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Cesium-137 sediment depth profiles and inventories in Adirondack Lake sediments

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Abstract. Depth distributions and inventories of ^{137}Cs (mCi km^2) were determined in sediment from several fresh water lakes in the New York State Adirondack Preserve. Included were Big Moose and Darts Lakes, part of the North Branch of the Moose River system, as well as North, Sagamore, South, and Woods Lakes and the 'seepage pond', Tamarack Lake. Comparisons were made betwen the ^{137}Cs inventories in these lakes and large inpoundments in the Adirondacks (Hinkley, Great Sacandaga, Stillwater and Cranberry Lake Reservoirs) and other large impoundments and lakes located in various regions of the U.S., especially Cayuga Lake, Ithaca, NY.

None of the Adirondack Lakes had ^{137}Cs distributions with depth in sediment that closely resembled the deposition pattern of weapons testing as a function of time. All of the natural lakes and small impoundments, including the seepage pond, were found to have significantly lower inventories of ^{137}Cs than expected; while the large reservoirs were generally enhanced in ^{137}Cs. We suggest that more than one mechanism may be responsible for the low sediment inventories: for the majority of lakes, flushing of ^{137}Cs out of the lakes during periods of thermal stratification and ice thaw; and for the seepage pond, remobilization of ^{137}Cs into the water column due to biological recycling.

Introduction

The radionuclides ^{137}Cs and ^{210}Pb have been used by a number of investigators to determine the chronologies of pollutant deposition into the sediments of a variety of aquatic ecosystems (Robbins and Edgington, 1975; Pennington et al., 1976; Galloway and Likens, 1979; Durham and Joshi, 1980; Heit et al., 1980, Norton et al., 1980; Jaakkola et al., 1983). However, in recent years, evidence has been presented which suggests that the sediments from soft-water (low alkalinity, low conductivity) lakes, such as those that occur in the Adirondack Preserve, cannot be dated with confidence solely using radioactive isotopes (Binford, 1984; Heit et al., 1984; Parkhurst, 1984). Several reasons for this have been suggested, including: possible nuclide mobility and diffusion (Cornett and Chant, 1983; Davis et al., 1984; White and Driscoll, 1986); sediment redistribution (Von Damm et al., 1979); ion exchange (Evans et al., 1983); delayed watershed input (Davis et al., 1984); and subjectivity in rejecting anomalous data points and applying appropriate models (Parkhurst, 1984; Evans and Rigler, 1983).

Another factor that is not often considered when determining whether lake sediments are appropriate for reconstructing pollutant chronologies is the ^{137}Cs inventory, which is defined as the total or integrated ^{137}Cs activity per unit area (mCi/km^2) in a sediment core (Heit et al., 1984). Determining the degree of enhancement or depletion of ^{137}Cs is of possible significance for estimating atmospheric fluxes into an ecosystem through the use of sediment coring. This is because the sediment distribution of ^{137}Cs in several lakes has been shown to be quite similar to that of other radionuclides (Edgington and Robbins, 1977; Jaakkola et al., 1983; Heit et al., 1984; Davis et al., 1984; Simpson et al., 1986), trace elements (Plato and Jacobson, 1976; Hesslein et al., 1980; Whalen and Thompson, 1980; Heit et al., 1981; Wong and Moy, 1984), and organic micro pollutants (Heit et al., 1981; Plato and Jacobson, 1976). Thus, factors affecting the inventory and distribution of ^{137}Cs may affect these substances as well.

It is our purpose here to evaluate the ^{137}Cs distributions and inventories in the sediments of several soft-water Adirondack lakes. Such lakes are currently of great scientific interest since considerable importance is being placed on the use of their sediments to reconstruct and evaluate the chronologies, histories and causes of acidification in several U.S. ecosystems (NAPAP, 1984; Charles et al.; 1987).

Materials and methods

Sample locations

Table 1 lists the general physical characteristics of the lakes studied. The water chemistry, geology and vegetation of these lakes are described in detail elsewhere (Driscoll et al., 1987; Goldstein, 1984; Bloomfield et al.,

Table 1. Limnological characteristics

Lake	pH[b]	Cond.[b] (μhos)	Surface area (km^2)	Drainage area (km^2)
Big Moose	5.19	26.6	5.2	95.6
Cranberry	6.46	33.5	28	345
Darts	5.23	27.5	1.4	107
Great Sacandaga	6.73	47.3	108	2706
Hinkley	6.95	55.6	11.3	
Mirror[a]	5.5–6.5		0.15	1.0
North	5.17	27		
Sagamore	6.07	31.5	0.7	49.7
South	4.98	25	202	
Stillwater	5.50	28	25	
Tamarack	5.2		0.2	
Woods	4.7–5.1		0.2	2.1

[a] All lakes located in the Adirondack Preserve except Mirror Lake, NH.

[b] All pH and conductivity data except for Tamarack, Woods and Mirror from surface samples taken September 1983. Analytical methodologies are described in EML (1983).

1980; Fuhs et al., 1981; Cronan et al., 1987; April and Newton, 1983; Heit et al., 1980). In general, the Adirondack Lakes are either acidic or sensitive to acidic episodes since most have very low acid neutralizing capacities. The watersheds are forested, sparsely populated and geologically Precambrian, consisting of granite gneiss interlayered with meta-sedimentary rock. Elevations of the lakes are between 500 to 700 m. The soils are generally podzols, with thin glacial till although there is considerable heterogeneity both within and between lake basins. The major vegetation is secondary growth hardwoods with conifers along the lakeshores.

Sediment sampling methodology

All of the sediment cores were taken with a 21 cm diameter sphincter corer with tripod modification, having the capacity to take 90 cm long cores (Burke, 1968; Burke et al., 1980; 1983). The locations for each of the

