



# Time-Scales for Sediment Focusing in Lake Michigan Based on Fallout <sup>137</sup>Cs Data: A Thirty-Year Study

## The Problem

Lake Michigan, the fifth largest lake in the world and second among the Laurentian Great Lakes, has a 100 year hydraulic residence time. As a result many varieties of contaminants, introduced since the mid-19th century, have remained in the lake and may be chronically available to biota. Particularly persistent are particle-associated contaminants such as lead, mercury, DDT, PCBs, PAHs plus fallout <sup>137</sup>Cs and plutonium isotopes. While they are efficiently adsorbed by fine particles that rapidly settle out, significant amounts return to the water column through annual cycles of re-suspension, horizontal transport, and re-deposition. As a result, the long-term removal of such contaminants from the lake often reflects the average time particles reside in a pool of suspendable sediments prior to burial in permanent deposits. Previous studies, using fallout radionuclides, have shown that the characteristic particle removal time is around 20 years for Lake Michigan.

With the establishment of the Lake Michigan Mass Balance (LMMB) Program in 1994, the Environmental Protection Agency (USEPA) began developing a refined mass balance model to account for the fate of contaminants in the lake with improved spatial and temporal resolution. The model will more realistically describe effects of currents, thermal stratification, fall overturn, and particle transport, particularly sediment resuspension, on the movement and ultimate removal of contaminants from the lake. A particularly difficult aspect of modeling is the proper model treatment of sediments as a source of particles to the water column. Thus a critical part of the LMMB Program has been to determine sediment physical properties and to generate state-variable data such as net sediment accumulation rates, depths and rates of near-surface sediment mixing and times of residence of particles in sediment mixed layers.

## Approach

One of GLERL's major collaborative roles in the LMMB program has been to provide such information based on sedimentary profiles of naturally occurring <sup>210</sup>Pb and fallout <sup>137</sup>Cs in cores collected from principal depositional basins of the lake (Fig. 1a). Of the 134 cores for this study (Fig. 1b), 79 were collected in 1992 by the Great Lakes WATER Institute (GLWI). An additional 55 were collected from 1994 through 1996 by GLERL, GLWI and the USEPA, Large Lakes Research Station. Coring sites are located within depositional basins where fine sediments preferentially accumulate. Coring operations were conducted from the R/V Lake Guardian (USEPA, Fig. 2a) and the R/V Neeskey (GLWI). Most cores were collected with a modified Sontar type box corer (Fig. 2b). A few were obtained in 1992 using an open tube gravity corer when sea conditions precluded box coring. Sub-cores were obtained by insertion of 10 cm diameter tubes into each acceptable box core sample. Sediments within tubes were hydraulically extruded and sectioned aboard ship into 1-cm sections (Fig. 2c). Within each section, sediments were quantitatively transferred to containers, sealed, returned to the lab, weighed, freeze dried, re-weighed and radiometrically analyzed.

## Fallout of <sup>137</sup>Cs and Concentrations in Water

Delivery of <sup>137</sup>Cs ( $t_{1/2} = 30.2$  years) to the Great Lakes began with the start of atmospheric testing of nuclear weapons in 1952, peaked in 1963-1964 and diminished substantially by the early 1970s following the test ban treaty (Fig. 3). Because <sup>137</sup>Cs strongly binds to particles, it was rapidly removed from the water column within a few years. However small amounts return to the lake annually through resuspension of sediments in the late fall when the water column mixes. Concentrations of decay-corrected <sup>137</sup>Cs in the southern part of the unstratified lake have declined exponentially with about a 20-year time constant during the post-fallout period from 1970 onward.

## Early Inventories of <sup>137</sup>Cs in Sediments of Southern Lake Michigan

The earliest map of the vertically integrated amount (inventory) of <sup>137</sup>Cs in sediments was based on gravity cores collected in 1972 at 50 sites in the southern part of the lake (Fig. 4, left panel). By the end of the fallout period, the inventory contour map resembles the pattern of fine sediment accumulation in the Waukegan Member of the Lake Michigan Formation (Fig. 5). This most recent member has an average <sup>14</sup>C age of 3500 years and represents historic and present day, strongly focused allocation of fine sediments to the bottom. Evidently <sup>137</sup>Cs has moved with particles and may serve as a tracer of their pathways from introduction to burial.

