

# Environmental Radiotracers

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## Background and Objectives

The Environmental Radiotracers (ERT) Project employs natural and artificial radionuclides to identify and model important particle transport processes in diverse systems including the Laurentian and other Great Lakes, smaller freshwater bodies, wetlands and coastal marine environments. The project is an outgrowth of the recognition that, in such systems, many contaminants and nutrients in the water move in association with inorganic and organic particles, including plankton species and their remains. Particle-associated constituents settle through the water column to underlying sediments, where they may be mixed and resuspended by currents or biological action, and are ultimately lost by burial in accumulating sediments. Along such pathways as these, rates of particle transport often can be determined using particle-associated radionuclides because of their built-in clocks, relative ease of measurement or accurately known loading histories. Radionuclide studies often inform development of quantitative process models and this aspect has been emphasized in the ERT Project.

Since its inception in the early 1980s, this technique-based project has given particular attention to radiometric dating of sediments. In addition to the role that sediments play in the regulation of nutrients and contaminants in aquatic systems, they frequently possess retrievable records of present and historical changes in ecosystem status and constituent loads due to natural or human causes. The development, application and evaluation of radionuclide dating methods address numerous scientific and public concerns about the changing status of ecosystems, environmental contamination and global climate change.

Changes in contamination, biological status and physical characteristics of lakes and coastal marine systems during the past century often lead to significant changes in the composition of accumulating sediments. Reconstructing the history of such changes from sedimentary records (paleolimnology) is an important part of present efforts to understand human impacts on ecosystems and undertake appropriate remedial strategies. To this end, it is critical to have reliable and accurate methods of dating sediments. The Great Lakes Environmental Research Laboratory has been at the forefront of developing and testing radiotracer methods for sediment dating.

The ERT Project consists of a series of sub-projects with differing duration, environments and collaborators, which generally share the above objectives. Sub-projects are a mix of internally-funded activities and formal externally-funded inter-agency agreements. Over the lifetime of this project there have been about 30 sub-projects in diverse systems and geographic areas.

## **Geographic Coverage**

Lake Athabaska (Canada), Lake Baikal (Russia), Lake Constance (Germany/Austria/Switzerland), Coeur D'Alene Lake (Idaho), Florida Bay, Lake George (US/Canada), Great Slave Lake (Canada), Lake Ladoga (Finland/Russia), Lake Oahe (S. Dakota), Lake Rockwell (Ohio), Lake Sniardwy (Poland), Lake Tahoe (California/ Nevada), Lake Winnipeg (Canada), Terrace Lake (Colorado), The Everglades (Florida), The Gulf of Mexico, The Keweenaw Waterway (Michigan), The Laurentian Great Lakes (US/Canada).

## **Formal and Informal Institutional Collaborations**

- Australian Institute of Marine Science, Queensland, Australia
- Argonne National Laboratory, Argonne, IL
- Canada Centre for Inland Waters, Burlington, Ontario, Canada
- Case Western Reserve University, Cleveland, OH
- Freshwater Institute, Winnipeg, Manitoba, Canada
- Department of Geology, University of Michigan, Ann Arbor, MI
- Great Lakes Research Division, Institute of Science and Technology, University of Michigan, Ann Arbor, MI
- Great Lakes Water Institute, University of Wisconsin, Milwaukee, WI
- Illinois Geological Survey, Champaign-Urbana, Illinois
- Limnological Institute, Irkutsk, Russia
- Ministry of the Environment, Toronto, Ontario, Canada
- Ministry of the Environment, Saskatoon, Saskatchewan, Canada
- Michigan Technological Institute, Houghton, Michigan
- Pacific Marine Environmental Research Laboratory, NOAA, Seattle, WA
- School of Public Health, University of Michigan, Ann Arbor, MI
- South Florida Water Management District, West Palm Beach, FL
- University of Florida, Gainesville, Florida
- University of Minnesota, Minneapolis, MN
- U.S. EPA: Duluth, MN, Large Lakes Research Station, Grosse Ile, MI, Region V, Enforcement, Chicago, IL
- U.S.G.S., Atlanta, GA, Denver, CO, Menlo Park, CA, Reston, VA, St. Petersburg, FL, Woods Hole, MA
- University of Constance, Constance, Germany
- University of Joensuu, Joensuu, Finland
- Dept. of Geological Sciences, University of Wroclaw, Wroclaw, Poland

## Radionuclides Emphasized in the Environmental Radiotracers Project



**Lead-210:** This naturally occurring radioactive isotope of lead ( $t_{1/2} = 22$  years) is part of the uranium decay series that includes radium. This latter radionuclide occurs widely in soils and rocks and produces the well-publicized radioactive, chemically inert gas, radon, which leaks into the atmosphere. Decay of radon ( $t_{1/2} = 4$  days) produces  $^{210}\text{Pb}$ , which is efficiently removed by precipitation, transferred to water bodies and rapidly incorporated into sediments where it decays on burial. As a result, a layer of sediment, for example containing half as much  $^{210}\text{Pb}$  than at the surface, would be considered 22 years older than sediments deposited at the surface and so forth. This simple idea has often proven to work well in many of the hundreds of studies that have employed the  $^{210}\text{Pb}$  method during the past several decades. However the method is never routine because sediments may accumulate at variable rates, be subject to near-surface mixing due to biological or physical processes, and even the rate of delivery of  $^{210}\text{Pb}$  to sediments may vary as a result of physical or geochemical processes in aquatic systems. Some of the sub-projects described involve verification of the method, offer approaches to improving its accuracy and attempt to link  $^{210}\text{Pb}$  dating to system specific physical and biogeochemical processes. In other sub-projects the method is applied to obtain modern rates of sediment accumulation, rates and ranges of sediment mixing and residence times of particles in sediment surface mixed layers. In some cases the  $^{210}\text{Pb}$  has been used to date cores where there is an interest in linking sedimentary records of water body contamination to historical loading from urban or industrial sources.



**Cesium-137:** This man-made radionuclide ( $t_{1/2}=30$  years) was delivered to aquatic systems primarily through atmospheric fallout of debris from above ground testing of nuclear weapons mostly in the mid-1960s. In the Great Lakes,  $^{137}\text{Cs}$  has attached to fine particles and cleared out of the water within a few years after fallout, but small amounts continue to be reintroduced through resuspension of surface sediments. In cores from selected areas of many lake and coastal marine sediments as well as wetland soils, there is a clear peak present at depths corresponding to the fallout maximum in 1963-1964. Thus the  $^{137}\text{Cs}$  is useful in verifying sediment cores dates based on  $^{210}\text{Pb}$ , but perhaps more important,  $^{137}\text{Cs}$  mimics the behavior of many non-degradable contaminants circulating in aquatic systems. Since its loading to the Great Lakes as well as other continental sites, is well-known and  $^{137}\text{Cs}$  is unambiguously and easily determined in sediments, this fallout radionuclide can be used to track the movements of contaminants through systems during the past 40 years or so. Some of the sub-projects described below exploit  $^{137}\text{Cs}$  to establish or verify sediment accumulation rates while others exploit its particle-tracking capability to characterize the long-term fate of contaminants. In the Great Lakes as in many other aquatic systems, particles that were labeled by  $^{137}\text{Cs}$  in the mid sixties are in the process of being buried and replaced by new, largely uncontaminated particles from erosion of shoreline of watershed sources. As a result  $^{137}\text{Cs}$  is useful in characterizing mechanisms and times of recovery of water bodies from past contaminant insults. Another fallout radionuclide mentioned in several sub-projects is plutonium, primarily the long-lived isotope,  $^{239}\text{Pu}$ , ( $t_{1/2} = 24,400$  years). Plutonium was co-dispersed with  $^{137}\text{Cs}$  in nuclear testing events and deposited essentially in fixed proportion to radiocesium onto land and water surfaces. Although Pu is chemically dissimilar to cesium, both elements have labeled suites of particles that have had comparable transport and fate in the Great Lakes and many other freshwater systems.

