the two locations of sampling suggests that contamination in the lake is uniform and probably not a result of local point sources. Averaged dissolved levels of congeners from the two sampled sites are shown in Figure 3 for comparison to other compartments. Concentrations of total dissolved PCBs in Lake Tahoe were comparable to those reported for Lake Baikal (0.56 \pm 0.18 ng/L) (18) and Lake Superior [(0.55 \pm 0.37 ng/L) (25); (0.32 \pm 0.03 ng/L in 1990; 0.18 \pm 0.02 ng/L in 1992) (26)].

The level of total dissolved PCBs in Marlette Lake was 0.67 ng/L (Table 2), nearly twice the concentration measured in Lake Tahoe water and similar to earlier studies in the Great Lakes (1, 27). In general, the dissolved t-PCB congener profile of Marlette Lake surface water closely matched that of Lake Tahoe, except that some of the more chlorinated congeners (hexa-through octachlorobiphenyls) not detected in Marlette Lake were present in Lake Tahoe water at trace

levels (Figure 4). This may be attributed to the larger sample size taken at Lake Tahoe or to the longer residence time of Lake Tahoe water. Especially noteworthy is that the concentration of many of the lighter weight congeners (18, 17, 16 + 32, 31 + 28, 33 + 21 + 53) were much more pronounced in Marlette water, while the concentration of medium and heavier weight congeners (74-202 + 171 + 156) was approximately equal in both of the lakes. This difference in concentration of PCB congeners may be due to the fact that Marlette Lake is solely atmospherically driven, whereas Lake Tahoe may be receiving contaminants from other minor sources such as general urbanization. Also, although the residence time of Marlette Lake water is not known, it is likely much shorter than Lake Tahoe and thus the surface water probably reflects better what is in its most recent snowpack water sources. In contrast, Lake Tahoe retains its water longer, so that lighter PCB congeners may volatilize while heavier ones accumulate over time. Results of congener patterns and concentrations measured at Marlette Lake decisively establish that airborne transport-deposition is of primary importance for PCB entry to lakes in this region of the Sierra Nevada Mountains.

Similarity of congeneric composition of dissolved PCBs in surface waters from the north and south regions of Lake

0.03

0.025

0.02

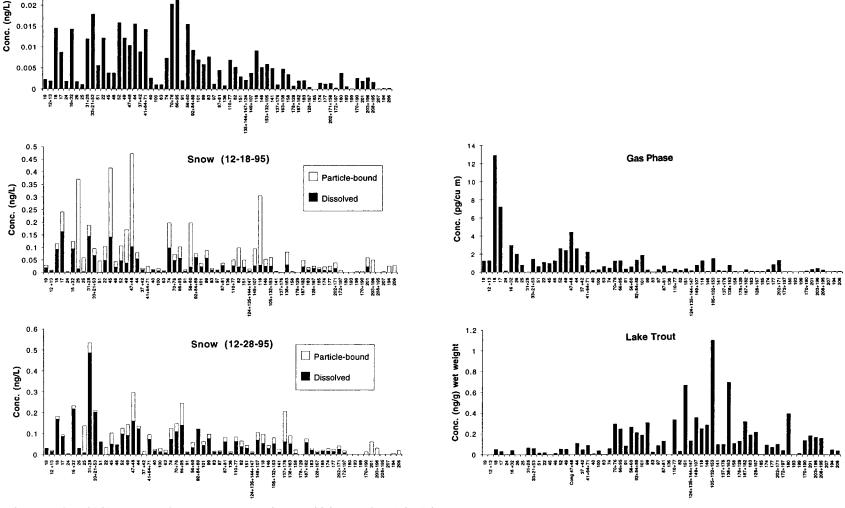


FIGURE 3. Concentration of PCB congeners in water, snow, gas phase, and lake trout from Lake Tahoe.