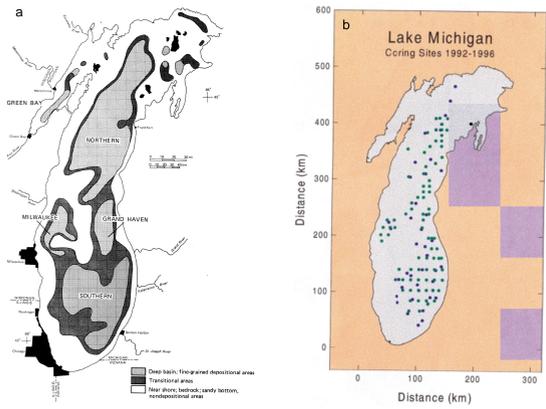


Fig. 1. (a) Generalized depositional environments of Lake Michigan (Cahill, 1981). (b) Box coring sites (1992-1996) for the Lake Michigan Mass Balance Program. The 134 sites are located within the principal depositional basins of the main lake. About 60% of the lake bottom has no net long-term accumulation of fine sediments.



Fig. 2. (a) R. Wood standing by the R/V Lake Guardian. (b) Crew members deploying the box corer. (c) N. Morehead preparing a box sub-core for sectioning.

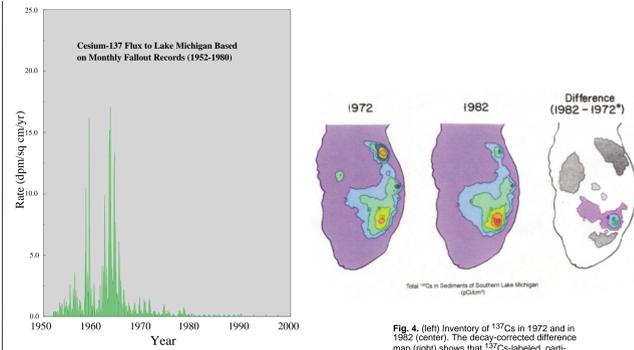


Fig. 3. A 25-year monthly record of <sup>137</sup>Cs fallout over Lake Michigan (Robbins, 1985). Over 90% of the deposition occurred between 1955-1970.

Fig. 4. (left) Inventory of <sup>137</sup>Cs in 1972 and in 1982 (center). The decay-corrected difference map (right) shows that <sup>137</sup>Cs-labeled particles continued to focus toward centers of depositional basins with inventory losses at the margins.

A survey of the area 10 years later (Fig. 4, center panel) showed that the inventory had increased toward the center of the depositional basin but was depleted around the margins (Fig. 4, right panel). An increase in the inventory at the basin center of about 50% in ten years corresponds to a rate of 0.05 per year or a time constant of about 20 years. The continuing post-fallout focusing of <sup>137</sup>Cs is due to cycles of re-suspension, horizontal transport, and re-deposition of <sup>137</sup>Cs labeled particles.

## Particle Removal Times in the Benthic Boundary Layer

For the past 25 years, settling particles have been periodically collected by (benthic) traps deployed from 1-20 m above bottom at four sites (12, 65, 100 and 160 m water depth) indicated in Fig. 5. Samples were collected by Argonne National Laboratory at the 65m site in 1976-1977 (D. Nelson and M. Wahlgren). Remaining samples were collected by GLERL. Traps were deployed, mainly during the period between late fall and early spring, when the water column was unstratified, for time intervals ranging from a few months to somewhat less than one year. Some trap deployments extended into the late spring-early fall season when the lake was stratified. In these cases samples contained small amounts of authigenic materials depleted in <sup>137</sup>Cs. Shown in Fig. 6, are decay-corrected <sup>137</sup>Cs/<sup>40</sup>K ratios vs time in the samples. This long-lived (1.3 x 10<sup>9</sup> years) naturally occurring isotope of potassium is principally a constituent of clay minerals which bind <sup>137</sup>Cs. Normalization to <sup>40</sup>K, co-determined with <sup>137</sup>Cs, removes small effects of dilution by authigenic materials such as biogenic silica, organic carbon and calcite.