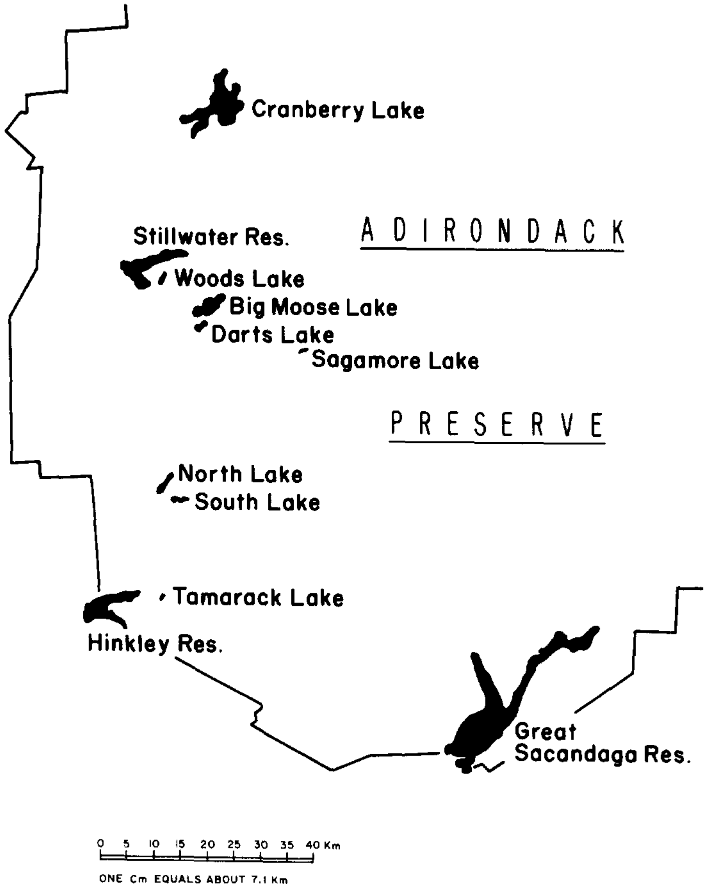

Figure 1. The locations of the Adirondack Lakes.

Adirondack Lakes are shown in Figure 1. The sediments were extruded immediately after retrieval. One centimeter increments were taken to 30 cm and 2 cm sections were taken thereafter. The outer 1 cm of each layer of sediment was descarded resulting in 19 cm in diameter sections. We have found that unless the outer sections are eliminated, contamination results from the surface sediment being 'dragged down' by the core barrel wall. A complete description of this method of extrusion is given in Heit et al. (1986).

Bathymetric surveys were made prior to coring. Replicate cores were taken from the deepest, hypolimnetic basins within each lake, away from areas of potential sediment slumping. Inlet and outlet areas were avoided. Where possible, relatively large, flat basins were selected for coring as a means of decreasing the potential impact of sediment focusing (Davis and Ford, 1982).

^{137}Cs analysis

The methodology we use for ^{137}Cs analyses had been described by Gogolak and Miller (1977). In this method, 100 g (wet) aliquots of sediment are gamma counted for 100 min in sealed 90 cm^3 Teflon-lined aluminium cans using a high resolution germanium gamma-ray detector, a 4000 channel analyzer, and computer. Calibration of the gamma spectroscopy system is described in EML (1983). The overall systematic error of the radionuclide measurements was estimated to be $< 5\%$ (EML, 1983). Based upon the analysis of replicate samples, the coefficient of variation was determined to be 1% to 8%. All of the analyses were performed at the Environmental Measurements Laboratory (EML).

^{137}Cs inventories

The inventories for ^{137}Cs were determined by the formula

$$I = k_i \sum_i \varrho_i t_i C_i$$

where I = the ^{137}Cs inventory for each sediment core increment in mCi/km^2,
C_i = ^{137}Cs (pCi g^{-1}) measured for each increment,
ϱ_i = the density for each sediment increment (g cm^3)
t_i = the thickness of each increment (1 or 2 cm), and
k = 10, a constant for converting pCi cm^{-2} to mCi/km^2.

Results and Discussion

History of ^{137}Cs in the environment and sediment dating of lakes

The history of ^{137}Cs inputs into the atmosphere from weapons testing is well documented (EML, 1977; Heit et al., 1984) and is shown in Figure 2

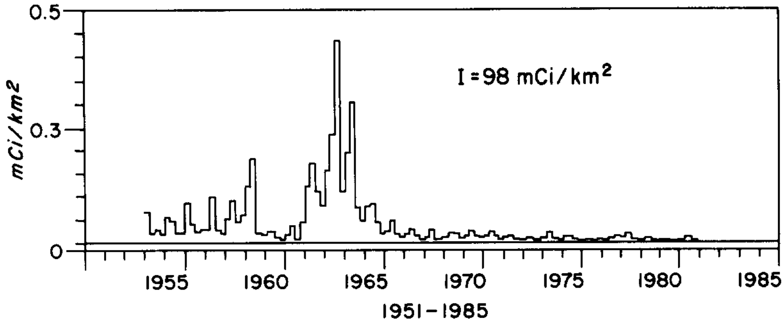

Figure 2. ^{137}Cs fallout (mCi/km^2) in 40 °N to 50 °N Latitude Band, 1951 to 1985. Values are derived from measurements of ^{90}Sr fallout and a conversion factor (Larsen, 1986).

for the 40° and 50° latitude band for the period of 1951 through to the present (Larsen, 1985). The presence of such a ^{137}Cs fallout pattern in a core from a highly resolved hypothetical mainland U.S. lake would allow accurate reconstruction of the sediment chronology for this time period. [It should be noted that mainland U.S. lakes should not be impacted by 'close-in' fallout originating from the Nevada Test Site NTS (P.W. Krey, EML, Personal communication, 1985).] Such a core would be characterized as follows. The core has two easily distinguishable peaks representing periods of significant weapons testing — a major peak, representing 1963 and a secondary peak occurring somewhat deeper in the sediment, representing 1958 to 1959. The ^{137}Cs activity in sediments deposited after 1963 declines rapidly, as did the atmospheric fallout since the signing of the Nuclear Test Ban Treaty. There would be no downward tailing of ^{137}Cs below the depth corresponding to 1951 (beginning of major weapons testing).

We have successfully used the methodologies described in this report to obtain highly resolved sediment cores from several lakes outside of the Adirondacks. As an example, Figure 3 plots the ^{137}Cs activity per unit area (mCi/km^2) vs. depth (cm) for a sediment core taken from Cayuga Lake, Ithaca, NY during August 1981. Clearly, the ^{137}Cs distribution closely follows the historic deposition pattern of ^{137}Cs from atmospheric weapons testing shown in Figure 2.