Surface Water (Dissolved)

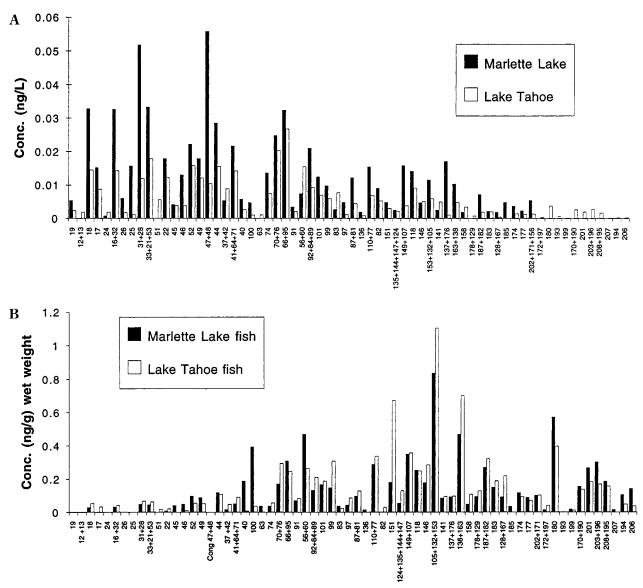


FIGURE 4. Comparison of concentration of PCB congeners measured in Marlette Lake and Lake Tahoe: (A) surface water (dissolved), and (B) fish.

Tahoe and the similarity of the congener distribution of surface water dissolved PCBs of Lake Tahoe and Marlette Lake suggest that loading of contaminants to the two lakes is coming from the same source, the atmosphere. Differences in concentrations of t-PCBs between the two systems (Lake Tahoe, 0.37 ng/L; Marlette Lake, 0.67 ng/L) may be attributed to differences in sampling time, lake depth, water residence time, and particle removal mechanisms. Levels of dissolved PCBs in surface water may also change with season. Lake Tahoe water was sampled in June, and Marlette Lake was sampled in December. Also, the total volume of Lake Tahoe is 350 times greater than that of Marlette Lake, so that chemical pollutants may become considerably diluted in the Lake Tahoe water. In addition, Lake Tahoe, having a lengthy residence time, will retain stable pollutants such as PCBs over a longer period of time relative to Marlette Lake. Sedimentation rates of particles in the water may vary between the two lakes and contribute to the differences in t-PCB concentration. Last, the contrast in the water column concentration of PCBs may be attributed to different sedimentation resuspension rates. Lake depth (Lake Tahoe, 505 m; Marlette Lake, 13 m) may influence the extent to which dissolved PCBs are released over time from sediment into the water column, especially surface water.

Total dissolved PCB concentrations found in surface waters of Lake Tahoe and Marlette Lake were within the range of other lakes, including Lake Baikal and Lake Superior. The roughly 2 times greater total dissolved PCBs in Marlette Lake versus Lake Tahoe finds analogy with atmospheric deposition patterns in the Upper Great Lakes (1, 27). Surface water samples from the smaller reference lake, Siskiwit on Isle Royale in Lake Superior, contained higher t-PCBs than Lake Superior. Swackhamer and co-workers (27) reported that Siskiwit Lake had 2.5 times greater concentration of t-PCBs compared to Lake Superior.

Air. High-volume air samples taken in duplicate on the northern shore of Lake Tahoe (Figure 1) on July 6-7, 1997, had t-PCB vapor concentrations of 62 and 83 pg/m³ with congeners 18, 17, 52, and 49 having the highest levels (Figure 3). Surface water-dissolved PCBs of Lake Tahoe and gasphase PCBs in air samples from the Lake Tahoe basin are high in the tri- and tetrachlorinated biphenyls, suggesting that cycling of these congeners may be occurring between the atmosphere and surface waters. Baker and Eisenreich (21) made this observation for air and surface water samples from Lake Superior.

Vapor phase t-PCB levels measured in Lake Tahoe air are at the lower range of concentrations reported in Lake Superior (110–460 pg/m³) (28) and Green Bay, Lake Michigan (65–2200 pg/m³) (29). Lake Tahoe may be receiving reduced loadings compared to these regions since it is a subalpine lake and is further removed from industrial sources of input. Moreover, high-volume air sampling at Lake Tahoe represents a snapshot picture of contamination levels and is not representative of monthly or annual concentrations such as were determined in the Great Lakes area. Long-term air sampling data from the Lake Tahoe basin needs to be acquired for more detailed comparisons of the two systems.