**Beryllium-7:** In contrast with the above radionuclides, naturally occurring  $^7\text{Be}$  has a comparatively short half-life ( $t_{1/2} = 53$  days) and results from the smashing (spallation) of stratospheric nitrogen and oxygen nuclei by cosmic rays. Tropospheric concentrations of  $^7\text{Be}$  and rates of deposition onto surface water have an annual cycle that depends on a combination of its seasonally variable efficiency of transfer from the stratosphere and on regional weather

patterns. In the Great Lakes area, the amount of  $^7\text{Be}$  transferred to water varies almost sinusoidally each year, with maximum deposition in May and minimum in December-January. In several recent sub-projects, the seasonal variability and short half-life of this particle-tracing radionuclide has been used to characterize transport processes and their rates on weekly to monthly time scales.

### **SP-00 Saginaw Bay**

Between 1975 and 1978, with the support of the USEPA (Large Lakes Laboratory, Gross Ile, Michigan) we collected sediment cores from Saginaw Bay to determine sediment mixing and accumulation rates, and to establish, if possible, a history of contaminant metal accumulation. In addition surveys were made of the distribution and abundance of benthic macroinvertebrates. Their influence on sediment-water exchange of nutrients was determined at the time by measurement of concentrations in water overlying intact, incubated sediment cores. The results were published as a final report.

### **Accomplishments**

An old study was resurrected at the request of Don Schloesser. Determined profile of radiocesium in a core analyzed for Hexagenia tusks. Developed and applied a model to obtain sediment chronology.

We contributed to a chapter (Soster, Matisoff, McCall and Robbins) entitled "In situ effects of organisms on pore water geochemistry in Great Lakes sediments."

### **SP-1 Lake Erie Reference Sites**



This project, which predates the existence of the Environmental Radiotracers Project at GLERL, was the outgrowth of sediment deposition studies in Lake Erie initiated in about 1970 by the Canada Center for Inland Waters (CCIW). The Canadian group showed that, at selected sites in the deep eastern basin of the Lake, sediments accumulate at a higher rate than in any other profundal areas of the Great Lakes - more than a cm/year. Sediment cores from this area were ideally suited for high-resolution reconstruction of the dramatic ecological and biogeochemical changes that have occurred in Erie during the past century. With the start of the ERT project at GLERL in 1980, sediment materials and results of radiometric measurements for cores collected (by JAR then at U. Michigan) in 1976 and 1978 became available to the project as well as core materials collected in 1971 by the Canadian group. Subsequently, gravity and box cores were collected from the Eastern Basin Reference (EBR) sites in 1981, 1982, 1983, 1985 and 1988 from the C/S Limnos with generous support from CCIW, and in 1991 from the R/V Neeskay, Great Lakes WATER Center, U. Wisconsin. These ship- of-opportunity expeditions and subsequent radiometric analyses of sediment samples have created an archive of well-dated sediment cores spanning a period of more than twenty years. While several papers have been published based on these archives, the resource has been underutilized due to the pressures in recent years to conduct externally funded research projects.

### **2005 Plans**

- Complete analysis of sediment samples for  $^{239}\text{Pu}$  from a core collected in 2003. Kent Orlandini at Argonne National Laboratory is doing the analysis.
- Finish writing a paper on the history of accumulation of long-lived fallout radionuclides at the eastern basin reference site (EBRS) in Lake Erie: J. A. Robbins, K. A. Orlandini, N. R. Morehead, D. N. Edgington and A. Mudroch, Accumulation of fallout  $^{137}\text{Cs}$  and  $^{239+240}\text{Pu}$  in sediments at a long term monitoring site in eastern Lake Erie. J. Great Lakes Research. (presently 50% complete).
- Analyze the 2003 core for selected trace metals by neutron activation analysis. Work may be done by L. Minc, Oregon State University.

### **2004 Accomplishments**

- Completed processing core collected in 1993.
- Completed analysis for  $^{137}\text{Cs}$  and Total phosphorus (Johengen)
- Prepared samples and began counting samples for  $^{210}\text{Pb}$  and  $^{226}\text{Ra}$
- Ran STA2 Model (time-dependent integration) for  $^{137}\text{Cs}$  profile.
- Contracted with K. Orlandini (ANL) to measure Pu-239 activity
- Initiated INAA with L. Minc, Oregon State U., NAA Laboratory
- Provided BJE and PM with mass accumulation rate data for their study of in situ degradation rates of selected organics.

## Accomplishments

Continuing our investigations of Mn profiles at the eastern basin reference sites (EBRS), we have discovered that, between 1850 and 1960, periodic fluctuations in concentrations of sedimentary manganese were inversely and strongly correlated with water level. Most likely this occurs because fine sediments reaching the coring site variably dilute sources of manganese that are at least partially independent of water level. Sediments in the eastern basin are derived largely from water-level sensitive erosion of high bluffs along Long Point and the Canadian shoreline further west. After 1960, the Mn- water level relation breaks down. Much more Mn is present in the sedimentary record than can be accounted for by water level fluctuations alone. This is due to our previously discovered connection between Mn (and excess Pb-210) profiles and hypolimnetic oxygen depletion above sediments in the central and western depositional basins.

Synthesis of sediment accumulation rate and surface elemental concentrations published by others. Our investigation has produced a contour map of excess Mn to sediments in Lake Erie that reveals very significant rate of accumulation of excess Mn in the eastern basin. This result strongly supports our central thesis of anthropogenic modification of biogeochemical processes affecting  $^{210}\text{Pb}$  dating of sediments at EBR sites. Finally, we have a paper in press for the Proceedings of the South Pacific Environmental Radioactivity Association.

All radiometric and elemental analyses were completed. Model calculations were completed, as well as most figures for publication and presentations.

We examined possible reasons for systematic irregularities in sediment profiles of excess  $^{210}\text{Pb}$  that previously had been ascribed to near-surface mixing or to recent increases in rates of sediment accumulation. Our investigations of certain inconsistencies in these two explanations lead to an alternative more self-consistent theoretical treatment the lead-210 data.

The alternative model is based on the observation that, when hypolimnetic waters over western and central basins of Lake Erie are depleted in oxygen during the late summer and early fall, concentrations of manganese increase substantially as a result of dissolution of recently deposited detrital materials and near-surface sediments. According to speculations of Nriagu and Burns, excess soluble Mn should re-associate with particles as waters oxygenate during overturn. Then, because of prevailing currents, particle-bound Mn should be transported to and accumulate in sediments of the eastern basin. Since lead is known to associate with Mn, it seemed likely that the deposition of the radioactive isotope of lead,  $^{210}\text{Pb}$ , used as a sediment dating tool, might also be perturbed by recurrent hypoxic episodes in Lake Erie. Our highly-resolved profiles of Mn demonstrate the connection with  $^{210}\text{Pb}$ . To construct accurate chronologies based on this radionuclide, the influence of Pb-Mn interactions had to be quantitatively modeled. Having done that, we were able to use the Mn record as a surrogate to estimate the degree of near-bottom hypoxia in Lake Erie for the last century. We also illustrate sharp, transient increases in sediment Mn associated with pronounced hypoxic episodes in the 1960s and 1970s as determined from lake-wide surveys by others.