In all four cases, ratios decline exponentially over about a 25 year period with time constants at the 12, 65, 100 and 160 m sites of 13, 20, 40 and 60 years, respectively. These constants reflect the time for replacement of <sup>137</sup>Cs-labeled particles by pristine or <sup>137</sup>Cs-deficient material from eroded shoreline bluffs or watershed soils. Thus labeled particles clear out of the water near the bottom almost five times faster at the 12m inshore site than they do at the deepest point (160 m site). Since <sup>137</sup>Cs is a surrogate for the behavior of atmospherically delivered, particle seeking trace constituents, the time constants characterize exposure of biota to a host of non-degradable contaminants in these benthic habitats. The trap results are consistent with the observations of changes in the inventory of <sup>137</sup>Cs in this area between 1972 and 1982. Most important, the results show that the atmospheric flux is strongly time averaged prior to delivery of <sup>137</sup>Cs to sediments and that the time constant is site specific but not a property of sediments themselves. Rather it's the constant characterizing first-order time averaging of the atmospheric <sup>137</sup>Cs flux as "seen" from a particular location on the lake floor. This is an important finding for modeling sedimentary profiles of <sup>137</sup>Cs.

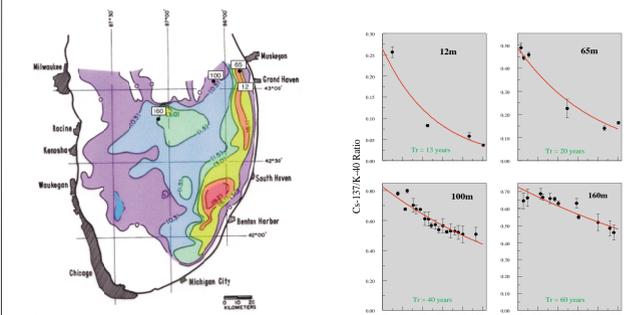


Fig. 5. Thickness of the Waukegan Member of the Lake Michigan Formation (meters) in southern Lake Michigan (Lineback and Gross, 1972). Mean age of the layer is about 3500 years b.p. (Lineback et al., 1971). Long-term benthic trap sites at 12, 65, 100 and 160-meter water depths are indicated as solid red circles.

Fig. 6. Decay-corrected <sup>137</sup>Cs/<sup>40</sup>K ratio in sediments collected by near-bottom traps at the 12, 65, 100 and 160 m sites (Fig. 5) about 1975 and 2000. Declines are essentially exponential (solid red lines) with time constants of 13, 20, 40 and 60 years, respectively. These characterize replacement of the cohort of <sup>137</sup>Cs-labeled particles by pristine or <sup>137</sup>Cs-depleted material eroded from shoreline bluffs and watershed soils.

## The Inventory of <sup>137</sup>Cs in Lake Michigan (1992-1996)

The recent lake-wide inventory of <sup>137</sup>Cs (Fig. 7) is expressed as a focusing factor (FF) defined as the ratio of the sediment inventory at a coring site to the time-integrated atmospheric flux up to the date of core collection. The pattern of accumulation in the southern part of the lake still (after 20 years) resembles previously determined patterns (Fig. 4, left and center). At the site of most intense focusing, more than 4 times as much <sup>137</sup>Cs has accumulated than expected from direct atmospheric fallout (i.e. FF>4). The pattern of small areas with high FFs centered within depositional basins continues northward parallel to the shoreline and then moves toward the center of the lake. By 1995 about 70% of the entire <sup>137</sup>Cs inventory can be accounted for by cores from depositional basins. If the 134 cores adequately represent depositional basin inventories, then a remaining 30% must still reside somewhere in "non-depositional" areas indicated approximately by light gray shading in Fig. 7. To proceed with estimating time scales for focusing of particles in the lake, it is essential to consider vertical distributions (profiles) of <sup>137</sup>Cs and the meaning of their lack of correspondence with the atmospheric fallout record.