Snow. Analysis of two snowstorm events sampled on December 18 and 28, 1995, in Ward Valley, located within the Lake Tahoe basin upland from the western shore, showed approximately 7-fold higher levels of dissolved t-PCBs compared to surface water of Lake Tahoe (Table 2). Total PCB concentrations, particle-bound plus dissolved, were similar in the two snow samples (5.1 and 4.8 ng/L), but the distribution of the congeners between the two samples was notably different (Figure 3). The first significant snow event sample contained a larger fraction of the total PCBs associated with the particle phase than the second snowstorm sample. This may have occurred because the December 18 snowstorm, the first significant precipitation event of the season, scavenged more of the available particles from the air. Alternatively, since the December 28 snow sample contained both fresh and aged snow, some redistribution between the particle and surrounding snow and/or ice may have occurred (30). The PCB distribution of congeners in the dissolved fraction in snow was comparable to the surface lake water and favored the lower chlorinated biphenyls (Figure 3).

Few studies have analyzed snow or rain samples in freshwater ecosystems. Precipitation (rain and snow) was analyzed for PCB congeners in Siskiwit Lake, Isle Royale, Lake Superior (1). Average dissolved t-PCB concentrations in rain and snow samples were 13 and 17 ng/L, respectively, and similar to air samples in distribution of the congeners. More recently, however, precipitation monitoring for pesticides and PCBs at three locations around the Great Lakes as part of the Integrated Atmospheric Deposition Network (31) has resulted in much lower PCB concentrations. The annual average t-PCB concentration for 1993 was 1.8 ng/L at Lake Superior, 2.4 ng/L at Lake Michigan, and 0.73 ng/L at Lake Erie with a maximum observed concentration of 7.7 ng/L (31). Conducting a survey of Canadian Arctic snow at lakes, rivers, and coastal stations, Gregor and Gummer (32) reported t-PCB concentrations in the range of 0.02-1.76 ng/ L. Results of both the Great Lakes study (31) and the Canadian Arctic snow measurements (32) compared well with the levels observed at Tahoe in this study. Levels of t-PCB in snow (the sum of particle and dissolved phases) found in the Lake Tahoe basin in 1995 are less than half of those reported in 1976 samples from the Lake Superior basin (1). Significantly, snow samples contained approximately 7-10 times the concentration of dissolved PCBs measured in the surface waters of Lake Tahoe and Marlette Lake. These data correspond to the findings of the Lake Superior study (28) in which precipitation levels were in the order of 6 times that of the surface water concentrations.

Fish. Organic contaminants were measured in two juvenile lake trout from Lake Tahoe, of 12.0 and 13.5 cm length and 400 and 520 g mass. Body burdens of t-PCBs in lake trout were 9 and 14 ng/g wet weight (Table 2). Rainbow trout (*O. mykiss* ssp.) from Marlette Lake, of comparable size and weight to juvenile lake trout sampled from Lake Tahoe, had similar distribution and concentrations of PCB congeners (Figure 4) despite differences in species of fish sampled from the two lakes. Total PCB congeners in Marlette Lake rainbow trout ranged from 3 to 11 ± 3 ng/g wet weight, while for Lake Tahoe the range was 9-14 ng/g wet weight (Table 2). Prominent congeners in fish from both lakes were 118, 146,

180, 105 +132 +153, 138 +163, and 149 +107.

In our previous analysis of adult fish (7-9) years old) from Lake Tahoe ranging in fork lengths of 44.7 to 52.8 cm and weights of 960 to 1400 g (14), average concentrations of total Aroclor residues were roughly 13 times greater than those of immature lake trout of this study (Table 2). These contrasting measurements of residues are most likely due to age differences between the two groups of fish studied since, with increasing age, fish usually acquire more fat tissue and higher levels of pollutants (33).

Exposure from diet may also be significant in explaining differences in contaminant levels. Adult lake trout in Lake Tahoe feed on mysid shrimp and other fish, such as Tahoe suckers (*Catostomus tahoensis*) and Paiute sculpins (*Cottus beldingi*) and occasionally Lahontan redsides (*Richardsonius egregius*), Tui chub (*Gila bicolor*), Kokanee salmon (*Oncorhynchus nerka*), Lahontan speckled dace (*Rhinichthys osculus*) and mountain whitefish (*Prosopium williamsoni*) and even other lake trout (*34*). Juvenile lake trout rely mainly on mysid shrimp for food, thus they receive less exposure from higher trophic species compared to the adult lake trout. Additionally, juveniles, having a faster growth rate than the adult fish, may dilute persistent pollutants in their growing mass (*35*), reflected in a reduced body burden of residues.