Our results show that increases in the temporal-spatial extent of hypoxia in Lake Erie started around the turn of the century and were not just limited to the recent period of major phosphorus-induced eutrophication of the system.

## **SP- 2 Composition and Flux of Settling Matter**

The general aim of this internally funded project is to gain insights into the characteristics of particle transport, particularly in Lake Michigan, by radiometric analysis of trap-collected materials. Traps are generally moored collectors of settling particles which, in this study, were deployed for times ranging from weeks to months. Our current goal is to estimate removal times of  $^{137}\text{Cs}$ -labeled particles resuspended into near-bottom (benthic) traps located 5-10 meters above bottom at selected sites in the southern part of Lake Michigan. During the past several decades, concentrations of  $^{137}\text{Cs}$  and plutonium in surface waters from diverse locations in the southern part of the lake have decreased exponentially with a characteristic time of about 20 years. This decrease, which refers to long-term changes in surface waters when the lake is isothermal and well-mixed vertically, most likely reflects the average time required for  $^{137}\text{Cs}$ - (or Pu-) labeled particles to leave the pool of resuspendable sediments by burial or exportation to other areas of the lake. However, since the pool of sediments in question appears to be highly focused on the lake bottom, horizontal variations might occur in benthic particle residence times that were not seen at the top of the water-column.



## Accomplishments

We have completed most of the measurements of plutonium (Pu) activities of samples collected over 25 years from the southern part of the lake. The results indicate that, despite differences in geochemical behavior of Pu and  $^{137}\text{Cs}$ , these radionuclides that were delivered in fixed ratio to the lake mainly during the mid-1960, have behaved virtually identically in terms of their reintroduction to the lake from bottom sediments. An appropriately decay-corrected ratio of Pu/ $^{137}\text{Cs}$  in trap samples is essentially the same as the ratio in atmospheric loading to the lake. Thus in southern Lake Michigan at least, these fallout radionuclides are tracking the “cohort” of particles recycling through the system en route to burial in sediments or loss, to a much smaller degree, by outflow.

We also obtained the first sets of concentration data for 20 elements as determined by instrumental neutron activation analysis (INAA). The most significant result to date is the finding that (1) elemental composition near-bottom trap samples is comparable to underlying surface sediments, (2) manganese concentrations in near-bottom trap samples at a 250 m site in northern lake Michigan are markedly enriched in manganese. This highly significant finding is discussed further below (SP-44).

Additional data (from mid 1990s onward) have been added to the results for the four study sites as well as data from an additional mid-lake, 250 m site.

During the past several years a number of benthic trap samples from four selected sites (12, 65, 100 and 150 m) have been analyzed for  $^{137}\text{Cs}$  to develop a precise indication of changes in its concentration at and between sites over about a 15-year period. At these sites decay-corrected concentrations have declined quasi-exponentially with time constants of 10, 16, 35 and 80 years respectively. These time constants translate into removal rates of about 10, 6, 3 and 1.2 percent per year, respectively. The inshore-offshore trend evidenced in the data results from the more efficient resuspension of labeled-particles from inshore regions of the lake, their net exportation to deeper areas and replacement by unlabeled particles eroded from shoreline sources. The net outward propagation of labeled particles is a long-term consequence of cross-margin transport processes in this lake.

Since  $^{137}\text{Cs}$  is a surrogate for the behavior of many particle-associated contaminants, the time constants determined by this study may characterize spatial variations (and trends) in the persistence of non-degradable contaminants in benthic habitats. The results will be of interest to the scientific community, coastal transport process modelers, physical limnologists, oceanographers, benthic ecologists and organizations including EPA and the U. S. Geological Survey. The results have been used to formulate new approaches to interpreting and modeling contaminant profiles in recent lake and coastal marine sediments.

### **SP-3 Recent Sedimentation Rates in Lake Ontario**

A series of sediment box cores were collected from Lake Ontario in 1987 as part of an EPA sponsored project to determine sedimentation rates and focusing factors for mass balance calculations. At least two adjacent sub-cores were collected at the time from each box core, one sub-core for radionuclide analysis at our laboratory and a second sub-core for analysis of toxic organic contaminants (notably PCBs and dioxins) by EPA. The radionuclide measurements and initial models were developed and applied to the radionuclide data. A final report was delivered to EPA in 1989. Since that time, analytical results for PCBs and dioxins in selected cores were obtained and have been combined with information on concentrations in lake trout and the history of their abundance in Lake Ontario.

#### **Accomplishments**

We have completed the first draft of a major paper for submission to Environmental Science and Technology (Cook, Robbins, Endicott, Lodge, Walker, Zabel, Guiney, and Peterson) entitled "Effects of Ah Receptor Mediated Early Life Stage Toxicity on Lake Trout and Survival in Lake Ontario During the 20th Century".

We reviewed critical literature on sediment radionuclide chronologies and developed revised vertical sediment transport models as a result. In addition were compared <sup>137</sup>Cs distributions between adjacent sub-cores to obtain the most precise and accurate core date assignments.

### **SP-9 Sediment Focusing in Lake Erie**

The original project was based on analysis of <sup>137</sup>Cs of cores collected from the principal depositional basins of Lake Erie in 1976 and again in 1982. The changes in patterns of accumulation showed that toward the basin margins <sup>137</sup>Cs was lost while the storage toward the centers had increased, despite the absence of any significant new loading to the lake during this period. The results indicate that sediment focusing of particle-associated transient tracers continued long after the loading has ceased, presumably because of the seasonal cycle of resuspension and deposition.

More recent work has concerned the history of focusing of <sup>137</sup>Cs in the eastern depositional basin based on cores obtained as part of the Lake Erie Reference Site Study [SP-1] between 1971 and 1991. In these cores, radiocesium profiles resemble the history of fallout to the lake. During the 20-year observation period the depth of maximum <sup>137</sup>Cs concentration, corresponding to peak atmospheric fallout in 1964, has moved downward by more than 20 cm due to burial by new sediment. In post-fallout sediments concentrations of radiocesium have declined exponentially partly due to radioactive decay and partly due to decreases in the amount available to be delivered to the eastern basin site by resuspension of sediments from adjacent or remote areas of the lake bottom.

When corrected for radioactive decay, radiocesium profiles show that the time of residence of <sup>137</sup>Cs-labeled particles in the relevant reservoir of sediments contributing to the coring site is about 4 years. This is the time required for the concentration to decrease exponentially to about

37% of its initial value. Since radiocesium is a surrogate for many non-degradable contaminants, this time may characterize the recovery of the profundal benthic habitat following cessation of external loads to the lake.

