

# Contaminant Concentrations in Adirondack Soil and Sediment

By JEFFREY CHIARENZELLI, J. PAGANO, M. MILLIGAN, T. HOLSEN, J. GLOVAK, AND K. ALTHOUSE

## Abstract

*Paired soil and sediment samples were collected along a transect across northern New York from Lake Ontario to the Tug Hill Plateau to Indian Lake in the Central Adirondack Mountains. The samples were analyzed for four classes of compounds including polychlorinated biphenyls, polycyclic aromatic hydrocarbons, select chlorinated pesticides, and inorganic elements and metals with different origins, usage history, and chemical properties. In general, the concentrations of these contaminants were higher in soil than sediment but relatively low compared to other more developed areas, consistent with the remoteness of the Adirondack region and limited permanent population. Trends in the data suggest that the concentrations of contaminants, particularly high molecular weight organic compounds, are generally greater closer to Lake Ontario. The reason(s) for this remain unclear, but are likely related to the large amounts of lake-effect precipitation the Tug Hill area receives and scavenging of contaminants by rain and snow. Levels reported for contaminants investigated in this study are generally lower than those reported elsewhere in the United States, and likely reflect the low population density of the region and less intensive land use.*

*Jeffrey Chiarenzelli is with the Departments of Geology at St. Lawrence University and Clarkson University; J. Pagano, the Department of Chemistry, SUNY Oswego; M. Milligan and J. Glovak, the Department of Chemistry, SUNY Fredonia; T. Holsen, the Department of Civil and Environmental Engineering, Clarkson University; and K. Althouse, the Department of Geology, SUNY Potsdam.*

## Introduction

The far-reaching impact of anthropogenic activities has been convincingly documented in the world's remote regions including the poles (Corsolini et al., 2006) and mountain tops (Blais et al. 1998). One consequence of the modern human footprint is the global scale redistribution of chemical compounds (Wania and MacKay, 1996). These compounds include both natural materials (e.g. metals, inorganic elements, ions) and manufactured compounds (e.g. pesticides, dioxins, polychlorinated biphenyls) not normally produced by natural processes. Regardless of their origin, anthropogenic activity has led to the redistribution of these compounds and many others on a scale few might have anticipated just several decades ago (cf. Rappe, 1974).

Parts of the Adirondack Park are considered by many as some of the most remote areas in the United States east of the Mississippi River. This distinction is further advanced by regulated land-use categories that include 'wilderness areas', encompassing over one million acres and 18.4% of the Adirondack Park, where human activity is strictly controlled (Adirondack Park Agency, 2001). Nonetheless, the region is a near equal mixture of public and private land, surrounded by large population centers, and has millions of visitors a year (Jenkins, 2004). Through atmospheric deposition the area receives far-traveled contaminants including acidic species, mercury and other metals, and persistent man-made organic contaminants, some of which may ultimately be derived from areas many thousands of kilometers away (Simonin and Meyer, 1998). However, intensive forestry, mining, agriculture, and tourism-related activities all take place within and peripheral to the Blue Line (Jenkins, 2004) and could

conceivably result in local sources adding to contaminant loading.

Four different classes of compounds were selected for analysis because of their different origins, behavior in the environment, production and usage histories, and possible links to anthropogenic activities within, or adjacent to, the Park. These include polychlorinated biphenyls (PCBs), chlorinated pesticides (including mirex, hexachlorobenzene (HCB), dichlorodiphenyltrichloroethane (DDT) and its metabolites DDD and DDE), polycyclic aromatic hydrocarbons (PAHs), and metals. A detailed discussion of these compounds, their behavior in the environment, and possible health implications is beyond the scope of this paper but can be found in literature and websites (e.g. Morrison and Murphy, 2006; ATSDR website - <http://www.atsdr.cdc.gov/>).

These contaminants have long been known to have a variety of adverse health impacts on humans and animals (e.g. Robertson and Hansen, 2001). For example, the toxicity of DDT was widely publicized by Rachel Carson in 1962 in her seminal book on environmental toxins *Silent Spring*. In general, toxicity increases with concentration in the environmental media and through synergistic effects (the net effect of two or more compounds acting in concert). People and animals are exposed through ingestion of food and water, inhalation, and, in some cases, through dermal contact. Exposure to elevated levels of most of these contaminants is typically related to proximity to a potential source or dietary choices.

Most of these contaminants bioaccumulate and tend to be sequestered in fatty tissues because of their solubility in lipids; however, some like mercury are found in muscle tissue. All are generally found in highest concentrations in animals at the

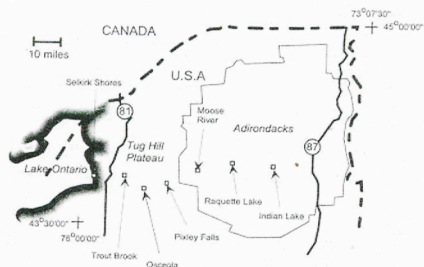


Figure 1. Sample location diagram for this study showing northern New York from Lake Ontario to the Central Adirondacks.

top of the food chain. Thus the fat and milk of top predators such as seals, walrus, bears, and humans often contain unsafe levels of many of these contaminants when exposure rates are high through ingestion (Dewailly et al., 1989). All of us carry a body burden of contaminants that serve as a fingerprint of where we live and what we eat. Some of the contaminants are readily passed on from mothers to offspring or eggs (Pagano et al. 1999). How these contaminants move through the environment, and their impacts and fate have been the subjects of numerous studies.