Concentrations of t-PCBs determined in fish in this study are similar to those reported for the same species and other species of fish from remote regions of Canada and Europe, such as Lac Brule (mean 5 ng/g wet weight, muscle) and Lac St. Augustine (range, below detection limit to 37 ng/g wet weight, muscle), southern Labrador, Canada (36), Turkey Lakes, Emmett Lake, and Bartley Lake (range 34.6 to 66.3 ng/g wet weight, whole fish), Ontario, Canada (5), and numerous pristine lakes of southern Scandinavia (0.34 to 64 ng/g wet weight, muscle) (35). Values reported in this study are, however, considerably lower than those reported for fish from Siskiwit Lake, Isle Royale, Lake Superior (2), which is closer to areas with past heavy PCB usage, and Lake Baikal in Siberia (18), which has received direct PCB surface discharge for many years. Total PCBs measured in fish from Lake Superior are more than 1 order of magnitude greater than those of the Lake Tahoe basin, even though the dissolved PCB levels in the two water bodies are comparable. An explanation for this inconsistency of water and fish levels may be due to differences in concentrations in food chain levels, age of the fish (33), or sediment loading. Variations in the bioavailability of the chemical to the organism or the efficiency of trophic transfer influence exposure of organic pollutants to fish, and hence fish concentration.

PCB Loading. A simple mass budget of PCBs in Lake Tahoe was constructed to determine the relative importance of atmospheric inputs. Mean annual estimates of tributary/ runoff, direct precipitation, and discharge (Truckee River) from two water budgets (37, 38) were averaged. The concentration of total (dissolved and particulate) PCBs measured in snow (5.0 ng/L) was multiplied by the mean yearly volume of tributary/runoff water (4357 m³) and direct precipitation (3216 m³) to yield annual loads of 2.2 and 1.6 kg, respectively. For the loss of PCB mass by the outflow of the Truckee River, the discharge volume (2975 m³) was multiplied by the concentration of dissolved PCBs in Lake Tahoe surface water (0.37 ng/L) and a conservative estimate of the total particulate PCB concentration of 0.07 ng/L to obtain a total discharge loss of 0.1 kg/year. Gas-exchange and local inputs are more difficult to assess. From the air sample measurements, gas-exchange values were calculated for some of the dominant congeners and showed that the net flux of these PCB congeners is out of the lake in summer. Without more air sample data, we can only conclude that gas-exchange represents a sink. Urban or local sources were assumed to be minimal, given that the Lake Tahoe basin

does not have known industrial discharges. Overall, acknowledging that there are some gaps of knowledge in the construction of this budget, it is apparent that atmospheric sources (direct and indirect) dominate loading of pollutants to Lake Tahoe. Further, the occurrence of PCBs in Marlette Lake surface water and fish confirms the hypothesis that atmospheric transport and deposition processes, rather than local sources, are responsible for much of the loading of PCBs to the Lake Tahoe basin.

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Literature Cited

- (1) Swain, W. R. J. Great Lakes Res. 1978, 4, 398.
- (2) Swackhamer, D. L.; Hites, R. A. Environ. Sci. Technol. 1988, 22, 543
- (3) Haines, T. A. Water, Air, Soil Pollut. 1983, 20, 47.
- (4) Heit, M.; Klusek, C.; Baron, J. Water, Air, Soil, Pollut. **1984**, 22, 403
- (5) Johnson, M. G.; Kelso, J. R. M.; George, S. E. Can. J. Fish. Aquat. Sci. 1988, 45, 170.
- (6) Jassby, A. D.; Reuter, J. E.; Axler, R. P.; Goldman, C. R.; Hackley, S. H. Water Resour. Res. 1994, 30, 2207.
- (7) Laird, L. B.; Taylor, H. E.; Kennedy, V. C. Environ. Sci. Technol. 1986, 20, 275.
- (8) Leonard, R. L.; Goldman, C. R.; Likens, G. E. Water, Air, Soil Pollut. 1981, 15, 153.
- (9) McColl, J. G. J. Environ. Qual. 1988, 17, 425.
- (10) Zabik, J. M.; Seiber, J. N. J. Environ. Qual. 1993, 22, 80.
- (11) Cory, L.; Fjeld, P.; Serat, W. Pestic. Monit. J. 1970, 3, 204.
- (12) Drost, C. A.; Fellers, G. M. Conserv. Biol. 1996, 10, 414.
- (13) Knapp, R. A. In *Status of the Sierra Nevada;* Sierra Nevada Ecosystem: Final Report to Congress, Science Team Special Consultants, Eds.; Centers for Water and Wildland Resources, University of California: Davis, CA, 1996; Vol. 3.