### **Accomplishments**

The single residence time model provided an excellent quantitative account of radiocesium profiles in cores collected through the early 1980s but, beyond that, it underestimates observed concentrations. This year we developed a variable residence time (VRT) model to account for the difference. The idea behind the model is that particles within the relevant reservoir most easily resuspended and moved to the coring site contribute first and are depleted. It thus becomes increasingly difficult to mobilize particles remaining parts of the reservoir. The region of resuspendable sediments may be visualized as a landscape internal to the lake and, in that sense, analogous to processes known to occur in watersheds which control loss of contaminants by tributary outflow. The VRT model provides a complete description of all radiocesium profiles collected from the reference site over twenty years through the use of only two parameters: an initial particle residence time and its subsequent rate of change. According to the VRT model the residence time has increased from about 2 years to 50 years during the past two decades.

### **SP-11 Sediment Records of Contamination and Biologic Responses in Lake George**

The St. Mary's River, starting as outflow from Lake Superior, after a few km passes through the locks at Sault Saint Marie in the vicinity of the twin Canadian and U.S. towns with the same name. Already in the 18th century, the towns had arisen as a result of the fur trade and in the 19th century the region was subject to extensive logging as well as the development of the system of locks in the river. Starting in the 20th century, populations of both cities increased greatly as a result of new industrial activities including steel and paper production as well as leather tanning. As a result, the river has received massive loads urban/industrial contaminants and, by the 1950s, was identified one of the most impacted areas in the Great Lakes connecting channels. In the past twenty years, the system has begun to recover as a result of economically-driven declines in industrial activity and because of environmental regulation.

This study, involving both Canadian and U. S. Scientists, reconstructs the history of human impacts on the St. Mary's system by measurement of contaminants, nutrients and biological components in dated sediment cores from Lake George. This small lake, fed by the north branch of the St. Mary's River, is situated about 20 km down-river from urban/industrial sources and is ideally suited for the study. Sediment cores were diver collected from this lake in 1986 (MOE-ONT/CCIW) as part of the Upper Great Lakes Connecting Channel Study. In 1993, we obtained additional cores using advanced box coring (GLWI) methods. Cores, dated using natural and fallout radionuclides, have revealed changes in nutrient levels starting during the early period of colonial settlement and accelerated rise in nutrient loading and stimulation of plankton productivity during the logging period. Starting in about 1900 with industrialization, productivity declined greatly, presumably as a result of toxic contaminants, and has begun to turn around only in the last decade.

Currently this study focuses on historic changes that have occurred in the abundance and composition of diatoms (siliceous plankton), the connection between the trace element bromine and diatom abundance, and the story of chromium contamination. We are developing the connection between the history of chromium contamination of the river and the record of operations of a leather tanning company located in the U. S. Sault Ste. Marie. The company opened its doors for business in 1900, began using chromium salts instead of hemlock bark as a tanning agent in about 1915, and closed in the late fifties. The abandoned factory burned down in 1960s and the property ultimately became a USEPA superfund site. Company discharges were the primary cause of intense chromium contamination of the river system that peaked around the end of W.W.II, close to the time of maximum production of military leather goods. We show that features in sediment chromium records match the occurrence of primary events in the history of company operations. We use the pronounced sediment chromium spikes around 1945 in cores from distinct locations within Lake George as one of several time markers for confirming radionuclide chronologies. The study shows the potential for using chromium to track the dispersion of particles from the St. Mary's river out into Lake Huron. The Lake George study is a textbook example of the use of dated sediments to identify system contaminants, reconstruct their loading histories and biologic effects, to pinpoint offending sources, to chronicle the beneficial results of environmental regulation and illustrates the possible forensic use of well-placed, high-resolution sediment receptor sites.

### **Accomplishments**

Our major accomplishment has been the completion of a first draft of a paper for submission to the Canadian Journal for Fisheries and Aquatic Science by Reavie, Stoermer, Robbins, Douglas and Segieda, entitled "Post settlement microfossil succession in Lake George, a fluvial lake downstream of Lake Superior."

A paper was published by Tenzer et al., 1999 in Organic Geochemistry on anthropogenic changes in the composition of organic matter in Lake George. This year we began a collaborative project with E. Stoermer (U. Mich.) and E. Reavie (U. Toronto) to produce historical records of anthropogenic impacts on diatom abundance and composition as recorded in sediment cores from Lake George (St. Marys River system). Dr. Reavie is currently doing a thorough taxonomic analysis of sediment core materials. With our collaboration, Dr. C. Kerfoot began preparing a paper on the association of bromine with freshwater diatoms and biogenic silica with examples from connecting channel lakes: Lake George and Portage Lake. Note that the exoskeletons of diatoms are composed mainly of silica (silicon dioxide).

### **SP-16 Gamma Scan System**

A automated version of the Gamma Scan System at GLERL is the result of years of development and application to studies of the effects of benthic feeders on tracer profiles. The system precisely and accurately determines the vertical distribution of gamma-emitting tracers in sediment microcosms. In many studies, conveyor-belt (C-B) feeders are introduced into sediment cells with an initial surface layer labeled with gamma emitting <sup>137</sup>Cs. Their feeding action causes the layer to be buried by non-radioactive sediment brought to the surface. The rate of marker burial is a measure of collective sediment ingestion. Precise determination of

position of the marked layer over time and the inferred sediment reworking rate is the basis for examining the response of organisms to ambient conditions (temperature, dissolved oxygen), sediment characteristics (grain size, organic carbon content, bacteria) and to sediment contamination. During the past decade the system has been used mainly measure the effects of contaminants on the rates of sediment reworking by benthic organisms. Typically, concentrations of compounds, such as DDT, Endrin, Deildrin, required to observe changes in reworking rates are many orders of magnitude lower than doses lethal for organisms.

### **Accomplishments**

A paper entitled “Biological mixing responses to sublethal concentrations of DDT in sediments by *Heteromastus filiformis* (Capitellidae) using a  $^{137}\text{Cs}$  marker technique” by Mulsow, Landrum and Robbins has been submitted for publication to *Marine Ecology - Progress series*.

## **SP-33 Copper Mining Impacts in the Keweenaw Peninsula and Lake Superior**

### **Background**

Initially supported by the Great Lakes Protection Fund from 198x-198y, this collaborative study with Michigan Technological University, continues to assess the impact of copper mining, beginning toward the end of the 19th century in the Keweenaw Peninsula of Michigan, on the Keweenaw waterway, Portage Lake and Lake Superior. As a result of a 50-year practice of crushing ores to physically separate copper from rock matrices, mining companies created huge piles of stamp “sands” along selected areas of shorelines of the Waterway, Portage Lake and Lake Superior. Through annually varying erosion of stamp sands, the water bodies received major loads of copper and other metals, particularly from dispersion of fine, micron sized particles. And during the period of major mining operations, even the accumulation of sediments in Waterway lakes such as Portage, was dominated by direct dumping and erosion of stamp sands.

Our initial work using  $^{210}\text{Pb}$  and  $^{137}\text{Cs}$  showed that in Portage Lake cores, varve structures, i.e. periodic layering of sediments revealed by x-radiographs of intact sediment cores, were indeed the result of annual erosion of stamp sands. We also showed, by analysis of zooplankton remains in sediments, that the abundance of certain species was suppressed during much of the mining discharge era. We subsequently showed that sediments contained viable “resting” eggs of certain zooplankton. These eggs could be reactivated in the lab to look (1) for morphological or possibly genetic changes over time that might be associated with variable sediment composition and (2) for systematic differences in copper toxicity to resting egg grown zooplankton. We also showed that stamp sand piles at different sites in the Waterway had distinctive elemental characteristics reflecting variations in source ore deposits. As a result, we could separate contributions of various stamp sand piles and natural, pre-mining sources to coring sites in the lake.