Confirmation of the accuracy of the ^{137}Cs dating of this core is shown by the use of the SNAP-9A (Systems for Nuclear Auxillary Power) plutonium isotope signature. In 1964, the U.S. satellite (SNAP-9A), which used ^{238}Pu as a heat source for generating power, disintegrated entering the atmosphere of the Southern Hemisphere. As a result, there was a sharp rise in the stratospheric concentration of ^{238}Pu relative to $^{239+240}Pu$ in the Southern Hemisphere. The pulse of ^{238}Pu from SNAP-9A did not reach the surface of the Northern Hemisphere until 1966 at which time a distinct rise in the ratio of ^{238}Pu to $^{239+240}Pu$ activity occurred. It is the increase in this ratio that is used as a geochronological marker for establishing the year

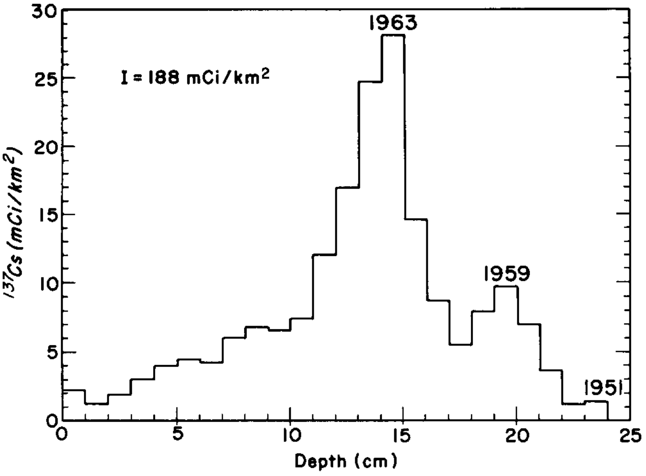

Figure 3. ^{137}Cs activity (mCi/km^{-2}) vs. sediment depth (cm) for Gayuga Lake.

1986 in a sediment core. This is clearly shown in Figure 4 where the ^{238}Pu/$^{239+240}$Pu activity ratio increases between 13 and 11.5 cm deep in the sediment core from Cayuga Lake, NY. The year 1966 is, therefore, assigned to 12.5 cm (the mid-point of the first recognizable increase in the ^{238}Pu/$^{239+240}$Pu activity ratio) which agrees well with the years assigned to this core by the use of ^{137}Cs (assuming that significant changes in sedi-

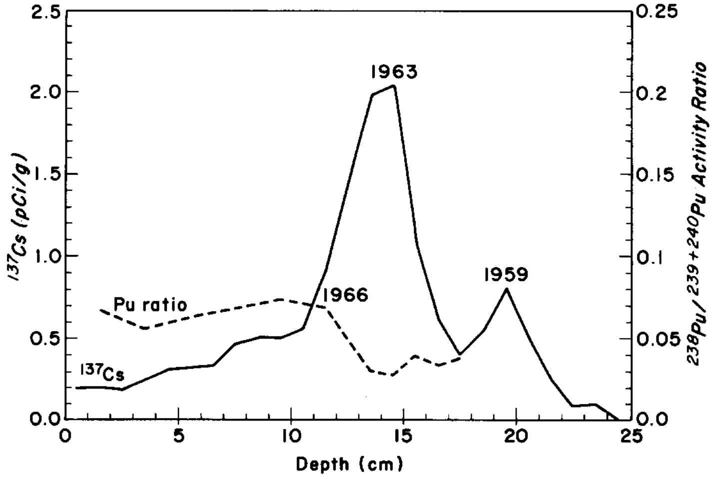

Figure 4. The occurrence of ^{238}Pu from the SNAP-9A satellite relative to ^{137}Cs in the sediments of Cayuga Lake.

mentation rate have not occurred within the time period 1959 to 1966). These two radionuclides have independent sources in the environment and the excellent agreement in chronologies between these two methods confirms that the dating of this core is valid.

The distribution of ^{137}cs in the sediments of Adirondack Lakes and reservoirs

Figure 5 shows the ^{137}Cs activities (mCi/km^2) vs. depth for Big Moose Lake and Darts Lake sediment cores taken in September 1983. The ^{137}Cs profiles from these North Branch Moose River lakes are typical of the other lakes described here and will serve as examples for this discussion. Since the watersheds of the North Branch of the Moose River have received only minor disturbances in the last 30 years (Charles, 1984), we will assume that dramatic changes in sedimentation have not occured and the sedimentation rate is relatively linear within this time period. As shown in the profile for Darts Lake, the highest ^{137}Cs activity occurred at about 9.5 cm. If we assign the year of maximum fallout, 1963, to this point the linear sedimentation rate is 0.48 cm yr^{-1}. If the sedimentation rate were constant within this lake and/or no mixing or diffusion were occurring, we would expect ^{137}Cs to reach zero at about 15 cm., e.g., 1951 (beginning of major weapons testing). This is clearly not the case since ^{137}Cs is detected down to 39 cm. If the 1963 sedimentation rate of 0.48 cm yr^{-1} is applied to this depth, the year would be 1901, which is prior to the first testing of nuclear weapons in 1945 and as such it is impossible. This anomaly could be explained by a variable sedimentation rate. Although this can occur in lakes, the ^{137}Cs profile for Darts Lake shows evidence against this. For example, the documented historical fallout record (Larsen, 1985) clearly shows that there has been a dramatic decrease in ^{137}Cs deposition since the major testing period of the early 1960's. This is not reflected in Darts Lake sediment where the ^{137}Cs levels from 1963 up until the present are quite

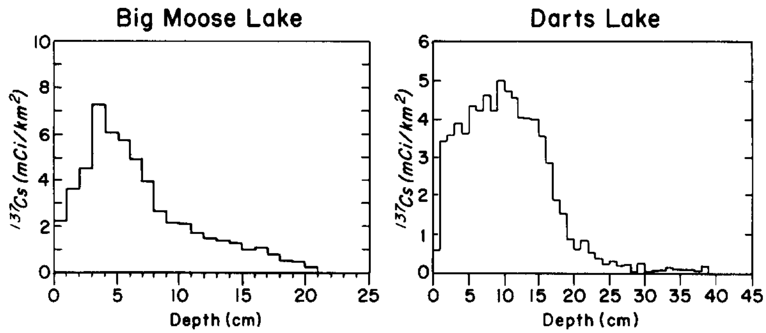

Figure 5. ^{137}Cs activity (mCi km^{-2}) vs. sediment depth (cm) for Big Moose and Darts Lakes.

high relative to the 1963 maximum. Such a distribution in the sediment suggests that mechanisms such as mixing (Robbins and Edgington, 1975; Edgington and Robbins, 1977) diffusion (Pennington et al., 1973, 1976; Jaakola et al., 1983) or resuspension (Davis and Ford, 1982) have occurred and that the core cannot be time resolved to better than 20 yr [sediment from 1963 must be mixed with sediment from the present] (Miller and Heit, 1986).