The objectives of study were as follows: 1) to provide an initial survey of baseline concentrations of several classes of contaminants in paired soil and sediment samples across an east west transect (Figure 1) across northern New York from Lake Ontario to Indian Lake; 2) compare the concentrations of contaminants found in the Adirondacks to other areas; and 3) determine if any spatial trends in the contaminant mixture (fingerprint) occur across the sampling traverse.

### Methodology

Seven sampling locations forming a 160-kilometer east west transect across northern New York were selected (Figure 1, Table 1). The transect extended from the eastern shore of Lake Ontario to the central Adirondacks near Indian Lake. Each locality was selected to allow sampling of soil and stream sediment within close proximity. To minimize differences in substrate composition extensive deposits of glacial lacustrine or outwash sands in undisturbed, open areas were chosen as sampling sites. In all cases sampling were collected >100 yards from roads to minimize the impact of automobile traffic.

Sandy soil and sediment was collected using a stainless steel trowel and immediately sealed in pre-cleaned glass jars. The jars were wrapped in aluminum foil and chilled in a cooler. Both soil and sediment were taken from the upper 2-3 centimeters of substrate. Any extraneous organic matter (e.g. leaves, roots, twigs, etc.) was removed. Our sampling goal was to obtain soil and sediment samples representative of the area with similar grain-size distribution (primarily sand) and organic content for comparative purposes.

Individual samples were homogenized and split into subsamples at SUNY Potsdam. The Environmental Research Center at SUNY Oswego (J. Pagano) carried out PCB and select pesticide analysis by gas chromatography with an electron capture detector (GC-ECD); PAH and DDT analysis was carried out at the Environmental Chemistry Laboratory at SUNY Fredonia (J. Glovak and M. Milligan) by gas chromatography mass spectrometry (GC-MS). Major and trace element analysis was carried out at ACME Analytical Laboratories in Vancouver, British Columbia utilizing inductively coupled mass spectrometry (ICP-MS) and organic carbon was determined by LECO analysis. Grain size analysis and moisture content were measured at SUNY Potsdam (K. Althouse and J. Chiarenzelli). Except for samples utilized for grain-size analysis and moisture content, all samples were analyzed without drying or size splitting.

Analysis of organic compounds is fairly complicated and the details of the analytical procedures are published elsewhere (Hopke et al., 2003; Chiarenzelli et al., 2001, 2002; Pagano et al., 1999). In general, it involves the extraction of the analytes of interest from a mass of sediment, cleaning of the extract to remove compounds causing potential analytical interference, and condensing of the final solvent carrier. The sample is then injected into a gas chromatograph and passes through a long thin (capillary) column utilized to separate related compounds of different molecular weights by retention time (how long it takes them to travel through the column). Further separation can be employed using a mass spectrometer that allows definitive identification of compounds by their mass in addition to retention time.

The analysis of inorganic elements and metals in geologic samples is more straightforward and involves the digestion of powdered samples. Homogenized samples of rock powder were analyzed by ICP-ES (major elements) and ICP-MS (trace elements) after lithium metaborate/tetraborate fusion and nitric acid digestion. A separate split for base metals utilized aqua regia digestion after fusion. During ICP-MS analysis the sample is injected into a stream of plasma where all bonds are broken and the characteristic mass to charge ratio of each element and/or ion can be measured as intensity on a mass spectrometer.

Table 1. Information on soil and sediment samples analyzed for this study.

Location	Latitude	Longitude	Altitude	%OC		%fines	
				soil	sediment	soil	sediment
Selkirk Shores State Park	N43° 33.244'	W76° 12.774'	298'	0.31	0.1	2	2
Trout Brook Fishing Area	N43° 32.730'	W76° 02.761'	545'	2.7	0.14	8	1
Osceola (Salmon River)	N43° 27.683'	W75° 35.890'	1298'	0.79	0.22	3	0
Pixley Falls State Park	N43° 24.202'	W75° 20.567'	944'	5.05	0.18	1	2
Moose River	N43° 36.777'	W75° 06.090'	1501'	0.28	0.1	1	0
Raquette Lake Picnic Area	N43° 48.497'	W74° 36.146'	1810'	0.18	0.09	0	0
Indian Lake Public Beach	N43° 47.494'	W74° 14.031'	1642'	1.96	0.44	6	1

OC = organic carbon

% fines = silt + clay fraction, the remainder consists entirely of sand

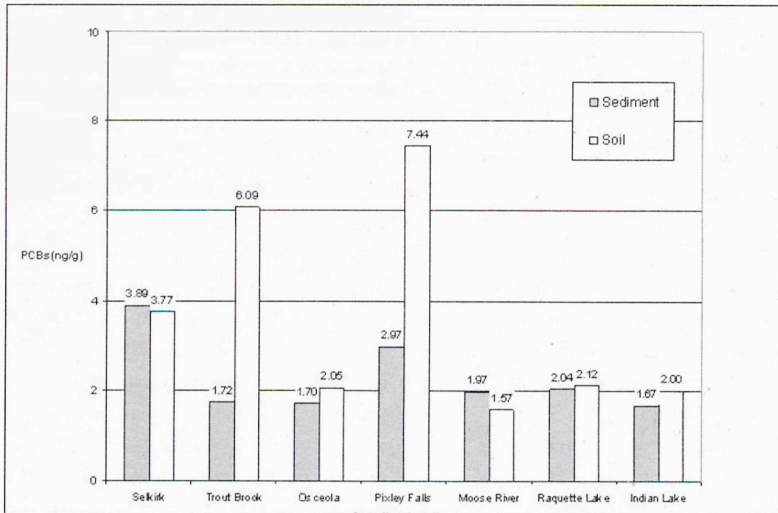


Figure 2. PCB concentrations in sediment and soil from west to east across sampling traverse.