## Profiles of <sup>210</sup>Pb and <sup>137</sup>Cs in Un-Mixed Sediments: An Example

To understand the <sup>210</sup>Pb data, it is necessary to consider a few primary facts concerning the origin of this radionuclide and its transfer of sediments. The source of <sup>210</sup>Pb is the long-lived radionuclide <sup>226</sup>Ra that occurs widely in crustal rocks and soils. Decay of <sup>226</sup>Ra produces the short-lived, unreactive gas <sup>222</sup>Rn, some of which escapes from mineral matrices into the atmosphere and decays through a series of extremely short-lived progeny to long-lived <sup>210</sup>Pb. This generalized pathway is shown here:

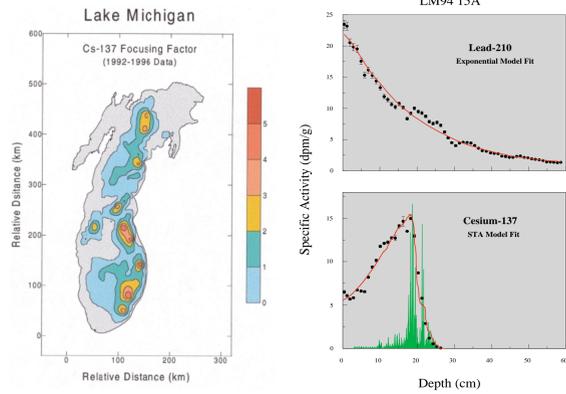
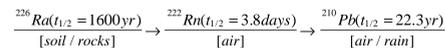


Fig. 7. <sup>137</sup>Cs focusing factor contour map for Lake Michigan (1992-1996). Three decades beyond the fallout period, labeled particles have focused intensely within depositional basins. In the southern part of the lake, focusing factors are greater than 4 at some sites. To create this contour map, the inventory has been set equal to zero at the shoreline.

Fig. 8. (upper) Distribution of <sup>210</sup>Pb (solid black circles) in sample core LM94-15A where there is no evidence of near-surface sediment mixing. The activity decreases to a background supported by in situ radium decay and is well described (dashed red line) by a simple exponential model (Eq. 1). (lower) Distribution of <sup>137</sup>Cs (solid black circles). Even in the absence of sediment mixing, the profile bears little resemblance to the loading of <sup>137</sup>Cs to the lake (green line). A system time averaging (STA) model (Eq. 2) provides an excellent description of the profile (solid red line) for a time constant (T<sub>s</sub>) of 21.1 years.

Once formed, <sup>210</sup>Pb is rapidly removed from the air by precipitation and dry deposition and, when delivered to water bodies is most often rapidly transferred to sediments. Since sediments themselves usually contain <sup>226</sup>Ra, there will be some <sup>210</sup>Pb supported by *in situ* radium decay. In many cases the rate of supply of <sup>210</sup>Pb to sediments can be safely assumed to be constant so that, in un-mixed sediments accumulating at a constant rate, excess <sup>210</sup>Pb (total minus supported) should decrease exponentially down core as a result of radioactive decay. A total <sup>210</sup>Pb profile may be calculated from constant net rates of supply to sediments of excess <sup>210</sup>Pb as F<sub>0</sub> (dpm cm<sup>-2</sup> yr<sup>-1</sup>) and mass, m (g cm<sup>-2</sup> yr<sup>-1</sup>), respectively. Total <sup>210</sup>Pb activity (dpm g<sup>-1</sup>) is then,

$$A_s(g) = (F_0/r_s) \exp(-\lambda g/r_s) + A_0 \quad (1)$$

where g (g cm<sup>-2</sup>) is the cumulative weight of sediment at a given depth, A<sub>s</sub> is the supported activity (dpm/g) and λ is the radioactive decay constant (0.69315/t<sub>1/2</sub> = 0.03114 yr<sup>-1</sup>). The age of a sediment layer relative to the core collection date is t (years) = g/r<sub>s</sub>.