In Big Moose Lake, ^{137}Cs peaks at about 3.5 cm. If we assign 1963 to this peak, the linear sedimentation rate is 0.18 cm yr^{-1}. If the sedimentation rate were constant within this lake and mixing or diffusion were not occurring, we would then expect ^{137}Cs to reach zero at 5.6 cm, i.e., 1951. Again, this is clearly not the case, ^{137}Cs is detected to 20 to 21 cm. Using the same rational as for the Darts Lake, this is also impossible, since the age of this sedimentation would be 1863. As in the case of Darts Lake, we estimate the time resolution to be on the order of 20 years (Miller and Heit, 1986).

^{137}Cs sediment distribution profiles are shown for the other Adirondack lakes and large impoundments in Figures 6 and 7. Also displayed is the ^{137}Cs profile for Mirror Lake, an oligotrophic lake located in the Hubbard Brook/White Mountain region of NH. Again, the ^{137}Cs profiles only

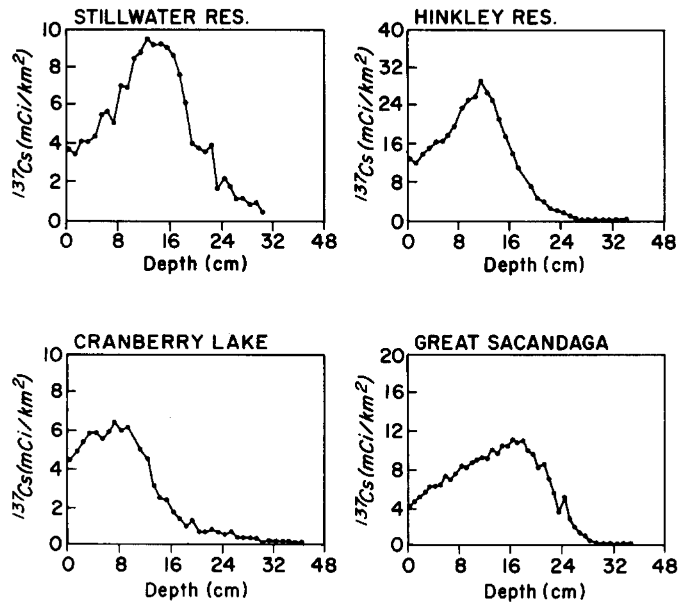

Figure 6. ^{137}Cs activity (mCi km^{-2}) vs. sediment depth (cm) for large Adirondack impoundments.

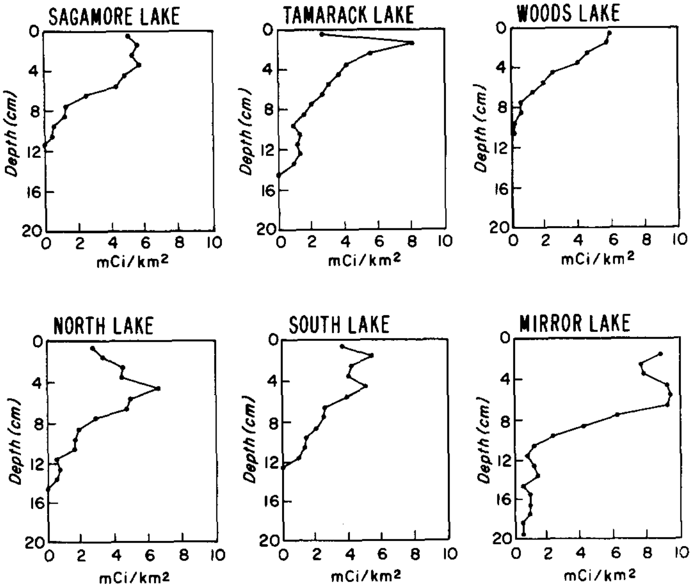

Figure 7. ^{137}Cs activity (mCi km^{-2}) vs. sediment depth (cm) for Mirror, North, Sagamore, South, Tamarack and Woods Lakes.

remotely resemble actual weapons fallout history indicating that the time resolutions are no better than those of Darts and Big Moose Lakes. Significant differences between these cores and actual fallout chronology include the presence of single ^{137}Cs peaks. The ^{137}Cs activities remain quite high from the sediment-water interface to the depth of maximum activity (1963), and in several cases, the ^{137}Cs extends down well into the sediment as a long ‘rat-tail’.

Table 2. ^{137}Cs soil inventories in central New York State (mCi/km^2)[a]

Location		Inventory
Albany		90 ± 2
Troy		92 ± 3
Aurora		98 ± 4
Ludlowville		98 ± 3
Taughannock		110 ± 4
	Average	98 ± 8

[a] Decay corrected to September 12, 1983 sampling date. All data from C.S. Klusek (EML, Personal communication, 1985). Samples collected by methodology described in EML (1983).

Determination of ^{137}Cs inventories

It has been shown that if carefully selected, soils that are protected from erosion (overlain with vegetation) and flooding (Loughran et al., 1982) and that are undisturbed since 1951 will provide accurate and precise inventories per unit area of radionuclides, including ^{137}Cs, deposited at those sites from atmospheric weapons testing. In this regard, ^{137}Cs inventories in soil taken from undisturbed sites in New York State (see Table 2) have an average of 98 ± 8 mCi km^2 (C. S. Klusek, EML, New York, NY, Personal communication, 1985). This value is in excellent agreement with the cumulative surface deposition of ^{137}Cs estimated to be 98 ± 11 mCi km^2, decay corrected to 1983, from fallout within the 10° latitude band, which incorporates central New York State (Larsen, 1986; Simpson et al., 1986).

Besides latitude, the amount of precipitation may influence the inventory of ^{137}Cs. The average precipitation for central New York State is 75–100 cm/yr compared to 100–130 cm/yr for the Adirondack region (Pack, 1972). Hence, one would expect the cumulative surface deposition of ^{137}Cs for the Adirondacks to be greater than the 98 ± 8 mCi/km^2 determined for central New York. However, to be conservative, we will assume that the cumulative surface deposition for the Adirondack region is also 98 ± 8 mCi/km^2.

If lake basins acted in a manner similar to undisturbed soils, then sediment cores would also be expected to serve as accurate integrators of atmospheric debris. Unfortunately, the situation with sediments is not the same as for soils. Sediments are susceptible to disturbances not usually associated with soils covered by vegetation, such as: slumping, resuspension and redistribution both within and between lake basins from water currents, storms and seasonal overturns (Davis and Ford, 1982; Edgington and Robbins, 1977), as well as from natural and anthropogenic disturbances to the watershed from logging, fire, heavy storms and agricultural practices.