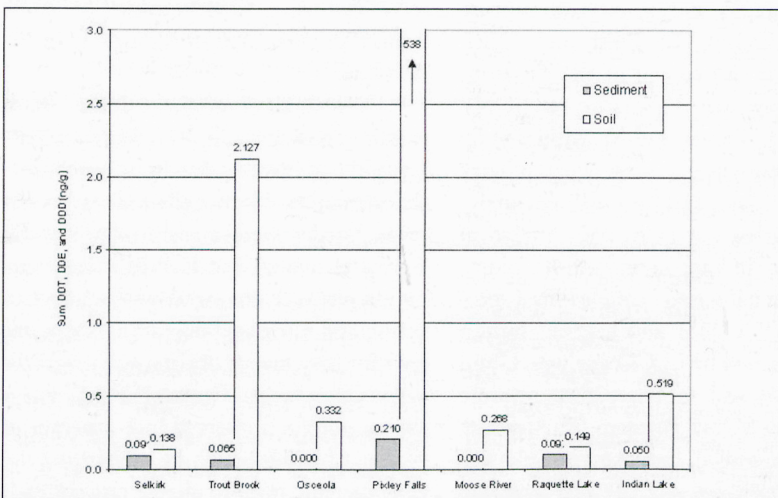


Figure 3. The sum of DDT, DDE, and DDD concentrations in sediment and soil from west to east across the sampling traverse. Note that the DDE concentration of the soil sample from Pixley Falls is > 500 ng/g and plots well above the upper limit of the diagram.

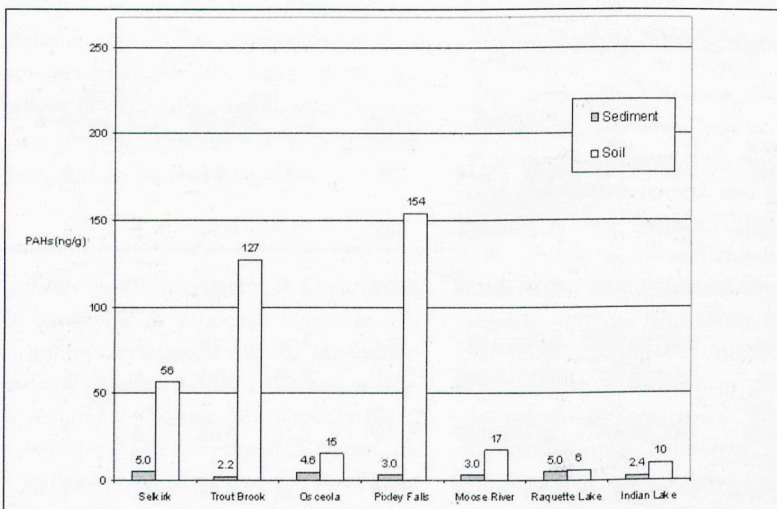


Figure 4. PAH concentrations from in sediment and soil west to east across the sampling traverse. The greatest concentration at the top of the diagram is 267 ng/g, the National Median value for sediment from 46 drainage basins NWQAP studies (Chalmers, 2002).

As with all analytical work, a number of steps and samples were taken to insure quality control. In this study quality control was particularly important given complicated subsampling procedures and shipping of samples to various laboratories. The quality control samples include blank samples, reagent blanks, method blanks, duplicate samples, the addition of surrogate compounds and internal standards, and the repeat analysis of certified standards with the samples. In addition, we also compared analytical methods for the analysis of DDE in two different laboratories by different analytical techniques. In all cases the quality control samples indicated that our work was precise and accurate, exceeding standards typically applied for quality control in studies of this type.

For each class of analytes (PCBs, Chlorinated pesticides, PAHs, and metals) at least two samples were duplicated to test reproducibility. For example, PCBs analysis was duplicated for sediment from Indian Lake (1.41 and 1.43 ng/g) and soil from Pixley Falls (6.17 and 5.19 ng/g). PCB congener specific patterns were nearly identical for duplicate samples ( $R^2 > 0.9995$ ). For DDT and its metabolites (DDD and DDE), ten duplicate samples were analyzed and concentrations generally were within 80%. The reproducibility of metals was also high, generally >95%, except for trace elements at small concentrations (ppb) which were all >90%. These results indicate that the results are reproducible and the data of high quality; however, the relatively small number of samples limits the use of statistical comparisons.

Method blanks for PCB analysis ( $n = 4$ ) ranged from 1.29 – 2.01 ng/g with a mean 1.81 +/- 0.35. Method blanks represent the sum total of the analyte in question introduced into the sample during handling, transport, extraction, and analysis. Several of the samples analyzed had PCB concentrations at or below what could be introduced during the analysis; essentially indicating that the PCB concentrations were at or below our effective detection limit in some samples.