- (14) Datta, S. Ph.D. Dissertation, University of California, Davis, CA, 1997.
- (15) Goldman, C. R. Limnol. Oceanogr. 1988, 33, 1321.
- (16) Sater, E. M. M.S. Thesis, University of California, Davis, CA, 1994
- (17) Ko, F. C.; Baker, J. E. Mar. Chem. 1995, 49, 500.
- (18) Kucklick, J. R.; Bidleman, T. F.; McConnell, L. L.; Walla, M. D.; Ivanov, G. P. *Environ. Sci. Technol.* 1994, *28*, 31.
- (19) Majewski, M. U.S. Geological Survey, Sacramento, CA. Personal communication, 1995.
- (20) Leister, D.; Baker, J. E. Atmos. Environ. 1994, 28, 1499.
- (21) Baker, J. E.; Eisenreich, S. J. J. Great Lakes Res. 1989, 15, 84.
- (22) Ribick, M. A.; Dubay, G. R.; Petty, J. D.; Stalling, D. L.; Schmitt, C. J. Environ. Sci. Technol. 1982, 16, 310.
- (23) Mullin, M. D. Presented at PCB Workshop, U.S. EPA Large Lakes Research Station, Grosse Ile, MI, June 1985.
- (24) Mullin, M. D.; Pochini, C. M.; McCrindle, S.; Romkes, M.; Safe, S. H.; Safe, L. M. *Environ. Sci. Technol.* 1984, 18, 468.
- (25) Baker, J. E.; Eisenreich, S. J. Environ. Sci. Technol. 1990, 24, 342.
- (26) Jeremiason, J. D.; Hornbuckle, K. C.; Eisenreich, S. J. *Environ. Sci. Technol.* **1994**, *28*, 903.
- (27) Swackhamer, D. L.; McVeety, B. D.; Hites, R. A. Environ. Sci. Technol. 1988, 22, 664.
- (28) Hornbuckle, K. C.; Jeremiason, J. D.; Sweet, C. W.; Eisenreich, S. J. Environ. Sci. Technol. 1994, 28, 1491.
- (29) Achman, D. R.; Hornbuckle, K. C.; Eisenreich, S. J. *Environ. Sci. Technol.* 1993, 27, 75.
- (30) Hoff, J. T.; Wania, F.; Mackay, D.; Gillham, R. Environ. Sci. Technol. 1995, 29, 1982.
- (31) Sweet, C. W.; Harlin, K.; Gatz, D. F.; Bauer, S. Great Lakes Integrated Atmospheric Deposition Network (IADN) Data Report 1993–1994; Illinois State Water Survey, Champaign, IL, March 1996; p 152.
- (32) Gregor, D. J.; Gummer, W. D. Environ. Sci. Technol. 1989, 23, 561.
- (33) Larsson, P.; Backe, C.; Bremle, G.; Eklor, A.; Okla, L. Can. J. Fish. Aquat. Sci. 1996, 53, 62.
- (34) Allen, B. Tahoe Research Group, University of California, Davis, CA. Personal communication, 1996.
- (35) Larsson, P.; Collvin, L.; Okla, L.; Meyer, G. Environ. Sci. Technol. 1992, 26, 346.
- (36) Lockbrie, D. M.; Clair, T. A. Bull. Environ. Contam. Toxicol. 1988, 41, 625.
- (37) Lind, R. A.; Goodridge, J. D. Lake Tahoe Water Balance. Presented at the Conference on Sierra Nevada Meteorology, June 19–21, 1978, South Lake Tahoe, CA, American Meteorological Society.
- (38) Myrup, L. O.; Powell, T. M.; Godden, D. A.; Goldman, C. R. Water Resour. Res. 1979, 15, 1499.

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