Since all of the lakes discussed in this report were either sampled in 1978 (Segamore and Woods), 1979 (North, South, Sagamore, Mirror and Tamarack), or 1983 (Big Moose, Cranberry, Darts, Great Sacandaga, Hinkley, and Stillwater), we would expect sediment cores from these water bodies to have the decay corrected, conservative ^{137}Cs inventory of 98 ± 8 mCi/km^2 if they were all perfect integrators.

^{137}Cs inventories in large impoundments and factors affecting retention

Among water bodies most likely to have enhanced ^{137}Cs sediment inventories are major impoundments (reservoirs) with large drainage areas. Such systems would be expected to have ^{137}Cs inventories higher than expected from direct atmospheric deposition because of both soil erosion within the watershed due to intermittent flooding (Ritchie et al., 1971; Mitchell et al., 1983; McHenry et al., 1978; Campbell et al., 1982) and resuspension of previously deposited sediments in the basin due to seasonal "draw-down" followed by wind and wave action upon the exposed sediments (Krey et al., 1980). Sediment focusing may also be an important mechanism in larger lakes. Edgington and Robbins (1977) have shown that some areas of the great lakes are depleted in ^{137}Cs while other areas are greatly enriched due to the funneling of sediments between basins by currents and or storms.

In order to avoid such bias from sediment focusing, here, bathymetric surveys were made prior to coring and replicate cores were taken from the largest and flatest basins, away from areas of potential sediment slumping. Inlet and outlet areas were avoided. For example, in Cayuga lake, 10 cores were taken in this manner over a distance of 24 km and the ^{137}Cs inventory

Table 3. ^{137}Cs inventories in selected U.S. reservoirs, lakes and soils[a] (mCi km^2)

Lake/Reservoir[b]	Location	Sediment inventory	Soil inventory	Ratio (%)[l]
Hinkley[c]	Grant, NY	335	98 ± 8	342
Great Sacandaga[c]	Northville, NY	251	98 ± 8	256
Stillwater[c]	Town of Webb, NY	151	98 ± 8	154
Cranberry[c]	Clifton, NY	91	98 ± 8	93
Other U.S. lakes and reservoirs[a]				
Upper Enterprise[d]	Enterprise, UT	750	101	750
Nottely[3]	Murphy, NC	687	[120]	570
Lake Mead[f]	Overton Beach, NV	> 583		
Deer Creek[g]	Heber City, UT	469	128	370
Echo[g]	Coalville, UT	361	(79)	(500)
Hansen[h]	Los Angeles, CA	> 432		
Standley[i]	Jefferson County, CO	319	85–110	330
Grand[g]	Grand Lake, CO	260	82–98	300
Chatuge[e]	Murphy, NC	235	[120]	200
Santeetlah[e]	Robbinsville, NC	209	[120]	170
Cayuga[i]	Ithaca, NY	183	97	190
Blue Ridge[e]	Blue Ridge, GA,	176	[120]	150
Gull Pond[i]	Cape Cod, MA	142	97	146
Utah Lake[k]	Provo, UT	110	118	93

[a] All sites sampled were analyzed by EML.
[b] Major impoundments in the Adirondacks (basin > 10 km^{2}0.
[c] This report. [d] Krey (1980). [e] Miller and Heit (1986).
[f] Lake Mead ^{137}Cs inventory for 0–56 cm only (^{137}Cs was detected at bottom of core).
[g] Hardy (EML, Personal communication, 1983).
[h] Hansen reservoir ^{137}Cs inventory for 0 to 55 cm only (^{137}Cs was detected at bottom of core).
[i] Hardy (1980). [j] Heit et al. (1986). [k] Krey and Beck (1981).
[l] Ratio = [(^{137}Cs inventory in sediment/^{137}Cs inventory in undisturbed soil)] × 100%.
Note:
[] Estimated ^{137}Cs soil inventories based upon ^{90}Sr deposition (Larsen, 1985)
() Suspect soil inventory for Coalville, UT due to flooding.

remained almost constant at 177 ± 17 mCi/km^2, coefficient of variation = 10%. The uniformity of the ^{137}Cs inventory over this relatively large distance suggests that our precautions in selecting coring locations does reduce the potential bias from sampling in areas of enhanced sedimentation due to focusing.

Table 3 is a listing of the ^{137}Cs inventories in sediment cores from representative large impoundments and lakes taken by EML over the last several years using the same or a similar coring methodology. Also shown are the inventories of the large Adirondack impoundments (basins > 10 km^2) and undisturbed soils taken near the water bodies. As shown here, sediment inventories ranged from 750 mCi/km^2 in the largely desert environment of Upper Enterprise Reservoir, to 91 mCi/km^2 in the forested Cranberry Lake. It is clear from this table that the majority of large Adirondack impoundments (Hinkley, Great Sacandaga, Stillwater) are typical of those found in other areas of the U.S., since their sediment inventories are generally greater than the ^{137}Cs inventories from the direct deposition of nuclear weapons fallout as reflected in undisturbed soils.

An extreme example of enhancement is Upper Enterprise Reservoir, which had a ^{137}Cs sediment inventory of 750 mCi/km^2 and an undisturbed soil inventory of 101 mCi/km^2. This gives a ratio [(sediment ^{137}Cs inventory/fallout ^{137}Cs inventory) × 100%], of 750%, indicating significant enrichment from erosion. Krey et al. (1980) have shown that the ^{137}Cs enrichment in this reservoir is a direct consequence of resuspension since several anomalous ^{137}Cs peaks correspond with known periods of reservoir draw-down. Hinkley, Great Sacandaga and Stillwater reservoirs had ratios of 350%, 260% and 160%, respectively. Similar results have been reported for sediments from other Northeastern lakes outside of the Adirondacks, where the ^{137}Cs inventories were all enhanced compared to the cumulative fallout inventory of 98 ± 8 mCi/km^2. Inventories for these lakes ranged from 132 mCi/km^2 in Sylvan Lake, NY to 281 mCi/km^2 in Lake Champlain, NY (Whalen and Thompson, 1980; Heit et al., 1986).

The ^{137}Cs inventory in Cranberry Lake sediment was similar to Utah Lake, Provo, UT in that both were approximately equal to that from direct fallout. The lack of ^{137}Cs enhancement in these systems may be explained by the fact that the coring locations were several km away from areas of enhanced sediment accumulation such as dams (Cranberry) or deltas (Utah). As such, the influence of terrestrial erosion would be expected to be considerably reduced, resulting in sediment inventories similar to that from the atmosphere. An alternative interpretation is that other mechanisms, such as sediment redistribution or focusing, are in operation in these systems.