Sixteen PAHs were analyzed in this project, ranging from acenaphthylene (three aromatic rings) to coronene (seven aromatic rings). Naphthalene (two aromatic rings) was measured in all samples, but in many cases was present in concentrations statistically below what was measured in method blanks. As a result, naphthalene has not been included in the sum. For the PAH analyses, sample concentrations were well above those measured in method blanks. For the sixteen PAH compounds measured in this project, the method blank total concentrations ranged from 0.2 – 0.7 ng/g ( $n = 2$ ), with an average of about 0.5 ng/g. All of the soil and sediment samples analyzed for this work had total PAH concentrations of at least

five times the method blank average. As a result, we believe that the PAH concentrations reported here are accurate representations of the environmental burdens, and accordingly have not been blank-corrected.

**Results and Discussion**

*Polychlorinated Biphenyls (PCBs)* – The concentration of polychlorinated biphenyls (PCBs) ranged from 1.57-7.44 ng/g (parts per billion - dry weight) in soil samples and from 1.67-3.89 ng/g (dry weight) in sediment samples. In all but two sample locations (Selkirk and Moose River) the soil had more PCBs than the corresponding sediment sample (Figure 2). In general, as discussed below, the concentrations measured are relatively low but higher west of the Adirondack Park.

*Chlorinated pesticides* – Several recalcitrant chlorinated pesticides and chlorinated byproducts were analyzed during the study. These include Hexachlorobenzene (HCB), Mirex, and DDT and its degradation products DDD, and DDE. As is the case with PCBs, these chlorinated compounds are found in considerably higher concentrations in soil than sediment samples. For example, Mirex was detected in four of the soil samples but in only one sediment sample. In general, the concentrations of each of these compounds is low (HCB = ND (not detected) – 0.097 ng/g; Mirex = ND – 0.300 ng/g; DDE = ND – 1.756 ng/g; DDD = ND – 0.155; and DDT = ND – 0.655). The soil sample from Pixley Falls had relatively high concentrations of DDE (538 ng/g) and is considered an anomaly for reasons presented later (Figure 3).

*Polycyclic Aromatic Hydrocarbons (PAHs)* – The concentration of PAHs ranged from 5.16-139.65 ng/g (dry weight) in soil samples and from 1.58-4.03 ng/g (dry weight) in sediment samples (Figure 4). In each location soil samples had more PAHs than sediment samples; at some locations (Trout Brook and Pixley Falls) soil contained more than 50x the amount of PAHs as paired sediment samples. In general, these concentrations are relatively low and this was particularly true for Adirondack sampling locations (Moose River, Raquette Lake, and Indian Lake).

*Organic Carbon, Inorganic Elements, and Trace Metals* – Comparison of total carbon with organic carbon analyses indicated that nearly all of the carbon was in organic form. Soil organic carbon (Table 1) concentrations (1.61 +/- 1.79%) were significantly higher than sediment concentrations (0.18 +/- 0.11%). The concentrations of

major and trace elements in paired soil and sediment where nearly identical; however, a slight enrichment in most trace metals was found in the soil samples (Figure 5a). The highest concentrations of some elements of anthropogenic interest, such as Pb, are found in the Tug Hill Plateau region (Figure 5b).

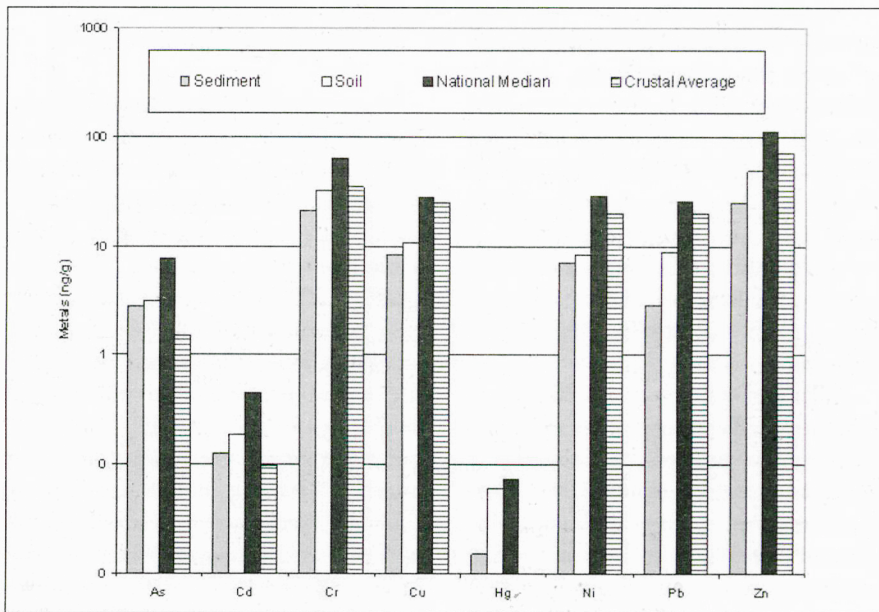
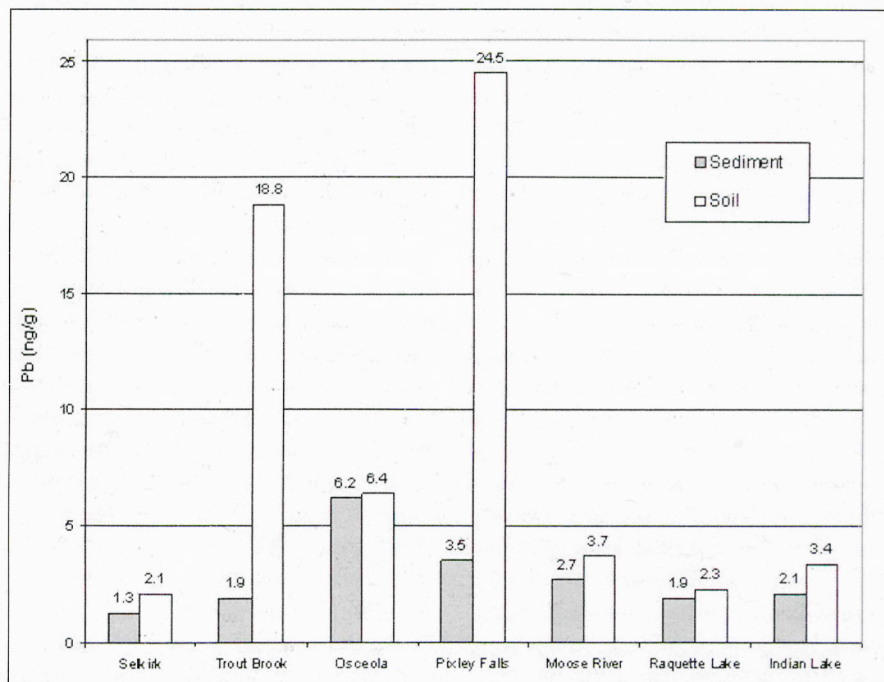