An example of <sup>210</sup>Pb and <sup>137</sup>Cs profiles in unmixed sediments is given here (Fig. 8) for site LM94-15A at the center of the high depositional basin in the southern part of the lake. Out of 134 sites, only 18 have un-mixed sediments and these typically occur in regions of maximum sediment deposition. Total <sup>210</sup>Pb (excess-supported) decreases with depth (Fig. 8, top panel, solid black circles) to a level supported by in situ radium decay. The theoretical curve (Fig. 8, solid red line) based on Eq. 1 yields a least-squares optimized mass accumulation rate of 0.25 g cm<sup>-2</sup> yr<sup>-1</sup>. The <sup>137</sup>Cs profile (Fig. 8, lower panel, solid black circles) has little resemblance to the atmospheric fallout record plotted on the appropriate depth scale (Fig. 8, green line). The reason is provided by the results of the trap study: <sup>137</sup>Cs fallout has been time averaged with a time constant (T<sub>s</sub>) specific to this location. Thus the theoretical <sup>137</sup>Cs profile in unmixed sediments should be proportional to the <sup>137</sup>Cs flux to sediments, F<sub>s</sub> (dpm cm<sup>-2</sup> yr<sup>-1</sup>), obtained as a solution to a first-order system time-averaging (STA) model,

$$dF_s/dt = \lambda_s F_s - (\lambda_a + \lambda_s) F_s(t) \quad (2)$$

where F<sub>s</sub>(t) is the time-dependent, atmospheric <sup>137</sup>Cs flux, λ<sub>s</sub> is the radioactive decay constant (=0.023 yr<sup>-1</sup>), and λ<sub>a</sub> = 1/T<sub>s</sub>. The red curve shown in Fig. 8 is the optimized STA model fit to the data for a time constant T<sub>s</sub> = 21.1 years. Note that time-averaging causes significant amounts of <sup>137</sup>Cs to remain in surface sediments more than 20 years after the fallout period. Values of T<sub>s</sub> derived from STA analysis, should be considered residence times of particles in the benthic boundary layer at specific sites. They are comparable to times determined from the long-term trap study.

## Profiles in Near-Surface Mixed Sediments: An Example

Profiles of <sup>210</sup>Pb and <sup>137</sup>Cs in core LM94-24A (Fig. 9) exemplify the many cases where near-surface sediments have been biologically or physically mixed. In the upper 4.5 cm (Fig. 9, upper panel, dashed vertical green line), the distribution of <sup>210</sup>Pb is flatter than the exponential shape expected from Eq. 1. Similarly, the profile of <sup>137</sup>Cs above the same depth is flatter than expected from the exponential decrease predicted by the STA model Eq. 2 (Fig. 9, lower panel). A remedy is to treat near-surface sediment mixing as a steady state diffusive process and to replace the simple vertical sediment transport (VST) model above (Eq. 1), with a second-order equation of the form:

$$(\partial A / \partial t) = \partial [D_b(\partial A / \partial z)] \partial z - \partial(wA) / \partial z - \lambda A \quad (3)$$

where A is the activity of the radionuclide (dpm cm<sup>-3</sup>), z is the sediment depth below the interface (cm), D<sub>b</sub> is the depth-dependent mixing coefficient (cm<sup>2</sup> yr<sup>-1</sup>) and w is the depth dependent linear accumulation rate (cm yr<sup>-1</sup>). In the revised VST model, D<sub>b</sub> is essentially constant above some critical mixing depth, Z<sub>cr</sub>, and zero below it. The boundary condition is that the flux of tracer to the sediment interface, F<sub>s</sub>, derived from the STA model (Eq. 2) must match the flux into sediments:

$$F_s(t) = [D_b \partial A / \partial z + wA]_{z=0} \quad (4)$$

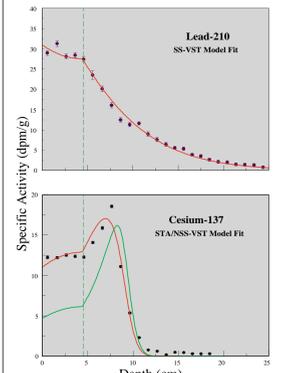


Fig. 9. (upper) Distribution of <sup>210</sup>Pb (solid black circles) in sample core LM94-24A where near-surface sediments appear to be mixed. A 4.5-cm zone of mixing, common to both radionuclides, is shown as dashed vertical green lines. A steady-state vertical sediment transport (SS-VST) model (Eq. 3 and 4) is used to generate the optimized fit to the data (solid red line). (lower) Distribution of <sup>137</sup>Cs (solid black circles) with the non-steady-state NSS-VST model fit (solid red line). The optimized STA model fit (T<sub>s</sub>) is 10.4 years. The model profile of <sup>137</sup>Cs without system time averaging (i.e. T<sub>s</sub> = 0), is shown as the green curve.

For excess <sup>210</sup>Pb the atmospheric flux is essentially constant and it is assumed that the flux to sediments is constant as well. Since, in principle, the flux of <sup>210</sup>Pb to the lake is also subject to time-averaging, its effect is to smooth out short term (< yearly) variations in its delivery to sediments. Hence, a steady state form of the VST model (i.e. SS-VST) is used for <sup>210</sup>Pb, i.e. ∂A/∂t = 0. For <sup>137</sup>Cs, the non-steady state VST model (Eq. 3 and 4) above is used (i.e. NSS-VST).

Optimized SS-VST model results for <sup>210</sup>Pb are shown in Fig. 9 (upper panel, solid red line) with D<sub>b</sub> = 2.48 cm<sup>2</sup> yr<sup>-1</sup>; Z<sub>cr</sub> = 4.13 cm, corresponding to a mass per unit area in the mixed layer, g<sub>mix</sub>, of 0.23 g cm<sup>-2</sup>, and r<sub>s</sub> = 0.67 g cm<sup>-2</sup> yr<sup>-1</sup>. If the mixed layer is a steady-state feature then the residence time of particles in it, T<sub>mix</sub>, equals g<sub>mix</sub>/r<sub>s</sub>. The mixed-layer residence time also is a measure of the best time-resolution achievable in reconstructing historical records from core profiles at the site. To apply the NSS-VST model to <sup>137</sup>Cs, values of D<sub>b</sub> and Z<sub>cr</sub> from <sup>210</sup>Pb were kept fixed while r<sub>s</sub> was allowed to vary. Mean sediment accumulation rates based on the two methods may be expected to differ slightly. However the principal parameter obtained for <sup>137</sup>Cs is the STA time constant. The result of this approach is shown in Fig. 9 (lower panel, solid red line) for optimized parameter values of r<sub>s</sub> = 0.053 g cm<sup>-2</sup> yr<sup>-1</sup> and T<sub>s</sub> = 10.4 years. If there were no time-averaging (i.e. T<sub>s</sub> = 0) the calculated profile (green curve) cannot match the data.

## Distributions of Particle Residence Times T<sub>s</sub> and T<sub>m</sub>

This study recognizes two categories of site-specific particle residence times in benthic boundary layer of Lake Michigan. One (T<sub>s</sub>) characterizes the time for removal of a cohort of fine particles from the benthic boundary layer and their replacement by particles originating from external sources, and the other (T<sub>m</sub>) characterizes the time for transfer of particles from the mixed layer within depositional basins to deeper un-mixed sediments. If resuspension of particles from sediment mixed layers re-supplies the benthic boundary layer, these time constants are related and should have comparable magnitudes. The distribution of T<sub>s</sub> (Fig. 10, upper panel) is grouped around a geometric mean of 15 years with small numbers of values greater than 30 years. The distribution of T<sub>m</sub> (Fig. 10, lower panel) is comparable, centering around a geometric mean of about 12 years.