^{137}Cs inventories in soft-water Adirondack Lakes and small impoundments

As previously stated, soft water Adirondack Lakes having 100% ^{137}Cs retention within the lake basin(s), would be expected to have a minimum cumulative ^{137}Cs inventory of 98 ± 8 mCi/km^2. As shown in Table 4, this is clearly not the case because all of the lakes in this category are deficient in ^{137}Cs. Again, using the lakes in the North Branch of the Moose River as examples, the ^{137}Cs inventory in the sediment of Big Moose Lake was 56 mCi/km^2, or 57% of the expected, assuming the unlikely possibility that no erosion or sediment focusing were operating upon the system to enhance the inventory. The results were similar, but somewhat higher, for Darts Lake where 75 mCi/km^2 was present in the sediment, which is 77% of the expected. The lowest inventories were found in the sediments of Woods lake (27 mCi/km^2), with the remaining Adirondack Lakes having inventories of between 36 and 41 mCi/km^2. Tamarack lake, with an inventory of 38 mCi/km^2 is a seepage lake with no inlet or outlet (J. N. Galloway, University of Virginia, Charlottesville, VA, Personal communication, 1979). Similar inventories of 36 and 38 mCi/km^2 were found for two separate basins of Sagamore Lake, sampled in succeeding years by related

Table 4. ^{137}Cs inventories in soft-water lakes and small impoundments

System	Location	Inventory	Ratio[f] (%)
Big Moose Lake[a]	Adirondacks	56	57
Darts Lake[a]	Adirondacks	75	77
North Lake[a]	Adirondacks	41	42
South Lake[a]	Adirondacks	40	4
Tamarack Lake[a]	Adirondacks	38	39
Sagamore Lake[a,b]	Adirondacks	36	37
Sagamore Lake[a,c]	Adirondacks	38	39
Woods Lake[a]	Adirondacks	27	28
Little Long Pond[d]	ME	86	88
E. Richardson Pond[d]	ME	80	82
Unnamed Pond[d]	ME	70	71
Coburn MTN Pond[d]	ME	42	43
Tumbledown Pond[d]	ME	39	40
Rangley MTN Pond[d]	ME	38	39
Speck Pond[d]	ME	28	29
Second Pond[d]	ME	23	23
Klondike Pond[d]	ME	22	22
Mirror Lake[e]	NH	40	42
Solitude Pond[d]	NH	38	40
Carter Pond[d]	NH	44	46
Dream Lake[d]	NH	30	32
Branch Pond[d]	VT	43	45

[a] This work. [b] Core taken in 1978. [c] Core taken in 1979. [d] Davis et al. (1984). [e] Heit (1986). [f] Ratio = [(^{137}Cs inventory sediment/^{137}Cs inventory from fallout) × 100%] cumulative ^{137}Cs inventory from fallout = 98 ± 8 (This report).

but different coring methods. This reproducibility indicates that these inventories are not artifacts of the methodology. It should be noted that the method for taking the 1979 Sagamore cores is the same used to obtain the high resolution cores from Cayuga lake. Similar results were found for North and South Lakes, two small impoundments that are located a few km from each other (e.g., ^{137}Cs = 41 and 40 mCi/km^2, respectively).

These results are in marked contrast to the values previously shown for the large impoundments and the other representative lakes that were shown on Table 3. The depleted ^{137}Cs inventories in the Adirondack soft-water lakes suggests that material is not reaching the sediment or is being remobilized and redistributed elsewhere. This phenomenon is not confined to soft water Adirondack Lakes. Using the data of Davis et al. (1984) we have examined, in a similar way, data for soft-water lakes from Maine, New Hampshire and Vermont, as well as Mirror Lake, in the Hubbard Brook region of New Hampshire (Heit, Unpublished data, 1979). Table 4 shows that the inventories of these lakes were also depleted, ranging from a low of 22 mCi/km^2 (Klondike pond) to a high of 86 mCi/km^2 (Little Long Pond). The majority of lakes had ^{137}Cs inventories between 20 and 40 mCi/km^2, which is quite similar to that found in the Adirondacks. The depleted (40 mCi/km^2) ^{137}Cs inventory in sediment cores from Mirror Lake, NH is interesting in that the cores were taken in the deepest basin of the lake, which has been shown to be the site of significant focusing of organic-rich, low density sediments (Davis and Ford, 1982). Thus, one would have expected the cores from Mirror Lake to be enhanced in ^{137}Cs rather than depleted (Longmore et al., 1983).

Factors affecting ^{137}Cs binding in Adirondack Lake sediments

Some factors that have been suggested which could influence ^{137}Cs adsorption to sediments are: the presence of clay, organic content, dissolved oxygen in the hypolimnion, and the concentration of hydrogen ions (pH). Each of these factors is discussed relative to the Adirondack Lakes.

Clay content

The ability of a substrate, soils or sediments, to retain ^{137}Cs appears to be directly related to the quantity and quality of the clay mineraology. For example, substrates that contain relatively high levels of layer-silicates such as micas, vermiculites and interleaves micas (formerly called illites) have been shown to preferentially adsorb or 'fix' trace quantities of ^{137}Cs (Francis and Brinkley, 1976). ^{137}Cs bound by such minerals, is only extractable by ions of similar charge and size, i.e., NH_4^+, K^+, and Rb^+ (Evans et al., 1983). In soils and sediments containing a higher proportion of minerals, such as kaolinite, the binding is significantly weaker and the ^{137}Cs can be displaced by exchange with several other cations, e.g., Na^+, Ca^{++}, Mg^{++}, and probably Mn^{++} and Fe^{++} (Evans et al., 1983). The differences between binding capacities of clay minerals appears to be related to the dimension of the interlattice layers (Tamura and Jacobs, 1960).

April and Newton (1983) determined the clay mineralogy of the soils in the vicinity of three soft-water Adirondack Lakes, including Woods and Sagamore. Their results show that vermiculite was the dominant clay mineral in the podzols of the watersheds of all the lakes, with lesser amounts of illite, kaolinite and mixed layer-clays (e.g., illite/vermiculite.) Since Tamura and Jacobs (1960) have shown that vermiculite is only slightly less binding with ^{137}Cs than illite and considerably more binding than kaolinite, these results suggest that the Adirondack soils should absorb ^{137}Cs. Thus, watershed disturbances such as logging, which would be expected to result in enhanced soil erosion, should also lead to enhanced ^{137}Cs inventories rather than the depleted sediment inventories shown in Table 4. However, because the watershed disturbances for the lakes in this study were either relatively minor (Charles, 1984) or occurred prior to, or considerably after the major periods of atmospheric weapons testing, enhanced ^{137}Cs inventories from erosion appear to be of little importance. For example, while the Sagamore lake watershed was logged in the period 1975 to 1979 (S. A. Norton, University of Maine, Orono, ME, Personal communication, 1984), the ^{137}Cs inventories were lower than for most of the lakes listed on Table 4, 36–38 mCi/km^2.