Figure 5. Select metal concentrations from west to east across the sampling traverse; above: a) comparison of sediment and soil samples from this study with National Median values for sediment (NWQAP studies; Chalmers, 2002) and average upper continental crust (Taylor and McLennan, 1985); an average crustal value for mercury was not determined. Note that the y-axis is logarithmic; below: b) variation of lead in soil and sediment across the sampling traverse. The greatest concentration at the top of the diagram is 25.9 ug/g, the National Median value for sediment from 46 drainage basins NWQAP studies (Chalmers, 2002).



*Baseline Concentrations of Contaminants in the Transect* – Concentrations of virtually all contaminants measured in this study are at low levels (Chiarenzelli et al. 2002; see Figures 3, 4, and 7) consistent with the geographic and demographic factors in the area of the transect. These factors include relatively large distances to population and industrial centers, limited use of these compounds in the area, and prevailing wind directions. It should be noted that the three Adirondack sampling locations were within a major west-east traffic corridor through the Central Adirondacks (Route 28). Presumably samples taken in wilderness areas would have even smaller concentrations that could only be effectively measured by increasing the sample size and/or reducing method blanks and detection limits utilized in this study.

PCBs and other banned chlorinated compounds and their derivatives were measured at a few parts per billion (ng/g) or below at Adirondack sampling sites. These concentrations are slightly above, at, or below method blank and/or detection limits. Some compounds, such as Mirex, produced in relatively small amounts for a limited timeframe, and although detected elsewhere in the traverse (e.g. Tug Hill) were not detected in the Adirondack samples. In general, concentrations of these compounds in soil, where detected, are generally slightly higher than in corresponding sediment samples. In general, contaminants show a correlation with organic carbon content and the soil samples had more organic carbon. Polycyclic aromatic hydrocarbons were found in each of the soil and sediment samples, reflecting their continuous production by combustion and natural processes throughout the region (Tan and Heit, 1981). Nonetheless concentrations are fairly low with maximum values of 154 ng/g measured in soil at Pixley Falls, a State Campground. This may be a function of historic and current PAH production in campfires, charcoal grills, or idling automobiles.

The concentrations of metals of anthropogenic interest are also fairly low (e.g. Chalmers, NAWQA, 2002); ranging from a low parts per million for As, Cr, Cu, Ni, Pb, and Zn, and low part per billion concentrations for Cd, and Hg. In addition, the chemical leach data (not presented

here) suggests much of the metals measured in the samples are tightly bound within mineral grains, requiring digestion to liberate them, rather than as adsorbed particles on the surface of mineral grains. As with the other contaminants, slightly more metals were found in soil than sediment and in the Tug Hill samples relative to the Adirondack samples.

*Comparison of Contaminant Concentrations with Other Areas* – Samples collected along the traverse generally have relatively low concentrations for all the analytes measured, consistent with the rural to remote locations selected for sampling (Chalmers, NAWQA, 2002; Chiarenzelli et al., 2002). Pixley State Park is the sole exception. Soil from Pixley State Park has fairly high (> 0.5 ug/g or ppm) concentrations of DDE. It is possible that its precursor, DDT, was utilized at the park in the past for mosquito control. This was a common practice in many areas of the State prior to the ban of DDT and those old enough may remember the fogging trucks or lawn mower foggers utilized to distribute the pesticide. The ratio of DDT to DEE in the samples is small and suggestive of an older, weathered source, rather than recent and illegal use of banned DDT based pesticides.