## Evolution of <sup>137</sup>Cs Focusing Maps

If the STA/VST models accurately describe <sup>210</sup>Pb and <sup>137</sup>Cs profiles, then three simple steps permit construction of past and future focusing factor maps. First, the <sup>137</sup>Cs flux to sediments, F<sub>s</sub>(t), at each site from 1950 onward may be calculated from Eq. 2 using the site-specific time constant, T<sub>s</sub>. Second, the relative inventory, I<sub>s</sub>(t), may be determined by time integrating decay corrected values of F<sub>s</sub>. The absolute inventory, I<sub>a</sub>(t) = F<sub>s</sub>I<sub>s</sub>, may then be determined by choosing the normalization factor F<sub>n</sub> so that I<sub>a</sub> equals the measured inventory at the time of core collection. Finally the inventory delivered to the lake, I<sub>l</sub>(t), is the time-integrated, decay-corrected atmospheric flux up to core collection time. Hence FF = I<sub>a</sub>(t)/I<sub>l</sub>(t). A final step may be taken in creating contour maps. Since the sum of focusing factors over the whole lake must equal its area, the mean focusing factor over "non-depositional" areas may be calculated as FF<sub>nd</sub> = (1 - I<sub>dep</sub>/FF<sub>dep</sub>)/(1 - I<sub>dep</sub>). FF<sub>nd</sub> is the average focusing factor over the "depositional" region and I<sub>dep</sub> is the corresponding fraction of the lake area.

The resulting contour maps show the striking feature that, during the period from 1955 through 1965 (Fig. 11, uppermost three panels) when <sup>137</sup>Cs was mainly delivered to the lake, most was stored over non-depositional areas. During subsequent years, focusing has intensified as inventories build up in central areas of depositional basins with losses from the margins and non-depositional regions. In the absence of further loading of <sup>137</sup>Cs, STA/VST models predict that focusing will continue as illustrated by the contour maps for years 1970-2050 (Fig. 11). Even by 2050 <sup>137</sup>Cs and Pu isotopes should remain easily detectable in sediments and the predictions made here could be rather readily verified. A final result of interest is the essentially exponential decrease in FF<sub>nd</sub> (Fig. 12) indicating a residence time of about 42 years for <sup>137</sup>Cs stored over non-depositional areas. This value accords well with known changes in trap-determined inshore-offshore changes in particle residence times within the benthic boundary layer.

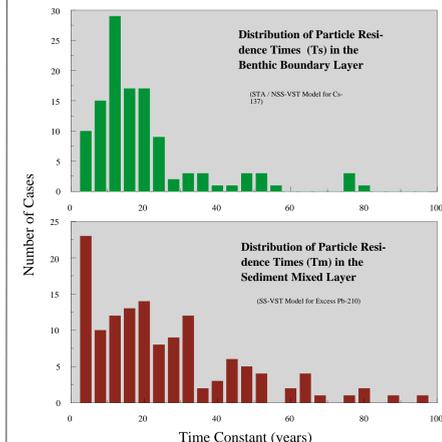


Fig. 10. (upper) Distribution of particle residence times (T<sub>s</sub>) in the benthic boundary layer inferred from fitting <sup>137</sup>Cs profiles. The geometric mean time is about 15 years. At about 10% of the sites (bar not shown) times are in excess of 100 years. (lower) Distribution of particle residence times (T<sub>m</sub>) in the sediment mixed layer inferred from fitting excess <sup>210</sup>Pb profiles. The geometric mean time is 12 years. At approximately 10% of the sites (leftmost vertical bar) profiles exhibit little or no mixing (T<sub>m</sub> < 1 year).