Unfortunately, little information is currently available on the sediment clay mineralogy of these soft-water lakes. However, the information that is available suggests minimal levels of clay minerals present. For example, Galloway et al. (1983) state that while ^{137}Cs binding minerals (illite, chlorite and mixed-layered clays, i.e., illite/vermiculite) were in the sediments of Sagamore and Woods Lakes, they were present at barely detectable levels. Similar results ($< 0.1\%$ clay) were reported for Woodhull Lake (Galloway and Likens, 1979). Davis et al. 91984) found that clay minerals were not detected in most of the 19 lakes soft-water lakes examined in New England. (It should be noted that severe interferences due to the presence of high concentrations of diatoms may lead to erroneous results when quantifying clay-minerals in such sediments (R.H. April, Colgate University, Hamilton, NY, Personal communication, 1985). These results suggest that the lack of clay-minerals in the sediments may allow ^{137}Cs to mobilize in the pore water. In this regard, we are now studying the sediments from ~ 100 lakes to clearly define the association of clay mineralogy and other geochemical factors with radionuclide binding.

Organic carbon content

Besides the presence of clay minerals, various investigators have suggested that the presence of organic ligands may also affect the binding capacities of soil and sediment for ^{137}Cs, although this work often appears to be poorly understood and the results somewhat contradictory.

For example, Lehr and Kirchmann (1973) showed that organic matter in soil can temporarily retain a portion of fresh fallout deposition. Hinkley (1978) reported that although ^{137}Cs in soils is associated with organic matter, it appears that soil moisture is required for ^{137}Cs binding, while Russell (1966) states that in soils of high organic content with little clay content the ^{137}Cs is relatively mobile. Ritchie et al. (1970) reported that 80% to 96% of the ^{137}Cs in Smoky Mountain forest soils was associated with organic matter and the upper 3.8 cm of mineral soil, which presumably contains clay minerals. According to Longmore et al. (1983), sediments composed of fine, highly organic colloidal material, such as that present in the Adirondack Lakes, should be ideal for the adsorption of ^{137}Cs because of the small particle size of such material, even though little clay is present. Evans et al. (1983), however, found that ^{137}Cs is only mimimally associated with organic ligands in sediments, but is associated with micaceous clay minerals. It would appear from these divergent findings that while ^{137}Cs will associate with organics, without the presence of binding clays, it will be mobile.

Although little data are currently available, the percent of organic matter in the sediments from lakes deficient in ^{137}Cs is higher than that found in lakes and reservoirs with ^{137}Cs profiles closely reflecting historic fallout events and inventories. For example, Galloway et al. (1983) reported that the organic content of Sagamore and Woods lakes varied little with depth and ranged from 28% to 38%. Similar results have been reported for other lakes deficient in ^{137}Cs, including Mirror Lake, NH (Von Damm et al., 1979), Woodhull Lake in the Adirondacks (Galloway and Likens, 1979), and for several remote forested lakes in New England (Davis et al., 1984). Somewhat higher results have been reported by S.A. Norton (University of Maine, Orono, Personal communication, 1984) who found the sediment organic content of 10 Adirondack Lakes to range between 40 and 60%. By comparison, the organic content of the highly resolved Cayuga Lake sediments ranged from 0.4% to 3% (Heit et al., 1986). Thus, as stated by Davis et al. (1984), it is likely that highly organic sediments with considerable biological decomposition occurring (Rudd et al., 1986) and little evidence of binding clay (such as the Adirondacks Lakes) would be expected to have mobile ^{137}Cs.

pH effects

Based upon results for a perched seepage pond in Australia, depleted in ^{137}Cs with a pH of $\sim$4.0, Longmore et al. (1983) suggested that high concentrations of hydrogen ions (i.e., low pH) may contribute to the sedimentary release of ^{137}Cs, allowing diffusion in pore waters. A similar process has been proposed by Evans et al. (1983), where H^+, if viewed as the hydronium ion with a radius of 1.38 Angstroms, would be able to undergo a catio exchange with ^{137}Cs if binding clay were present. However, data presented by Sibley et al. (1981) showed that pH values from 4 to 9 had no discernable effect on the binding of ^{137}Cs to sediments taken from several fresh water U.S. locations. As shown in Table 1, all of the lakes reported here with depleted ^{137}Cs inventories had pH's > 5, suggesting that factors other than H^+ concentration are more important in causing ^{137}Cs mobility.

Dissolved oxyugen and redox potential

There is recent evidence from several investigations of lakes with anoxic hypolimnions that the Fe/Mn redox cycle may be responsible for increases in hypolimnetic concentrations of ^{137}Cs and Pu (Alberts et al., 1979; Sholkovitz et al., 1982; Sholkovitz, 1984). Evans et al. (1983) have shown that under anoxic conditions, NH_4^+ production in sediments may, via cation exchange, mobilize ^{137}Cs from binding clay minerals. Of the lakes reported here, Tamarack and Mirror Lakes have seasonal anoxic hypolimnions. However, it remains unknown whether such mechanisms contribute to the depleted ^{137}Cs inventories, since the sediments of these lakes appear to be composed primarily of organic debris with little clay present for cation exchange. The remainder of the lakes retain aerobic hypolimnions through stratification and, thus are thought not be affected in this manner. White and Driscoll (1987) have shown that Mn transport in aerobic Darts Lake is of minor significance compared to Pb and is unaffected by temporal changes in water chemistry.

Proposed mechanisms of ^{137}Cs migration and loss from Adirondack sediments

Our results indicate that there are at least two categories of multiprocess mechanisms causing the lower than expected ^{137}Cs inventories in the sediments — physical flushing of ^{137}Cs out of the lakes and biological recycling or resuspension of ^{137}Cs into the water column.