Polychlorinated biphenyls concentration in soil and sediment can be broadly divided into impacted or urban areas, rural areas, and remote areas. The PCB concentrations measured in this study are 7 ng/g and less and fall within the range most often found in rural to remote areas (Figure 2; Chiarenzelli et al., 2002). For example, recent fluvial sediment samples from Rice Creek in Oswego, New York and the Salmon River Reservoir in Redfield, New York have PCB concentrations from 2-5 ng/g (Chiarenzelli et al., 2001; 2002). Much greater concentrations, often measured in tens or hundreds of parts per million, are found in sediments impacted by PCB disposal or leakage (Sokol et al., 1994). In the National Water-Quality Assessment Program (Chalmers, NAWQA, 2002) conducted from 1991-1997 in 46 watersheds only two samples exceeded the 50-100 ng/g detection limits reported for PCBs. A composite sample of sediment from the New England Coastal Basin

(NECB) covering an area of 23,000 mi<sup>2</sup> had 155 ng/g of PCBs and that of the Oahu drainage basin on Hawaii had 80.9 ng/g. One study with lower detection limits yielded a concentration of 8.3 ng/g PCBs in the Potomac drainage basin. Adirondack samples had PCB concentrations considerably lower than measured in these drainage basins and often indistinguishable from the method blank values determine for our study (~2 ng/g).

The sum of DDT compounds (DDT+DDE+DDD) in stream sediments from the NAWQA study ranged from less than <2 ng/g to 25.9 ng/g. The highest value was measured in the densely populated NECB watershed. In this study, the sum of the same compounds in sediment sampled varied from 0.0 to 0.210 ng/g (Figure 3). Sediment values for DDT, DDD, and DDE are also low or non-detectable in the Adirondacks but slightly higher than in corresponding soil samples and in the Tug Hill Plateau.

Among the basins studied by NAWQA, polyaromatic hydrocarbons ranged from <50 – 21,764 ng/g. The national median for sediment concentrations of PAHs was 267 ng/g. All sampling sites in this study were well below the national median ranging between 2.2 – 5 ng/g and below most other drainage basins reported. Soil samples, considerably enriched above paired sediment samples, were also well below (6-154 ng/g) the national median value for sediment (Figure 4).

With very few exceptions (n = 4) all of the soil and sediment samples collected in this study were well below national median values for trace metals (Chalmers, NAWQA, 2002). Trace metals analyzed included many of anthropogenic interest including arsenic, cadmium, chromium, copper, mercury, nickel, lead, and zinc. In fact heavy metals in Adirondack and Tug Hill samples are among the lowest in the 46 drainage basins investigated by the NAWQA (Figure 5a, third set of columns from left). Further, the metals concentrations were usually below values for the upper continental crust (Figure 5a, fourth set of columns from left) determined by Taylor and McLennan (1985) and frequently used to identify anomalous metal concentrations for exploration or environmental purposes.

These data suggest that contaminant levels across the sampling traverse are consistently low for analytes measured in this study compared to a number ( $n = 46$ ) of drainage basins throughout the country. This is consistent with the land-use and population density in the Adirondacks. Previous studies have shown direct correlations between contaminants and population density and urbanization (Chalmers, NAWQA, 2002). Another predictor of contaminant concentrations is the amount of organic carbon in the sediment samples. The sediment samples in this study fall also well below all of the other drainage basins in terms of their content of organic carbon (0.09-0.44% vs. National Median of 2.5%).

*Spatial Trends in the Data* – Although concentrations of all contaminants are uniformly low throughout the sampling traverse, some intriguing trends in the data occur. These trends may be suggestive of regional meteorological processes controlling the deposition of contaminants, however, further work is needed to increase sample numbers and enable statistical testing of the preliminary hypotheses mentioned here. In general, the contaminant concentrations in the Adirondack portion of the sampling traverse were less than in the western portion (Tug Hill and Lake Ontario). This is most apparent for man-made organic contaminants such as PCBs, PAHs, and chlorinated pesticides. Adirondack samples were often at or below detection limits for these compounds. The rise and cooling of moist air masses passing over Lake Ontario and resulting precipitation may result in the preferential deposition of contaminants closer to the lake (Chiarenzelli et al., 2002).

In addition, spatial trends in the proportions of compounds (congeners or isomers) that make up the environmental mixture observed in the samples also vary. For PCBs there is a weak trend (not statistically validated) for heavier compounds (greater molecular weight) to be enriched closer to Lake Ontario and for mixtures to become progressively lighter as one travels eastward into the Adirondacks (Figure 6). These trends are most apparent in soil samples that generally have greater concentrations of contaminants than corresponding