More recently the study has pursued early indications of widespread dissemination of elemental contaminants in stamp sand fines into Lake Superior by measuring copper, other signature elements as well as radionuclide concentrations in a set of sediment cores in the GLERL

sediment “library” collected in 1983. These cores had not been previously analyzed. We are grateful to the Ontario Ministry of Environment and Canada Centre for Inland Waters for the opportunity to collect these cores from the CSS Limnos.

### **Accomplishments**

We contributed to a paper on contributions of mercury to Lake Superior from regional mining activities. The paper has been accepted for publication in a Special Volume on Metal Mining and the Environment of the Journal of Geochemistry by Kerfoot, Harting, Rossmann and Robbins, entitled “Amalgam mercury in copper, silver and gold ores: an unexpected contribution to Lake Superior”

Completed three papers with C. Kerfoot, one already published in L. & O (Kerfoot et al., 1999) and two in press for J. Great Lakes Research.

### **SP-37 Information Content of Sediments Accumulated in a Wrecked Ship**

Sediment cores were collected by diver (7/93) from the lavatory of the Grecian, a merchant ship which went down off the coast of Alpena in Lake Huron at the turn of the century. This study was determine if there is any interpretable and useful history related to conditions in the Lake which can be obtained from such material.

### **Accomplishments**

Distributions of  $^{137}\text{Cs}$ , determined for two cores, were not of any research value.

### **SP-39 Radionuclides, Metals and Organic Contaminants in Lake Ladoga Sediments**

In 1993 GLERL received a sediment core from Lake Ladoga, a major lake in Russia near the border with Finland. Samples from the core, collected joint Finnish-Russian expedition, were subsequently analyzed for  $^{134}\text{Cs}$ ,  $^{137}\text{Cs}$ ,  $^{210}\text{Pb}$  - by Dr. P. Van Hoof (GLERL), for polyaromatic hydrocarbons (PAHs).

GLERL participated in this study to determine if a history of contamination of this lake could be reconstructed from the available core. In samples, we found significant amounts of the short-lived radioactive isotope of cesium,  $^{134}\text{Cs}$  ( $t_{1/2} = 2.05$  yr). Much of landscape in Europe and in western part of the former Soviet Union, had been contaminated with both  $^{137}\text{Cs}$  and  $^{134}\text{Cs}$ , plus other radionuclides, from the reactor explosion in Chernobyl, Ukraine on April 26, 1986. The two radionuclides were released to the atmosphere in a distinctive ratio that enabled

researchers to discriminate between environmental contamination due to Chernobyl and other potential sources. Although the shorter-lived radionuclide was also produced and disseminated with  $^{137}\text{Cs}$  by above ground nuclear testing,  $^{134}\text{Cs}$  from that source had decayed into the background by the time of the Chernobyl event. As a result, we could use the sedimentary profile of  $^{134}\text{Cs}$  in combination with its characteristic Chernobyl ratio to  $^{137}\text{Cs}$ , to reconstruct the nuclear weapons part of the  $^{137}\text{Cs}$  profile.

We concluded that there were no significant sources of radiocesium contributing to our coring site in Lake Ladoga. We also used the radiocesium and  $^{210}\text{Pb}$  data to develop a vertical sediment transport (VST) model yielding a sediment chronology and the rate and range of near-surface mixing. Subsequently, the model was used to back-calculate the history of total PAH loading to the coring site and to account for the formation (diagenesis) of a naturally occurring PAH, perylene.

## **Plans**

Complete preparation of a paper for publication.

## **SP-43 Sediments in Florida Bay**

### **Background**

This project, funded by the South Florida Water Management District, has been a multi-agency collaboration (ANL, GEO-UM, SPH\_UM, USGS-SP, USGS-WH) to reconstruct ecological and biogeochemical changes in Florida Bay during this century using recently deposited sediments.

In Phase I of this project, the object was to determine if there were suitable archival sites in the Bay. In Phase II, dated sediment cores from such sites would be used to develop historical records for contaminants such as mercury, found in some bay fish, and nutrients, particularly phosphorus, that is thought to be a primary cause of accelerated eutrophication in some areas of the bay. GLERL's main role has been to use of radionuclides for dating and chronology confirmation (Phase I).

The study was successful in locating archival sites and developing sediment chronologies using the uranium-series radionuclides  $^{210}\text{Pb}$  and  $^{226}\text{Ra}$ . Sediment dates were confirmed by demonstrating that temporal records of lead in sediment cores matched previously measured concentrations of lead in annual layers of coral nearby on the ocean side of the Keys. However neither the sediment or coral records matched known long-term history atmospheric lead concentrations. In addition sediment records of fallout nuclides ( $^{137}\text{Cs}$  and plutonium) did not match the atmospheric fallout records. These discrepancies were removed by recognizing that atmospheric inputs lead and the fallout nuclides were time-averaged before incorporation into sediments. Most likely these constituents accumulated sediment mixed layers within the bay before being resuspended and horizontally redistributed to coring sites.

The analysis revealed that all atmospherically-delivered tracers in the study were subject to a time-averaging process with a common time constant of 16 years. On further examination it was evident that decade-scale time-averaging constants are probably characteristic of coastal marine sediments and reflect the residence time of particle-associated, non-degradable contaminants in many coastal environments.

## **Accomplishments**

We published a paper in J. Geophysical Research entitled "Time averaged fluxes of lead and fallout radionuclides to sediments of Florida Bay" in J. Geophysical Research - Oceans. We also contributed to a paper that has been accepted as a chapter in a special issue (book) entitled "Paleoecological Studies of South Florida", a publication of Bulletins of American Paleontology entitled "Sediment dynamics of Florida Bay mudbanks on a decadal time scale" by Holmes, Robbins, Halley, Bothner, ten Brink and Marot.

GLERL contributed to the preparation of a Final Report to the South Florida Water management District (1998).

## **SP-44 Accumulation and Mixing of Sediments in Lake Michigan**

### **Background**

With the establishment of the Lake Michigan Mass Balance Program, the EPA commenced development of a next-generation mass balance model to account for the fate of contaminants in the lake with improved spatial and temporal resolution. The model is expected to more realistically describe effects of currents, thermal stratification, fall overturn, and particle transport, sediment resuspension on the movement and ultimate removal of contaminants from the lake. In the program, model development and calibration has been supported by new estimates of inputs of selected inorganic and organic contaminants. The GLERL's ERT project, funded by EPA between 1994 and 1999, has had major role in providing lake-wide data on physical properties of coreable sediments and in generating sediment state variable data from profiles of  $^{210}\text{Pb}$  and  $^{137}\text{Cs}$ .