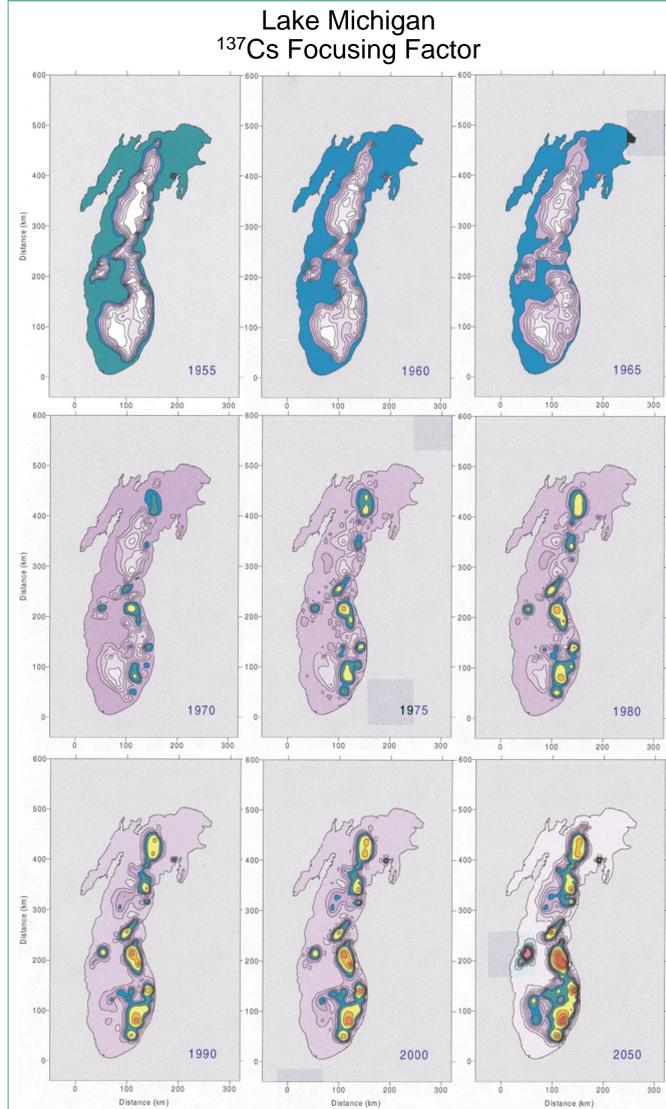


Fig. 11. Evolution of the focusing factor contour map for <sup>137</sup>Cs based on STA/VST analysis. "Non-depositional" areas outside the region of corable sediments, have been assigned color values corresponding to the mean focusing factor needed to make the lake-wide, contour-weighted average value equal 1.00. During the primary period of fallout (1955-1965), focusing factors within depositional basins are less than unity (areas with light violet hues). See color index on the right. The deficit is made up by temporary storage of <sup>137</sup>Cs over non-depositional areas (colored green and blue indicating FF values > 1). After about 1970, when atmospheric fallout is no longer significant, focusing of <sup>137</sup>Cs intensifies due to internal redistribution processes (note the projection to 2050).

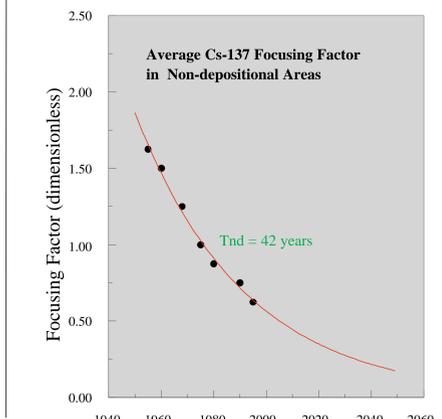


Fig. 12. Time-dependence of the average <sup>137</sup>Cs focusing factor in the "non-depositional" region (solid black circles). The exponential fit (solid red line) indicates a mean particle residence time of 42 years.

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