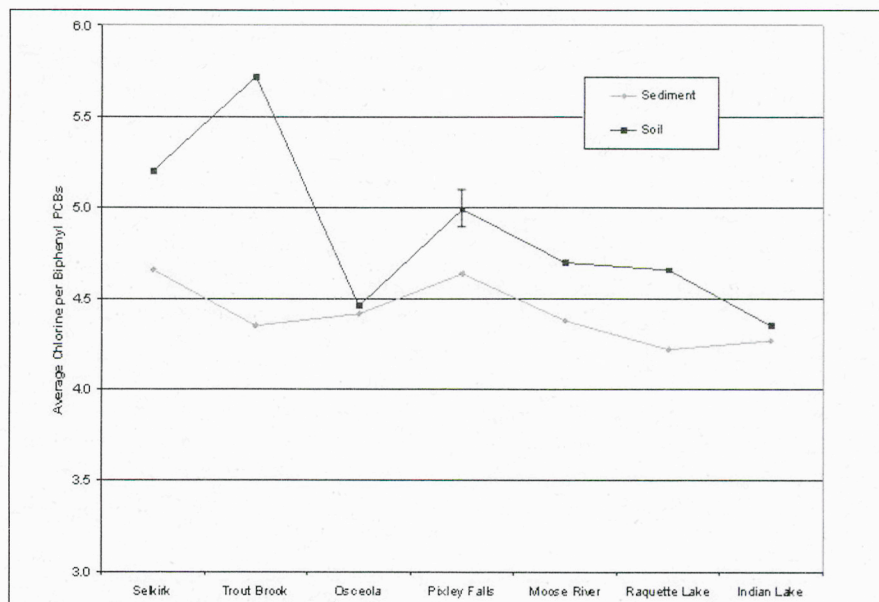


Figure 6. PCB average chlorine to biphenyl ratio in sediment and soil samples across sampling traverse from west to east.

sediment samples. Soil, as a residual deposits, develops and accumulates material for many years whereas stream sediment samples are often rapidly deposited and are subjected to burial or periodic reworking, and thus generally represent only restricted intervals of time. In addition, soil samples analyzed in this study have significantly more organic carbon than the sediment samples, which is often correlated with contaminant concentrations and hydrophobic compounds (Chalmers, NAWQA, 2002).

A “heavier” signature in the PCB mixtures observed suggests distinct changes in the proportion of individual congeners or isomers making up the mixtures (fingerprint) found in the environmental samples (Morrison and Murphy, 2006). In the case of PCBs, the mixtures with heavier signatures tend to be enriched in highly chlorinated congeners. This can be measured by determining the average number of chlorines per biphenyl molecule in a given environmental mixture. Figure 6 shows the trends in soil and sediment in terms of the average chlorine per biphenyl molecules across the sampling traverse. It should be noted that the trends displayed in Figure 6 are based on a limited number of samples; more data are needed to confirm the trend noted.

The PCB fraction in these samples is dominated by recalcitrant congeners that

are less mobile and have been shown to resist degradation in the environment and by metabolic processes within animals (James, 2001; Chiarenzelli et al., 2001). They represent a “weathered” fraction that has accumulated despite degradation and physical redistribution processes such as volatilization, solubilization, oxidation, and biodegradation. Since lighter PCB molecules (those with less chlorine) have a greater tendency to remobilize, soil samples are dominated by residual congeners.

### Summary

The following key points emerge from the data presented:

Contaminant (PCBs, chlorinated pesticides, PAHs, and metals) concentrations in paired sediment and soil samples are similar but low in the Tug Hill and Adirondack regions probably because of limited population density and relative geographic isolation. As might be predicted from their geography, Adirondack samples tend to have concentrations lower than those from the Tug Hill region. Factors such as elevation and watershed geology that may be important in determining the impact of acidic precipitation or mercury deposition appear to have limited influence on these classes of contaminants.

Despite active forestry, mining, and tourism activities within the Park, the contaminant concentrations measured are

among the lowest in the Nation, based on comparison with available databases and the literature. However, this also suggests that some contaminants not used in great abundance in the Park (e.g. PCBs, chlorinated pesticides) are likely derived from atmospheric transport from sources external to the region. Unfortunately despite limited local input of contaminants, the environmental health of the Adirondacks depends primarily on decisions and actions taken outside of the Adirondacks. Amelioration of existing problems such as acidic precipitation (Driscoll et al., 2003) and mercury deposition (Simonin and Meyer, 1998) are thus also dependent on external factors.

Spatial trends suggest that within PCB mixtures "heavier" molecules are relatively enriched in samples taken along the western end of the sampling traverse closer to Lake Ontario. One possible reason for this is the path of regional air masses from west to east over population centers and the scavenging effect of lake-effect precipitation (Franz and Eisenreich, 2000). Fine particles and their adsorbed contaminants in air may be partially removed by precipitation in the Tug Hill Region, effectively cleaning air masses prior to their movement over the Adirondack Region. Because the Tug Hill Region is closer to population centers and receives large amounts of lake-effect precipitation, it may serve as a harbinger of environmental change and foreshadow trends in contaminant deposition in northern New York. If so, the Tug Hill Plateau may provide important context for future contaminant studies in the Adirondack Park.

To our knowledge this is the first study of this kind conducted in the Adirondacks and perhaps elsewhere. Comparison of soil and sediment samples taken in the same area suggests soil, a residual deposit, tends to accumulate more contaminants, although this may also be a function of organic carbon content. The organic contaminants in soil have a highly "weathered" signature due to physical, chemical, and/or biological processes operating in the soil. However, because of their highly recalcitrant nature and resistance to redistribution the compounds remaining may be preserved in the soil profile for perpetuity.

### Acknowledgments

The authors would like to gratefully acknowledge financial support from the Great Lakes Research Consortium.