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 were determined by use of a standard, steady-state vertical sediment transport (VST-S) model to interpret  $^{210}\text{Pb}$  profiles. The VST-S model has treated mixing of sediment particles as a diffusive process with constant particle mixing rate occurring only above a sharply defined depth. VST-S considers  $^{210}\text{Pb}$  to be particle bound and the sediment accumulation rate to be invariant. To interpret profiles of  $^{137}\text{Cs}$ , the analogous non-steady vertical transport (VST-N) model was modified to include the first-order time-averaging atmospheric fluxes of  $^{137}\text{Cs}$  experience prior to accumulation of the radionuclide in sediments. Time-averaging means that quantities of  $^{137}\text{Cs}$  introduced at previous times are co-mingled in some proportion with those introduced to the system later. In the lake this occurs to some extent because it takes more than a year for a given month's load to clear out of the water column. As a result, subsequent monthly additions mix in the water with past inputs. But time averaging seems to be due mainly to cycles of deposition, sediment mixing, resuspension and re-deposition that occur over many years and result in large-scale horizontal redistribution of  $^{137}\text{Cs}$ -labeled particles moving toward their final burial sites. The trap measurements spanning more than 20-years (see SP-2) demonstrate that time-averaging is essentially a first-order process thus described by a single time constant that increases by roughly an order of magnitude between inshore and offshore sites. VST-N analysis with system time averaging (STA) provides a new state variable, the STA time constant, as well

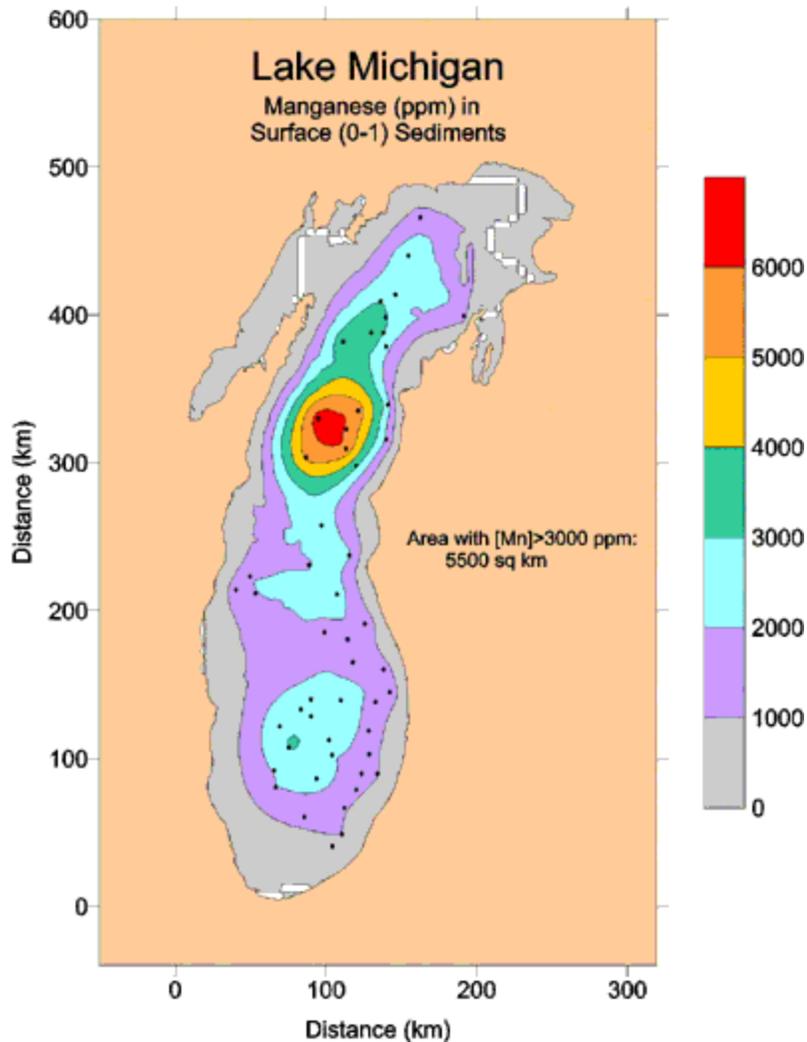
as additional measure of the net mass accumulation rate. Codification of the VST-N (STA) model, incorporation of optimization routines and addition of Monte-Carlo error estimation procedures commenced in 1994 with the initiation of field work.

Between 1994 and 1996, 55 box cores were collected from depositional areas processed at GLERL and analyzed here for fallout  $^{137}\text{Cs}$ . Portions of sediment from each of the cores were analyzed at GLWI for  $^{210}\text{Pb}$ . In addition  $^{137}\text{Cs}$  and  $^{210}\text{Pb}$  data, based on a set of 79 additional box cores collected in 1992 by GLWI, have been available for use in this study. Coring operations were conducted from the R/V Lake Guardian (USEPA) and the R/V Neeskay (GLWI).

### **Accomplishments**

A collaborative study with Argonne National Laboratory was funded on effects of transition metal coatings on the ability of natural and test particles to bind stable and radioactive tracers.

We completed INAA for surface concentrations of about 20 trace elements in 55 cores collected from Lake Michigan between 1994 and 1996 for the LMMB study. In addition, we determined INAA element concentration profiles for the upper 10 cm of five selected cores. Perhaps the most significant discovery was a very large, more than three-fold, elevation in concentrations of manganese (Mn) in surface sediments (0-1 cm) of northern Lake Michigan as compared with the southern part of the lake. It is well known that certain elements like Mn and iron (Fe) can become enriched in near-surface sediments by a natural process (diagenesis) in which redox-sensitive, transition metals can partly dissolve off sediment particle surfaces in a sufficiently reducing sedimentary environment and migrate toward less reducing (i.e. more oxidizing) near-surface sediments where they re-attach to particles. In the southern part of the lake, fine sediments may have concentrations of Mn of about 2000 ppm compared with underlying sediments of, say 1200 ppm, while in the northern deep basin the corresponding values are around 5000 to 1800 ppm. In most areas of Lake Michigan only Mn (but not Fe) exhibits this natural diagenetic enrichment. The area of strong Mn enrichment occurs in the deepest waters (ca 250 m) and is extensive, about 1000 square miles. The significance of our discovery is that (1) there is a natural process by which sediment particles in the deepest part of the lake are "labeled" with Mn; these particles are evidently resuspended at least into near bottom traps (see project SP-2) by an unknown process that seems unlikely to be shear stress determined; Mn labeling can be used to understand particle transport processes in the system and (2) Mn coated particles are efficient scavengers of certain constituents in the water column, namely radium, possibly plutonium, lead and other trace constituents. This year we also presented results (Robbins et al.) at the ASLO meeting (Albuquerque, NM) and collaborated with EPA scientists (Grosse Ile Lab) on the formulation and running of their first-order mass balance model.



Part I (1994-1996 Cores) of the Final Report was completed and delivered to EPA-Large Lakes Laboratory, Gross Ile (July 1999). Submission of this report completes an inter-agency agreement with GLERL. About 90% of the VST-S and VST-N (STA) model calculations (without M-C error estimation) have been completed for the 1992 cores.

For 1994-1996 cores, sample analyses and selected re-analysis in a few cases were completed, data and summary files were completed, models were calibrated and applied, state variable-data with Monte Carlo (M-C) errors were obtained. For the 1992 data from GLWI, considerable effort has been made this year to reconstitute the data set and check for errors. In some cases radionuclide activities had to be re-determined. The data set is now ready to use with transport model programs.