^{137}Cs flushing

We suggest the following hypothesis for the majority of the Adirondack Lakes. Due to the lack of binding clay-minerals, following thermal stratification in April and November (Driscoll et al., 1987), ^{137}Cs is mobilized from the sediments by diffusion into the hypolimnion, transported by currents through the outlet and redeposited downstream in areas of reduced current. Coupled with this, ^{137}Cs and other atmospherically transported pollutants deposited on the snow and ice (which remains on the lakes for the majority of the year) are released into the surface water during periods of thaw, which occurs during April and May. During these periods, significant fractions are flushed from the lakes prior to being deposited in the sediment by the substantial currents and water flow that occurs below the melting ice and snow pack (Driscoll et al., 1987). According to N.E. Peters (U.S. Geological Survey, Doraville, GA, Personal communication, 1985), groundwater discharge would not be a significant factor in ^{137}Cs loss from these lakes.

An ideal location to test this hypothesis would be at a series of contiguous lakes. If the hypothesis is correct, the first of the contiguous lakes would be depleted in ^{137}Cs compared to the next contiguous down-river lake, which in turn would have a lower inventory than the contiguous down-river lake, etc. Such a system is present on the North Branch of the Moose River, where the discharge from Big Moose Lake flows into Darts Lake, which in turn flows into Rondaxe Lake. As shown in Table 5, the inventory is in fact ~35% less in Big Moose than in Darts Lake (56 mCi km^2 vs. 75 mCi/km^2). Unfortunately, because of logistical problems, we were not able to verify this mechanism by taking sediment cores from Rondaxe Lake, which should, if our hypothesis is correct, have a higher ^{137}Cs inventory than Darts Lake. However, data presented by Durham and Joshi (1980) for three contiguous lakes in Saskatchewan clearly supports this hypothesis. As shown in Table 5, the inventory in the first of these contiguous lakes, Pasqua Lake, was 34 mCi/km^2, followed by Echo Lake with 59 mCi/km^2, while the last of the lakes, Katepwa Lake, had an inventory of 91 mCi/km^2. As described by these investigators, snow melt provides most of the annual discharge for these lakes and peak flows occur during the spring snow melt. This is quite similar to the situation in the Adirondacks. Unfortunately, Durham and Joshi (1980) present no information on the organic and clay mineralogy of the sediments. Interestingly, the authors note that the unsupported ^{210}Pb inventories in the first lake also appear to be depleted relative to that in the next contiguous lake, suggesting that constituents other than ^{137}Cs are flushed between basins as well.

Table 5. ^{137}Cs inventories in the sediments of contiguous lakes (mCi/km^2)

System	Location	Inventory (%)	Ratio[e]
Big Moose lake[a]	Adirondacks	56[c]	57
Darts Lake[a]	Adirondacks	75	77
Pasqua Lake[b]	Saskatchewan	34[d]	37
Echo Lake[b]	Saskatchewan	59	65
Katepwa Lake[b]	Saskatchewan	91	100

[a] This work.

[b] Durham and Joshi (1980).

[c] Cumulative ^{137}Cs inventory from fallout = 98 ± 8 for Adirondacks (This report).

[d] Cumulative ^{137}Cs inventory from fallout = 91 mCi/km^2 for Saskatechewan (UNSCEAR, 1977).

[e] Ratio = [^{137}Cs inventory sediment/^{137}Cs inventory from fallout × 100%].

Biological resuspension

We propose a second mechanism for the loss of ^{137}Cs from the sediments of seepage lakes such as Tamarack Lake. In such a system, pollutants enter primarily by direct atmospheric deposition. Since there are no exits from the pond other than potential ground water seepage, it is postulated that all ^{137}Cs that enters (disregarding radioactive decay) remains in the system. In a similar perched lake in Australia, Longmore et al. (1983) took a series of sediment cores from locations in the epilimnion and hypolimnion ranging in depth from 1.5 to 8.5 m. In all cases, the cores had depleted ^{137}Cs inventories relative to the cumulative inventory from fallout in the same latitude band in the Southern Hemisphere, although the cores taken from the deepest parts of the lake generally had the highest ^{137}Cs inventories.

Longmore et al. (1983) suggested that biological recycling is responsible for the depleted ^{137}Cs inventories in such seepage ponds. Rapid dissolution of the sediments by microbial activity at the sediment-water interface would, according to these investigators, allow the ^{137}Cs to be released into the water and be assimilated by plankton. During periods of plankton bloom, the ^{137}Cs in the water column would rise due to biological incorporation, followed by re-sedimentation at the death of the bloom or by deposition with fecal pellets (Hesslein et al., 1980). This would be followed by another cycle of microbial degradation, etc. According to Longmore et al. (1983), such a mechanism would result in a ^{137}C profile similar to that shown for Tamarack Lake (Figure 7), where cycling between the sediment and water would tend to enrich the upper sediment layers. It is also possible that aerobic production of NH_4^+ (Evans et al., 1983) in conjunction with cation exchange, would contribute to the mobilization of the ^{137}Cs from the sediments. However, this mechanism is unlikely since the sediments contain little if any binding clay minerals (R. April, Colgate University, Hamilton, NY, Personal communication, 1986).

Based upon the similarity between the Australian and Adirondack perched lakes and that rapid dissolution of sediments rich in organic matter has been reported to occur in the Adirondack Lakes (Rudd et al., 1986), we hypothesize a similar mechanism of biological recycling to account for the depletion of ^{137}Cs in Tamarack Lake. In this regard, we expect to collect water and plankton from Tamarack and several other Adirondack Lakes in order to verify this hypothesis.

Summary and Conclusions

(1) The ^{137}Cs distributions in the sediments of soft water (low alkalinity, low conductivity lakes) Adirondack Lakes do not follow the historic patterns of fallout from atmospheric weapons testing. Furthermore, the ^{137}Cs inventories in the sediments of these lakes

(including a seepage pond) are all depleted relative to decay-corrected cumulative inventories expected from fallout from weapons testing. In large impoundments, with relatively fast rates of sedimentation, the ^{137}Cs appears to be conserved in that the sediment inventories were either enhanced or at approximately the same levels as that expected from fallout from weapons testing.

(2) We suggest that more than one mechanism is responsible for the low sediment inventories: for the majority of lakes, flushing out of ^{137}Cs during period of thermal stratification and ice thaw; and for the seepage pond, remobilization of ^{137}Cs into the water column by biological recycling.

(3) Low or non-detectable levels of binding-clay minerals, such as illite, are suggested as the prime causes of ^{137}Cs mobility in the sediments of these lakes.

(4) The lack of agreement in the sediment distribution of ^{137}Cs and historical fallout events and depleted ^{137}Cs sediment inventories, indicates that this radionuclide should not be used to determine the sediment chronologies of soft-water lakes without other substantiating geochronological markers.

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