### References

- Adirondack Park Agency, 2001. Adirondack Park State Land Master Plan 2001 ([http://www.apa.state.ny.us/Documents/Laws\\_Regs/SlmpPDF2001.pdf](http://www.apa.state.ny.us/Documents/Laws_Regs/SlmpPDF2001.pdf) and <http://www.apa.state.ny.us/gis/colc0303.htm>).
- Blais, J., Schindler, D., Muir, D., Kimpe, L., Donald, D., and Rosenberg, B., 1998. Accumulation of persistent organochlorine compounds in mountains of western Canada. *Nature*, v. 395, p. 585-588.
- Carson, R., 1962. *Silent Spring*. Houghton Mifflin Company, Boston, MA, 368 p.
- Chalmers, A., 2002. Trace elements and organic compounds in streambed sediment and fish tissue of coastal New England streams, 1998-99: National Water-Quality Assessment Program: U.S. Geological Survey, Water Resources Investigations Report 02-4179.
- Chiarenzelli, J., Alexander, C., Scudato, R., Pagano, J., Falanga, L., Connor, B., and Milligan, M., 2002. Anomalous concentrations and chlorination of polychlorinated biphenyls in sediment downwind of Lake Ontario. *Journal of Great Lakes Research*, v. 28, p. 674-687.
- Chiarenzelli, J., Alexander, C., Isley, A., and Scudato, R., 2001. Polychlorinated biphenyls in non-accumulating, century-old sediments: Sources, signatures, and mechanism of introduction. *Environmental Science & Technology*, v. 35, p. 2903-2908.
- Corsolini, S., Covaci, A., Ademollo, N., Focardi, S., and Schepens, P., 2006. Occurrence of organochlorine pesticides (OCPs) and their enantiomeric signatures and concentrations of polybrominated diphenyl ethers (PBDEs) in Adelie penguin food web, Antarctica. *Environmental Pollution*, v. 140, p. 371-382.
- Dewailly, E., Nantel A., Weber J.P., and Meyer E., 1989. High levels of PCBs in breast milk of Inuit women from arctic Quebec. *Bull. Environ. Contam. Toxicol.*, v. 43, p. 641-646.
- Driscoll, C.T., Driscoll, K.M., Roy, K. M., and Mitchell, M. J., 2003. Chemical response of lakes in the Adirondack Region of New York to declines in acidic deposition. *Environmental Science and Technology*, v. 37, p. 2036-2042.
- Franz, T. P. and Eisenreich, S. J., 2000. Accumulation of polychlorinated biphenyls and polycyclic aromatic hydrocarbons in the snowpack of Minnesota and Lake Superior: *Journal of Great Lakes Research*, v. 26, p. 220-234.
- Hopke, P.K., Liu, W., Han, Y.J., Yi, S.M., Holsen, T., Cybart, S., and Milligan, M.S., 2003. Measured Summertime Concentrations of Particulate Components, Hg and Speciated Polycyclic Aromatic Hydrocarbons at Rural Sites in New York State. *Environmental Pollution*, v. 123, p. 413-425.
- James, M. O., 2001. Polychlorinated Biphenyls: Metabolism and metabolites: In *Recent Advances in environmental and toxicology and health effects* (editors Larry Robertson and Larry Hansen), The University Press of Kentucky, Lexington, KY, p. 35-46.
- Jenkins, J., 2004. *The Adirondack Atlas: A geographic portrait of the Adirondack Park*, Syracuse University Press, Syracuse, New York, 296p.
- Morrison, R. and Murphy, B. (editors), 2006. *Environmental Forensics Contaminant Specific Guide*, Elsevier, Amsterdam, 541p.
- New York State Department of Health, 2005. *Health advisories Chemicals in Sportfish and Game*. <http://www.health.state.ny.us/nysdoh/fish/fish.htm>
- Pagano, J. J., Rosenbaum, P. A., Roberts, R. N., Sumner, G. N., and Williamson, L. V., 1999. Assessment of Maternal Contaminant Burden by Analysis of Snapping Turtle Eggs. *Journal of Great Lakes Research*, v. 25, p. 950-961.
- Rappe, C., 1974. *Ecological Problems of the Circumpolar Area*; editors, Bylund, E, Linderholm, H., Rune, O.; p. 29-32, Luleå, Sweden: Norbottem Museum
- Robertson, L. and Hansen, L. (editors), 2001. *PCBs: Recent Advances in Environmental and Toxicology and Health Effects*. The University Press of Kentucky, Lexington, 461 p.
- Simonin, H. and Meyer, M., 1998. Mercury and other air toxics in the Adirondack Region of New York. *Environmental Science and Policy*, v. 1, p. 199-209.
- Sokol, R., Kwon, O., Bethoney, C., and Rhee, G-Y., 1994. Reductive chlorination of polychlorinated biphenyls in St. Lawrence River sediments and variations in dechlorination characteristics: *Environmental Science and Technology*, v. 28, p. 2054-2064.
- Tan, Y. and Heit, M., 1981. Biogenic and abiogenic polynuclear aromatic hydrocarbons in sediments from two remote Adirondacks lakes. *Geochimica et Cosmochimica Acta*, v. 45, p. 2267-2279.
- Taylor, S. R. and McLennan, S. M., 1985. *The Continental Crust: its Composition and Evolution*. Blackwell Scientific Publications, Oxford, p. 9-56.
- Wania, F. and Mackay, D., 1996. Tracking the distribution of persistent organic pollutants. *Environmental Science and Technology*, v. 30, p. 390-396A.