## **SP-45 Soil Accretion Rates in South Florida Wetlands**

### **Background**

This study, supported by the South Florida Water Management District, was initiated in 1994 to evaluate the use of  $^{137}\text{Cs}$  to date soil cores from the Water Conservation Areas (WCAs). These wetland areas, delineated by a network of levees and canals acting as flood control systems, are reservoirs that provide a reliable water supply for people living in predominately metropolitan areas on the eastern side of south Florida. In recent decades the WCAs as well as other parts of the Everglades wetlands, have been impacted by runoff of nutrients (especially phosphorus) from large-scale agricultural activities on adjacent land to the northwest. As a result expanses of natural sawgrass in the vicinity of nutrient inflows, have been slowly replaced by cattails that are better able to take advantage of excess phosphorus. Such vegetational succession has contributed to the degradation of wildlife habitats.

The role of phosphorus in stimulation of plant growth and succession has been established partly by observations of increased levels of this nutrient in soil cores dated by others using  $^{137}\text{Cs}$ . At sites where original sawgrass communities have given way to cattails, soil accretion rates above the 1964 soil horizon as delineated by the radiocesium maximum, have been shown to be considerably greater than rates at un-impacted sites. Indirectly, radiocesium has had an important role in establishing the phosphorus-soil accretion connection and informing Management District decisions on management of the wetlands system. Nevertheless a concern has remained about the validity of using radiocesium as a chronological marker in highly organic soils such like those in the WCAs. The radionuclide is subject to post-depositional migration in some organic soils so, at least in principle, considerable inaccuracies could be encountered in establishing chronological information by this method alone. To evaluate the method we first measured radiocesium profiles in cores collected in 1993 Dr. K. R. Reddy, University of Florida, Gainesville and subsequently conducted joint laboratory studies on the distribution of  $^{137}\text{Cs}$  between soil solids and pore water, between plants tops, roots and soils. In addition GLERL and U. Fla. conducted chemical extraction procedures on fresh soils to characterize the binding of radiocesium to various fractions. Also we investigated the long-term transfer of carrier-free radiocesium from pore water to soil solids. In 1996, we collected cores along a vegetational/nutrient gradient in Water Conservation Area 2A to compare radiocesium chronologies to those based on  $^{210}\text{Pb}$  as determined by the U.S. Geological Survey.

### **Accomplishments**

The results of our novel modeling approach to interpreting soil core distributions of  $^{210}\text{Pb}$  were presented at the SPERA 2000 meeting and discussed at the workshop. We contributed to summary of those workshop discussions in chapter entitled "Environmental Changes and Radioactive Tracers" by Hancock, Edgington, Robbins, Smith, Brunskill and Pfitzner in Proceedings of the South Pacific Environmental Radioactivity Association (SPERA) 2000. The book was published in 2002.

We completed laboratory studies and prepared a final report to the South Florida Water Management District. Submission of the report, in March 1999, completes terms of the

interagency agreement. The main finding of the report was that Cs-137 is not an ideal tool for dating soil cores from the Everglades but, as used to chronicle order-of-magnitude changes in soil accumulation rates between sites, it is an acceptable method. Therefore any management decisions based on conservative use of Cs-137 dating were probably justified. The study also showed that an alternative method of using Pb-210 is probably better but that the Everglades Wetlands is an inherently difficult system for establishing precise soil layer dates. This year we began developing new quantitative models for interpreting lead-210 profiles that circumvented use of the conventional CRS mapping method for dating that does not yield model Pb-210 profiles for comparison with data.

### **SP-47 Mississippi Basin Carbon Project**

The U.S. Carbon Cycle Science Program (CCSP) is an interagency partnership that draws on the expertise and ongoing research in seven agencies, with the objective of developing a whole-system predictive capability for the global carbon system. The ultimate goal is to provide integrated estimates of carbon sources and sinks. A flagship project of the U. S. Geological Survey aims to develop a comprehensive carbon budget for the Mississippi River drainage basin. It will assess effects of different of land characteristics (topography, vegetation, soil types etc.) and use (urban, rural, agricultural etc.) on storage and transport of carbon, on nutrient cycles and processes of erosion and sedimentation as they affect carbon cycling throughout the River and its drainage basin. Our role in the project is to estimate rates of accumulation of soils accumulating in the Mississippi Delta wetlands region since the mid-1960s using the fallout Cs137 peak as a time marker. The information will be used to estimate net carbon storage in various forms during the past 30 years. Our laboratory began receiving core samples in May of 1998 and has continued both analytical and interpretive work through the duration of the pre-synthesis part of the project.

### **Accomplishments**

We provided USGS-Atlanta with final results of radionuclide analysis with interpretation for inclusion in a research report on carbon sequestration in the Mississippi Delta wetlands.

A linear scaling algorithm was developed to align stratigraphic signatures in replicate core samples. This was provided to the USGS.

We received and analyzed two additional soil cores for <sup>137</sup>Cs and <sup>40</sup>K and obtained mass accumulation rates. This year we also revised our core data files to conform to new USGS bulk dry density values.

To date twelve soil cores have been analyzed and data reports have been forwarded to USGS. At a recent meeting in Menlo park we decided to prepare summary tables, figures and interpretive narratives for all cores based stratigraphic indications of a codetermined radionuclide, <sup>40</sup>K. This is currently underway.

## **SP-48 Episodic Resuspension Events in Lake Michigan (EEGLE)**

This multi-institutional project studies effects of short periods of intense, wind-driven water circulation on the transport of eroded and resuspended particles (plumes) and their possible ecological effects in the southern part of Lake Michigan. This program is described in detail elsewhere. One objective of the EEGLE project is to discover the role that episodic resuspension events may have on sediment focusing in the southern part of the lake. Fine-grained materials, mainly eroded from shoreline sources and post-glacial clay sediments on the western side, ultimately accumulate along a strip, roughly 50-100 m deep, on the eastern side of the lake. Inventories of fallout  $^{137}\text{Cs}$  in this high-deposition region, determined from gravity cores collected in 1972, had nearly the same pattern as sediments that had accumulated during the past 3500 years. Since  $^{137}\text{Cs}$ -labeled particles had been introduced in the mid-1960s, it was clear that sediments were concentrated into limited areas of the lake bottom (focused) in a matter of five years or less. In 1982, an area of particularly high sediment accumulation, near St. Joseph, Michigan, was re-cored. Between 1972 and 1982, when little new  $^{137}\text{Cs}$  entered the lake, inventories had increased toward the center of the high-deposition (HIDEP) area at the expense of inventories at the margins. Thus, even after the primary pattern of  $^{137}\text{Cs}$  was well established, radiocesium-labeled particles continued to be focused through cycles of resuspension and redeposition. Subsequently, between 1992 and 1996 all depositional basins of Lake Michigan were box cored as part of the Lake Michigan Mass Balance Project (SP-44). That study showed that the HIDEP area in the southern Lake Michigan was the outstanding sink for accumulation of particle-associated radionuclides in the entire lake.

To assess the contribution of particles to the HIDEP area during episodic events, we employed a short-lived radionuclide,  $^7\text{Be}$  ( $t_{1/2} = 53.4$  days) as a diagnostic tool (vide supra). Delivered to the lake surface from the air,  $^7\text{Be}$  rapidly attaches to particles in water and is conveyed to the bottom as particles settle (scavenging). At normal particle concentrations in water, about half of the  $^7\text{Be}$  is associated with particles; the remainder is present either in a dissolved state or associated with components having insignificant settling rates. When episodic resuspension events occur, it is expected that, because of the increased numbers of particles in the water, a greater fraction of  $^7\text{Be}$  will attach to settling particles. To test this idea, we planned to measure the inventory of  $^7\text{Be}$  (total amount per unit area of sediment) in the HIDEP area before, during, and after the period when late-winter and early-spring storms generally produce the most intense episodic resuspension events.

### **Accomplishments**

We completed construction and review of the radionuclide inventory data set consisting of surface concentrations of  $^7\text{Be}$ ,  $^{137}\text{Cs}$ ,  $^{40}\text{K}$  and inventories of  $^7\text{Be}$ ,  $^{137}\text{Cs}$  at 168 sites within the HiDep area. From the data we prepared contour maps of (1) the inventory of fallout  $^{137}\text{Cs}$  ( $t_{1/2} = 30$  years), (2) the net mass accumulation rate based the  $^{137}\text{Cs}$  inventories and (3) inventories of cosmogenic  $^7\text{Be}$  ( $t_{1/2} = 53$  days) before, during and after winter-spring resuspension episodes in 1999. These maps reveal patterns of mass and radionuclide accumulation with unprecedented spatial resolution.

The pattern of long-lived radionuclide ( $^{137}\text{Cs}$ ) inventories mimics the mean trajectory of satellite-imaged reflectance (turbidity) traversing the High-Dep area during episodic resuspension events. Maps of the short-lived radionuclide ( $^7\text{Be}$ ) reflect the seasonal cycle in its delivery from the atmosphere as well as a plume-specific enhancement in inventory following the winter storm events. The radionuclide data suggest that long term patterns of accumulation of sediments within the HiDep area are not simply the result of plume events. Rather, particles delivered to the area continue to be redistributed horizontally and focused by post-plume processes.

We presented results (Eadie et al.) at the ASLO meeting (Albuquerque) and prepared a poster for future presentations and display at GLERL.

We conducted box-coring expeditions in southern Lake Michigan area in September and November 1998 and in February, March and June 1999. On each occasion, box cores were collected from a rectangular array of 35 sites enclosing the HIDEP area. In some cases sea conditions (particularly in November 1998) did not permit complete collections. As of September 1999, all  $^7\text{Be}$  measurements have been completed. Analysis of  $^{137}\text{Cs}$  is still in progress. Contour maps for  $^7\text{Be}$  inventories indicate an increase after the episodic resuspension event period compared with other times. However the final interpretation of inventory maps depends on having a realistic box model for the relation between delivery of  $^7\text{Be}$  to the lake surface and the amount transferred to the bottom. It was expected concentrations of  $^7\text{Be}$  on particles and gross downward fluxes could be determined from sediment traps deployed during the Jan-June, 1999 period within HIDEP area. However the trap data will not be available because of equipment failure. The June cores were also analyzed by K. Orlandini (ANL) for Th-234, a short-lived decay product of dissolved U238.

### **SP-49 Vertical Sediment Transport Models for Lake Tahoe**

Lake Tahoe, on the border between Californian and Nevada, was impacted during the 19th century by deforestation that caused runoff of large quantities of soil eroded from the watershed. In more recent times, development of housing and commercial properties, particularly along the shore of the lake, has increased nutrient as well as sediment loads to the lake with resulting deterioration of water quality. The aim of the present un-funded study is to determine what changes in biogeochemical conditions of the lake during the past century or more might be inferred from sedimentary records. In collaboration with D. Edgington, GLWI, and individuals from the Tahoe Limnological Laboratory, we examined  $^{210}\text{Pb}$  records from a core collected in 1981 and subsequently developed several mathematical models to interpret radionuclide data in the 1981 core plus two cores collected in 1991.

### **Accomplishments**

We presented results (Edgington et al.) at the ASLO meeting (Albuquerque, N. M.).

The core collected in 1998 was analyzed at GLWI for  $^{210}\text{Pb}$  and plutonium (Pu) using acid extracts of sediments. At GLERL, whole dry core samples were counted in standardized geometry, each on two different well detectors to obtain maximum accuracy and high precision

in determining activities of  $^{210}\text{Pb}$ ,  $^{266}\text{Ra}$ ,  $^{235}\text{U}$ ,  $^{228}\text{Th}$ ,  $^{137}\text{Cs}$  and  $^{40}\text{K}$ . As a result, we showed that the  $^{137}\text{Cs}$  profiles were degraded (more diffuse without a subsurface peak) compared with the distribution of plutonium. This latter radionuclide did have a clear peak well below the sediment surface. We think this means that, relative to Pu,  $^{137}\text{Cs}$  has a very long residence time in the water of Tahoe. A few measurements of this radiocesium in the water should confirm or negate this hypothesis. We also discovered significant excess  $^{228}\text{Th}$  in sediments as deep as 4 cm. This particle-reactive radionuclide has a half-life of just 1.9 years and is produced in overlying water by decay of dissolved  $^{228}\text{Ra}$  [ ]. The addition of Pu and  $^{228}\text{Th}$  to the suite of radionuclides should resolve uncertainties about the extent of mixing which near-surface sediments experience. Because sediment accumulation rates are so low in Lake Tahoe, it is particularly important to correctly assess the extent to which mixing may have altered or even obliterated historical records.

The  $^{210}\text{Pb}$  data alone did not rule out the possibility of mixing in near-surface sediments. Ordinarily  $^{137}\text{Cs}$  is helpful in choosing between alternative vertical sediment transport models. But in this lake, activities of  $^{137}\text{Cs}$  were very low and exhibited no peak corresponding to the 1963-1964 fallout maximum. As a result, we returned to Tahoe this past summer to obtain additional material for radiometric and other analyses.

## **SP-50 Metal Contaminant Chronology of Recent Sediments near the Great Barrier Reef, Northern Australia**

### **Accomplishments**

We developed and applied several alternative models to account for distributions of  $^{210}\text{Pb}$  and  $^{137}\text{Cs}$  in a sediment core from coastal deposits near Townsville, Queensland, Australia. Previous work had shown a major spike in Hg concentrations down core during the time that gold was being extracted by the Hg amalgam-evaporation technique from ores present within the Burdekin River watershed. The Burdekin flows into the coastal area where the core was collected. This year we obtained INAA data for gold (Au) indicating that Hg increases down core were accompanied by commensurate increases in Au. While Hg may reach the coring by atmospheric pathways, it is likely that Au would be conveyed solely by tributary runoff.

## **SP-51 Sedimentary Evidence for Recent Die-Off of *Diporeia* in Lake Michigan**

Cores for biogenic Silica were analyzed. Worked with Andriesen and Stoermer to obtain a properly formatted and extensive array of diatom taxonomic data. Developed a computer program to interrogate the array for conventional and problem-specific statistical analysis.

## Products

### Posters

Robbins, J.A., C. Holmes, R. Halley, M. Bothner, E. Shinn, J. Graney, G. Keeler, M. tenBrink, K.A. Orlandini, and D. Rudnick; Dec. 2000. *Time-Averaged Fluxes of Lead and Fallout Radionuclides to Sediments in Florida Bay*

Robbins, J.A., D.N. Edgington, B.J. Eadie, N.R. Morehead, and R. W. Rood; Feb. 2001. *Time Scales for Sediment Focusing in Lake Michigan Based on Fallout 137Cs Data: A Thirty Year Study*

Robbins, J.A., B.J. Eadie, N.R. Morehead, and D. N. Edginton; Feb. 2002. *High Resolution Mapping of 137Cs and 7Be Inventories in the High Deposition Area of Southern Lake Michigan*

### Publications

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